Dynamique spatio-temporelle des flux naturels et anthropiques vers les hydrosystèmes littoraux tributaires des eaux souterraines : Investigations isotopiques et géochimiques pour la compréhension des interactions aquifères-lagune sur le site de Biguglia (Haute-Corse)

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Investigations isotopiques et géochimiques pour la compréhension des interactions aquifères-lagune sur le site de Biguglia (Haute-Corse)

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Jury

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Spatio-temporal dynamics of natural and anthropogenic flows towards coastal groundwater-dependent hydrosystems

Isotopic and geochemical investigations for the understanding of aquifer-lagoon interactions within the Biguglia system (Haute-Corse)

Abstract

The exacerbated anthropization of coastal hydrosystems poses a significant threat to groundwater and ecosystems that depend on it, then called "groundwater-dependent ecosystems". Like the Biguglia hydrosystem (Corsica, France), rapid and disorganized urbanization, as well as industrial and agricultural activities developed on the Marana plain are major sources of groundwater degradation. This strategic resource, used for the drinking water supply of the Bastia urban area, presents the markers of multiple and diffuse contaminations. In addition to being problematic for human water needs, the groundwater qualitative degradation of the Marana aquifer also constitutes a threat to the fragile ecosystem of the Biguglia lagoon and the sustainability of the ecosystem services it provides. In this context of increasing pressures, a strong knowledge of the Biguglia hydrosystem behavior is essential to ensure sustainable management of water resources. To this end, a multi-tracer geochemical and isotopic approach has been developed in order to better constrain the state of the groundwater resource, the hydrodynamic conditions and the nature of aquifer-river-lagoon interactions. The study of the stable isotopes of the water molecule (^{18}O, ^{2}H) has highlighted the recharge complexity. The Marana aquifer benefits from an indigenous recharge through direct infiltration of precipitation on the plain and an allochthonous recharge through precipitation from the schistous reliefs. The infiltration of river water from the Bevincu and Golu rivers and the lateral contribution of water from the schistous reliefs also contribute to the aquifer recharge. The developed mixing model (^{18}O, Cl^- and HCO_3^-) allowed a semi-quantitative estimation of the mixing processes. It demonstrates the complex aquifer behavior, with a significant difference in the contribution of schistous groundwater depending on the location and depth of the resource. It is also clear that the Biguglia lagoon is partially dependent on groundwater. The Marana plain is subject to qualitative degradation due to the excessive presence of nitrates (NO_3^-) and organic micropollutants. According to the NO_3^- (^{15}N-NO_3^-, ^{18}O-NO_3^-) and Boron (^{11}B) isotopes, the main sources of nitrogen are soil and wastewater. The correlation between NO_3^- concentrations and water residence time (^{3}H and CFCs) highlighted the storage and the groundwater archiving capacity. With regard to the evolution of land use, the progressive modification of the nitrogen sources recorded in the aquifer made it possible to trace the socio-environmental trajectory of the Biguglia hydrosystem. The contemporary degraded state of the resource mainly results from the pollution legacy linked to historical human activities. The conceptual model developed provides new elements that can help towards the implementation of relevant management strategies, to ensure the sustainability of water resources and associated ecosystem services.

Key words: Coastal aquifers; Multi-tracer approach; Groundwater archiving capacity; Nitrate legacy; Integrated water resources management

Résumé

L'anthropisation grandissante des bassins versants côtiers représente une menace importante pour les eaux souterraines et les écosystèmes qui en dépendent, alors appelés « écosystèmes tributaires des eaux souterraines ». A l'image de l'hydrosystème de Biguglia (Corse), l'urbanisation rapide et désorganisée ainsi que les activités industrielles et agricoles développées sur la plaine de la Marana sont autant de sources de dégradation des eaux souterraines. Cette ressource, pourtant stratégique et utilisée pour l'alimentation en eau potable de la région bastiaise, présente aujourd'hui les marqueurs d'une contamination multiple et diffuse sur l'ensemble de la plaine. En plus d'être problématique pour les besoins en eau humains, la dégradation qualitative des eaux souterraines de l'aquifère de la Marana, constitue également une menace pour l'écosystème fragile de la lagune de Biguglia et la pérennité des services écosystémiques qu'il prodigue. Dans ce contexte de pressions croissantes, la connaissance approfondie du fonctionnement de l'hydrosystème de Biguglia s'impose comme un élément essentiel pour garantir une gestion durable de la ressource en eau.
Dans ce but, une approche multi-traceurs géochimiques et isotopiques a été développée afin notamment de mieux contraindre l'état de la ressource, les conditions hydrodynamiques et la nature des interactions aquifères-rivières-lagune. L'étude des isotopes stables de la molécule d'eau ($\delta^{18}O$, $\delta^{2}H$) a mis en évidence la complexité de la recharge. L'aquifère de la Marana bénéficie d'une recharge autochtone par l'infiltration directe des précipitations sur la plaine et d'une recharge allochtone par les précipitations en provenance des contreforts schisteux. A ces mécanismes, s'ajoute également la recharge prodiguée par l'infiltration des eaux du Bevincu et du Golu et la contribution latérale des eaux en provenance des contreforts schisteux de la Corse alpine. Le modèle de mélange développé ($\delta^{18}O$, Cl^- et HCO_3^-) a permis une estimation semi-quantitative des mélanges. Il démontre la complexité du fonctionnement de l'aquifère, avec une différence notable de la contribution des contreforts schisteux en fonction de la localisation et de la profondeur de la ressource. Il apparaît également de manière claire que la lagune de Biguglia est partiellement tributaire des eaux souterraines. La plaine de la Marana est sujette à une dégradation qualitative liée à la présence excessive de nitrates (NO_3^-) et de micropolluants organiques. D'après les isotopes du NO_3^- (^{15}N-NO_3^-, ^{18}O-NO_3^-) et du Bore (^{11}B), les principales sources d'azote sont le sol et les eaux usées. La corrélation entre concentrations en NO_3^- et temps de résidence des eaux (^{3}H et CFCs) a mis en avant la capacité de stockage et d'archivage des eaux souterraines de l'aquifère de la Marana. Mise au regard de l'évolution de l'occupation des sols sur la plaine, la modification progressive des sources azotées enregistrée dans l'aquifère a permis de retracer la trajectoire socio-environnementale de l'hydrosystème de Biguglia. L'état contemporain dégradé de la ressource découle en grande partie de l'héritage des pollutions liées aux activités humaines historiques. Le modèle conceptuel élaboré grâce à ces travaux apporte de nouveaux éléments de compréhension qui pourront aider à l'instauration de stratégies de gestion pertinentes, assurant la pérennité future des ressources en eau et des services écosystémiques qui en dépendent.

Mots clés : Aquifères côtiers ; Approche multi-traceurs ; Capacité d'archivage des eaux souterraines ; Legs nitraté ; Gestion intégrée des ressources en eaux

Sunta

I litturali sò accampati da u sviluppu è l'attività umane chì inghjennanu un guastu quantitativu è qualitativu di e risolse d'acqua suttaranie. Di pettu à a crescita demugrafica è urbana, è à l'effetti di u cambiamentu climatticu, issi guasti puderiani accresce si in a zona mediterranea. Fattu assai preghjudizievule per u sviluppu sucio-ecunomicu di i liturali : u sgradamentu di e risolse in acqua hè anc'ellu assai prublematicu per l'ecosistemi detti « dipendenti di l'acqua suttarania ». Iss'ecosistemi, guasi o tutalamente dipendenti, annu bisognu di l'acque suttaranie per assume u so funziunamentu è l'inseme di e so pussibilità (pesca, turisimu, prutezzione di e coste…).

L'idrosistema di a laguna di Biguglia illustreghja bè isse prublematiche. L'urbanisazione di a piaghja di Bastia s'associa à una crescita demografica maiò, pratichendu una pressione tremenda nantu à e risolse in acqua suttaranie è nantu à a laguna chì ne dipende. In issu cuntestu di pressione chì s'accresce, una gestione adattata, di e risolse in acqua, hè essenziale per guarantisce a suddisfazzione futura di i bisogni in acqua, necessarii à l'attività umane è l'ambiente. Intantu, una bone gestione di a risolsa necessiteghja una cunniscenza acuta di u funziunamentu di l'inseme di l'idrosistemi. Ci tocca à identificà l'origine di i flussi suttaranii (chì cuntenenu acqua) è di capisce e dinamiche di corsa è l'interazzione chì asitenu trà l'acque suttaranie, l'acque di fiumi, salmatre (laguna) è salite (mare). Per ciò, una avvicinanza multi-tracciatori, utilizendu i dati geochimichi (chimica di l'acque) è isotopichi, hè stata sviluppata.

I risultati uttenuti anu permessu di custringhje megliu i prucessi di ricarica, i fenomeni di mischiu trà e sferente masse d'acqua è a qualità è u tempu di residenza di l'acque suttaranie. Un primu mudellu cuncettuale di funziunamentu di l'idrositema di a laguna di Biguglia hà pussutu esse cunstituitu. A messa in lea di i resultati idrologichi incù u studiu di l'occupazione di e terre hà permessu di ritraccià e traghjettorie sucio-ambientale di l'idrosistema. Spunta chì u statu cuntempuraniu guastu di a risolta vene, in mai parte, da a redita di e pulluzione leate à l'attività storiche umane. Cusì, i novi elementi di capiscitura di u funziunamentu di l'idrosistema di a laguna di Biguglia puderanu aiutà l'adattazione di e pulitiche di gestione di risolse affinchè guarantisce a salvezza di u sistema idrologicu sanu.

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« *C'est dans l'effort que l'on trouve la satisfaction et non dans la réussite. Un plein effort est une pleine victoire.* »

Lettres à l'Âshram – Gandhi

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- 2019-2020 : **Micro-LAG** : Impact touristique saisonnier sur la teneur en composés émergents et nanoparticules des hydrosystèmes côtiers en Méditerranée : investigations sur les micro-estuaires et lagunes de Haute-Corse ;
- 2018-2019 : **HydrArchive** : Existe-t-il un archivage hydrogéologique des ruptures hydrosystèmiques qualitatives brutales au sein du bassin versant de Biguglia ? ;
- 2017-2018 : **µPol-LAG** : Utilisation des micro-polluants émergents comme marqueurs de l'anthropisation côtière récente des flux superficiels et souterraines vers les lagunes ;
- 2016-2017 : **Nitro-LAG** : Dynamique socio-hydrogéologique du bassin versant de la lagune de Biguglia, dualité entre résilience et vulnérabilité ;
- 2015-2016 : **Hydrogeo-LAG** : Vulnérabilité hydrogéologique du bassin versant de la lagune de Biguglia en lien avec l'anthropisation côtière.

Le détail des activités de valorisation et des travaux menés au cours de la thèse, entre publications, communications et collaborations, est présenté ci-après.

Publications internationales

- Articles publiés dans des revues scientifiques internationales

- **M. EROSTATE**, F. HUNEAU, E. GAREL, S. GHIOTTI, Y. VYSTAVNA, M. GARRIDO, PASQUALINI, 2020. Groundwater dependent ecosystems in coastal Mediterranean regions: Characterization, challenges and management for their protection. *Water Research 172, 115461.* *https://doi.org/10.1016/j.watres.2019.115461*

- **M. EROSTATE**, F. HUNEAU, E. GAREL, Y. VYSTAVNA, S. SANTONI, V. PASQUALINI, 2019. Coupling isotope hydrology, geochemical tracers and emerging compounds to evaluate mixing processes and groundwater dependence of a highly anthropized coastal hydrosystem. *Journal of Hydrology* 578, 123979. https://doi.org/10.1016/j.jhydrol.2019.123979.

- J. JAUNAT, E. GAREL, F. HUNEAU, **M. EROSTATE**, S. SANTONI, S. ROBERT, D. FOX, V. PASQUALINI, 2019. Combinations of geoenvironmental data underline coastal aquifer anthropogenic nitrate legacy through groundwater vulnerability mapping methods, *Science of The Total Environment* 658, 1390-1403. https://doi.org/10.1016/j.scitotenv.2018.12.249.

- M. EROSTATE, F. HUNEAU, E. GAREL, M. F LEHMANN, T. KUHN, L. AQUILINA, V. VERGNAUD-AYRAUD, T. LABASQUE, S. SANTONI, S. ROBERT, D. PROVITOLO, V. PASQUALINI, 2018. Delayed nitrate dispersion within a coastal aquifer provides constraints on land-use evolution and nitrate contamination in the past. *Science of The Total Environment* 644, 928–940. https://doi.org/10.1016/j.scitotenv.2018.06.375.

- Chapitre de livre

- M. EROSTATE, E. GAREL, F. HUNEAU, V. PASQUALINI, 2017. Delineating the aquifer role in the anthropogenic fingerprint on the groundwater dependent ecosystem of the Biguglia Lagoon (Corsica, France). In: Groundwater and Global Change in the Western Mediterranean Area, M.L. Calvache et al. (eds.), Environmental Earth Sciences. https://doi.org/10.1007/978-3-319-69356-9_16

Communications internationales

- Congrès internationaux avec comité de relecture

- **M. EROSTATE**, E. GAREL, F. HUNEAU, V. PASQUALINI, 2019. Coupling geochemistry, environmental isotopes and organic pollutants to improve the understanding of anthropized coastal groundwater dependent hydrosystems. European Geosciences Union (EGU), Vienne (Autriche), 7-12 avril 2019. (Présentation orale)

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- Pr. P. BAUDRON, Polytechnique Montréal (Canada) : **Projet Ar^{14}Chive -** Lauréate d'une **bourse de recherche Mitacs Globalink**, co-financée Mitacs/Campus France (05-08/2018)

- Européens

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Introduction

1. Contexte général

A l'interface entre milieu continental et milieu océanique, les zones côtières abritent des écosystèmes uniques, d'une biodiversité exceptionnelle. Pourtant, ces milieux fragiles sont soumis à de fortes pressions naturelles et anthropiques qui menacent leur pérennité (Newton et al., 2014).

Les littoraux se caractérisent par un développement croissant des activités anthropiques couplé à une expansion démographique importante. Actuellement, 3,6 milliards d'habitants, soit environ 60% de la population mondiale, vivent à moins de 60 km du littoral (David, 2015). Une proportion qui devrait atteindre 75% d'ici 2050 (Neumann et al., 2015). La concentration des activités humaines entre urbanisation, agriculture, industrie et activités touristiques induit une pression sur l'ensemble des ressources naturelles et tout particulièrement les ressources en eau (Lotze et al., 2006). Pourtant, la capacité des zones côtières à satisfaire la demande en eau est une condition *sine qua non* au développement socio-économique des zones littorales (Zepeda Quintana et al., 2018).

Les ressources en eau de surface étant les premières concernées par les problèmes de disponibilité et/ou de vulnérabilité aux pollutions (UNEP/MAP, 2012; McGrane, 2016), les aquifères littoraux constituent alors des ressources d'eau souterraine essentielles tant pour les activités humaines (alimentation en eau potable, usages agricoles, industriels et touristiques) que pour l'environnement (zones humides littorales, marais et lagunes côtières situés à l'exutoire de ces nappes) (Michael et al., 2017). Souvent soumis à une sollicitation excessive, les aquifères côtiers sont fréquemment sujets à des dégradations quantitatives et/ou qualitatives irrémédiables. L'extraction d'importants volumes d'eau souterraine, en plus de réduire la ressource disponible, perturbe également l'équilibre entre l'eau douce souterraine et l'eau salée marine (Argamasilla et al., 2017; Jin et al., 2019). Cette déstabilisation facilite ainsi les processus d'intrusion saline et de salinisation durable des aquifères (Mahlknecht et al., 2017; Martínez-Moreno et al., 2017; Parizi et al., 2019). Les rejets de diverses natures, en lien avec les activités humaines, sont autant de sources potentielles de contamination susceptibles d'affecter les ressources en eau souterraine (Burri et al., 2019; Koelmans et al., 2019; Kurwadkar, 2019). Ces dégradations peuvent alors avoir des répercussions considérables sur la satisfaction des besoins en eau humains et environnementaux ainsi que sur l'état écologique des zones côtières.

Si pendant longtemps l'étude des eaux souterraines côtières était principalement dédiée aux processus et conséquences liés à l'intrusion saline, leur implication dans les bilans de masse et les bilans hydrologiques des zones côtières est de plus en plus reconnue (Burnett et al., 2003; Kaleris, 2006; Moore, 2006; Jolly et al., 2008; Gattacceca et al., 2011; Malta et al., 2017). Le caractère partiellement ou totalement dépendant des écosystèmes côtiers aux apports en eau souterraine est alors exprimé par la notion d'écosystèmes « tributaires des eaux souterraines » (Krogulec, 2016). Cette dépendance les place naturellement en situation de vulnérabilité. Les écosystèmes côtiers constituent en effet les collecteurs ultimes des bassins versants hydrologiques et hydrogéologiques. Ils sont donc impactés de manière directe ou indirecte par les prélèvements et contaminations des ressources en eau dans la partie amont des bassins versants. Les dégradations quantitatives et qualitatives des ressources en eau peuvent alors mettre en péril ces milieux fragiles ainsi que l'ensemble des services écosystémiques qu'ils prodiguent (PNUE, 2015; Newton et al., 2018). Parmi la diversité des écosystèmes côtiers, les lagunes côtières se distinguent tout particulièrement par leur importance écologique et économique. Considérées parmi les écosystèmes les plus productifs, elles fournissent une large diversité de services écosystémiques d'une grande importance (ressources nutritives, habitat, protection contre l'érosion ou les inondations, tourisme…).

Chaque espace côtier se distingue par des spécificités naturelles et anthropiques faisant de lui un cas unique. Etant donné la complexité et la multiplicité des interconnexions entre les eaux souterraines et les eaux de surface douces, saumâtres et salées, la gestion des hydrosystèmes côtiers requiert une appréhension globale du système. Il s'agit de préciser le fonctionnement de l'hydrosystème dans son ensemble et de comprendre les relations étroites entre les ressources en eau et les écosystèmes côtiers afin de développer des stratégies de gestion adaptées, durables et intégrées de la ressource en eau.

2. Contexte et enjeux de l'étude

La Corse s'illustre dans le bassin méditerranéen par la présence de nombreuses zones côtières encore préservées (Figure 1), en alternance avec des zones intensément urbanisées et/ou touristiques. Les lagunes de Corse constituent des zones d'intérêt majeur, tant écologique qu'économique, qui contribuent fortement à l'attractivité du littoral.

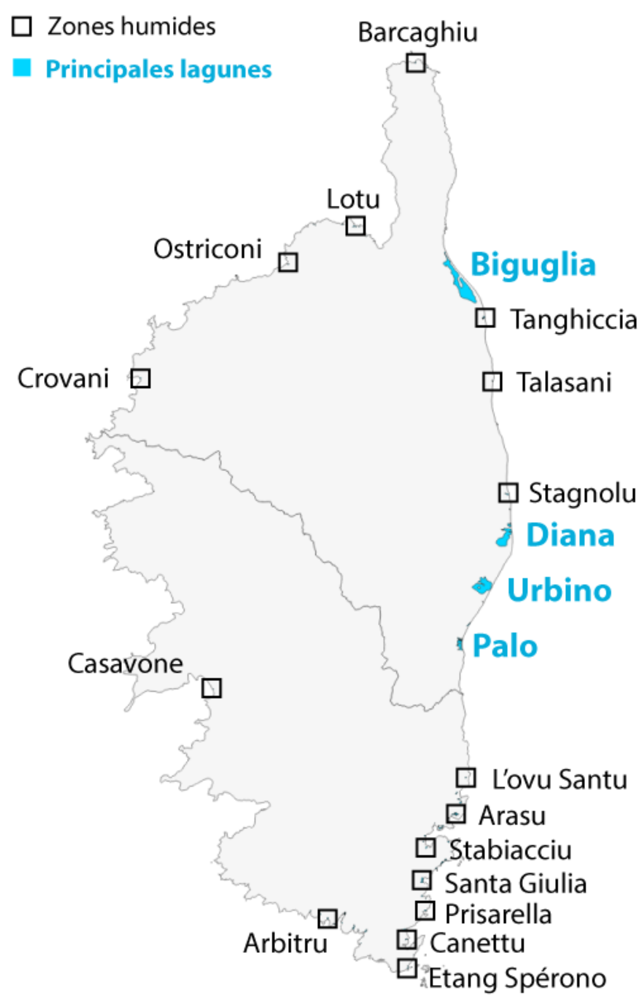

Figure 1 : Carte non exhaustive localisant les principales zones humides côtières et lagunes de Corse, réalisée à partir des données fournies par l'Office de l'Environnement de la Corse (en cours de validation).

Une importante pression est cependant exercée sur les littoraux pour l'extension des zones urbaines. Dans un même temps, la Corse détient au niveau national : le plus fort pourcentage de côte appartenant au Conservatoire du littoral, les taux de croissance démographique (+1,3% entre 2006 et 2019 contre seulement +0,5% sur le continent) et d'artificialisation les plus élevés (Tafani, 2010; Robert et al., 2015) ainsi que la plus forte croissance annuelle du parc de logements (+ 2,2%) (Insee, 2018). Réparties sur plus de 1000 kilomètres de linéaire côtier, les communes littorales occupent plus de 40% de la surface totale de l'île (Zaninetti, 2006) et 30% de l'urbanisation se concentre à moins d'un kilomètre du rivage (SDAGE, 2015). Les communes littorales regroupent 80% de la population corse, principalement répartie dans les deux grandes agglomérations de Bastia et d'Ajaccio (SDAGE, 2015). Dans ce contexte, l'agglomération bastiaise se distingue par des possibilités d'extension urbaine très fortement contraintes. Limitée au Nord par les reliefs du Cap Corse, la zone urbaine de Bastia n'a d'autre possibilité que de s'étendre vers le Sud, sur la plaine alluviale de la Marana. Cette plaine alluviale constitue à la fois le plus grand ensemble aquifère de la région, la source de l'apport en eau potable de l'agglomération bastiaise (Nguyen-Thé et al., 2003) et une part importante du bassin versant de la lagune de Biguglia.

La lagune de Biguglia a été l'objet de dégradations diverses depuis le début du 20ème siècle, en raison notamment de sa proximité avec la zone densément peuplée de l'agglomération bastiaise. L'urbanisation rapide et désorganisée, les zones industrielles et les activités agricoles développées sur la plaine environnante sont autant de facteurs mettant en péril la pérennité de cet écosystème. Pour tenter d'enrayer la dégradation de cette zone humide classée RAMSAR depuis 1991, la mobilisation des acteurs locaux a permis le classement en Réserve Naturelle en 1994. Une réflexion globale sur la gestion de l'eau est alors apparue comme indispensable afin de développer des solutions assurant dans un même temps le développement économique, l'aménagement du territoire et la gestion durable des ressources en eau. De cette concertation est né le schéma d'aménagement et de gestion des eaux (SAGE) de l'Étang de Biguglia (SAGE, 2012). Le SAGE s'adresse tout particulièrement aux aménageurs du territoire. Il vise la restauration ou la non dégradation des masses d'eau présentes sur le bassin Biguglia, dans l'optique d'atteindre le bon état écologique des masses d'eau prôné par la Directive Cadre Européenne (DCE). Deux points majeurs sont évoqués : la lutte contre toutes les formes de pollutions et la minimisation - voir l'absence totale – de l'impact des aménagements collectifs sur les milieux naturels (SAGE, 2012).

En dépit des efforts consentis pour la protection de la lagune de Biguglia, la considération portée aux eaux souterraines demeure restreinte et insuffisante. Les connaissances actuelles sur le fonctionnement hydrologique et hydrogéologique de cet hydrosystème sont encore très parcellaires. Une inconnue quasi totale subsiste sur l'origine des fortes teneurs en azote enregistrées dans les eaux lagunaires. Plusieurs études récentes ont déjà démontré le rôle majeur des eaux souterraines comme vecteur de contaminants d'origine anthropique vers les écosystèmes côtiers. Au vu de la connexion hydraulique fortement suspectée entre la lagune et les aquifères, la compréhension du fonctionnement global de l'hydrosystème et la détermination des sources de contaminations apparaissent comme essentielles. Le maintien de conditions hydrologiques optimales, qualitatives et quantitatives, autour de ces hydrosystèmes constitue un véritable enjeu pour la préservation du potentiel économico-patrimonial des lagunes de Corse et le développement des régions côtières. Devant l'absence d'informations scientifiques, une amélioration substantielle des connaissances s'impose afin de permettre la mise en place de politiques de gestion environnementale adaptées et durables sur ces ensembles littoraux.

3. Objectifs de l'étude et organisation du manuscrit

Les travaux de recherche menés dans le cadre de cette thèse s'intéressent à l'amélioration de la compréhension du fonctionnement global des hydrosystèmes côtiers tributaires des eaux souterraines. Il s'agit pour cela de fournir une compréhension détaillée de la composante hydrologique et hydrogéologique de l'hydrosystème de la lagune de Biguglia. Les résultats attendus de cette recherche sont donc de :

- **Préciser le fonctionnement hydrogéologique des aquifères en continuité hydraulique avec la lagune de Biguglia ;**
- **Caractériser et quantifier les interactions entre les eaux souterraines, les eaux de surface douces et les eaux lagunaires ;**

- **Retracer l'origine et l'historique de l'évolution des pollutions azotées ;**
- **Evaluer le caractère pérenne du fonctionnement de l'hydrosystème et sa vulnérabilité à moyen terme.**

Une approche multi-traceurs géochimiques et isotopiques a été mise en place, se basant sur des données hydrochimiques (paramètres physico-chimiques, éléments majeurs et traces) et isotopiques (^{18}O, ^{2}H) afin de caractériser les conditions de recharge, la qualité chimique des eaux et les phénomènes de mélange. Ces données ont été complétées par i) le tritium (^{3}H) et les gaz anthropiques (chlorofluorocarbures - CFCs et hexafluorure de soufre - SF_6) pour l'évaluation des temps de résidence des eaux souterraines, ii) les isotopes stables du NO_3^- (^{15}N-NO_3^-, ^{18}O-NO_3^-) et du bore (^{11}B) afin de préciser l'origine des polluants azotés dans les eaux souterraines, les eaux de surface douces et lagunaires et de retracer leur l'évolution temporelle dans l'aquifère et iii) les micropolluants organiques pour l'évaluation qualitative de la ressource mais également pour leur fort potentiel comme traceur hydrologique des flux d'infiltration rapide.

Ce manuscrit repose sur l'ensemble des articles réalisés au cours de ce doctorat, tous déjà publiés. Il s'organise en six chapitres. Les chapitres 1 à 3 contextualisent cette étude :

Le **chapitre 1** définit et explique les spécificités relatives aux hydrosystèmes côtiers et aux écosystèmes tributaires des eaux souterraines. Il établit notamment un état de l'art des connaissances et des considérations juridiques et managériales relatives aux lagunes côtières tributaires des eaux souterraines en climat méditerranéen.

Le **chapitre 2** présente le site d'étude de l'hydrosystème de la lagune de Biguglia en abordant notamment le contexte géologique, hydrologique et hydrogéologique ainsi que les pressions anthropiques affectant le bassin versant.

Le **chapitre 3** expose rapidement la stratégie d'étude mise au point et l'ensemble des méthodologies utilisées pour mener à bien la compréhension de l'hydrosystème de la lagune de Biguglia. Pour plus de précision, la présentation détaillée des outils et méthodes d'analyse est disponible dans la section « Méthodologie » des différents articles (Chapitres 4 à 6).

Les chapitres 4 à 6 présentent les principaux résultats acquis au cours de ce doctorat. Ces chapitres se composent d'un résumé étendu en français suivi des articles réalisés, présentés dans leur version finale de publication.

Le **chapitre 4** s'attache à la compréhension du fonctionnement global de l'hydrosystème grâce à une approche multi-traceurs géochimiques et isotopiques couplée au suivi des micropolluants organiques. Ces travaux ont permis d'élaborer le premier modèle conceptuel de fonctionnement de l'hydrosystème de la lagune de Biguglia.

Le **chapitre 5** concerne l'identification des sources de pollutions d'origine azotée et la compréhension de leurs dynamiques spatio-temporelles grâce à l'usage des isotopes du nitrate, du bore et du tritium. Il s'agit particulièrement de retracer les activités contemporaines et historiques induisant ou ayant induit une dégradation des ressources en eau et ainsi d'évaluer la capacité d'archivage des eaux souterraines.

Le **chapitre 6** s'intéresse à la vulnérabilité actuelle et passée des eaux souterraines en mettant en relation des cartographies de vulnérabilité - intrinsèque et spécifique - avec l'étude des contaminations azotées héritées d'activités anthropiques passées.

A travers les recherches appliquées à l'hydrosystème de la lagune de Biguglia, cette thèse propose des outils méthodologiques pour une meilleure appréhension des hydrosystèmes côtiers tributaires des eaux souterraines et fortement anthropisés. Les connaissances acquises sur le fonctionnement de l'hydrosystème de Biguglia permettront également d'apporter de nouveaux éléments d'aide à la prise de décision et à la gestion des ressources en eau à l'échelle locale.

Chapitre 1 :

Connaissances, considérations et gestion des hydrosystèmes côtiers tributaires des eaux souterraines

Ce chapitre définit et explique les spécificités relatives aux hydrosystèmes côtiers et aux écosystèmes tributaires des eaux souterraines. Il établit notamment un état de l'art des connaissances et des considérations juridiques et managériales relatives aux lagunes côtières tributaires des eaux souterraines en climat méditerranéen.

1. Connaissances, considérations et gestion des hydrosystèmes côtiers tributaires des eaux souterraines

1.1. Fondements de l'approche systémique en sciences de l'eau

1.1.1. De l'écosystème à l'hydrosystème

La compréhension des relations complexes entre l'homme et son milieu a clairement motivé l'évolution des approches utilisées en sciences environnementales. Les approches empiriques et descriptives, avec les concepts de « milieu » puis d' « environnement » ont posé au début du 20ème siècle les bases de l'appréhension globale des relations homme/milieu. Progressivement, ces concepts descriptifs et interprétatifs ont laissé place aux approches systémiques, dans le but de fournir des schémas explicatifs de fonctionnement plus intégrateurs. On assiste alors au développement de nouveaux concepts basés sur l'appréhension plus globale des systèmes (Figure 2).

Les écosystèmes sont définis comme des ensembles dynamiques formés des organismes vivants et de l'environnement non vivant dans lequel ils évoluent ; leur interaction constituant l'unité fonctionnelle de base de l'écologie (Lindeman, 1942). Les milieux naturels, et plus largement la biodiversité, se composent donc d'un ensemble complexe d'écosystèmes en interaction (Tansley, 1935), échangeant entre eux énergie, matière et organismes (Burel and Baudry, 1999). La compréhension d'un ensemble d'écosystèmes (appelé écocomplexe) a conduit, dans le cas des sciences hydrologiques, à l'élaboration du concept d'hydrosystème (Amoros and Petts, 1993).

Le concept d'hydrosystème, né dans les années 1980, est le produit d'une interaction entre scientifiques issus de trois disciplines majeures : géographie, écologie et hydrogéologie. L'hydrosystème se définit comme un « *système écologique complexe, organisé hiérarchiquement, et constitué de l'ensemble des biotopes et des biocénoses d'eau courante, d'eau stagnante, semi-aquatiques et terrestres, aussi bien épigés que souterrains, établis dans la plaine alluviale et dont le fonctionnement dépend directement ou indirectement du cours actif du fleuve. Il s'agit d'un ensemble d'écosystèmes en interaction qui forment un écocomplexe* » (Amoros and Petts, 1993). Plus simplement, l'hydrosystème peut être assimilé à l'ensemble des écosystèmes aquatiques liés à un cours d'eau et contribuant à son fonctionnement.

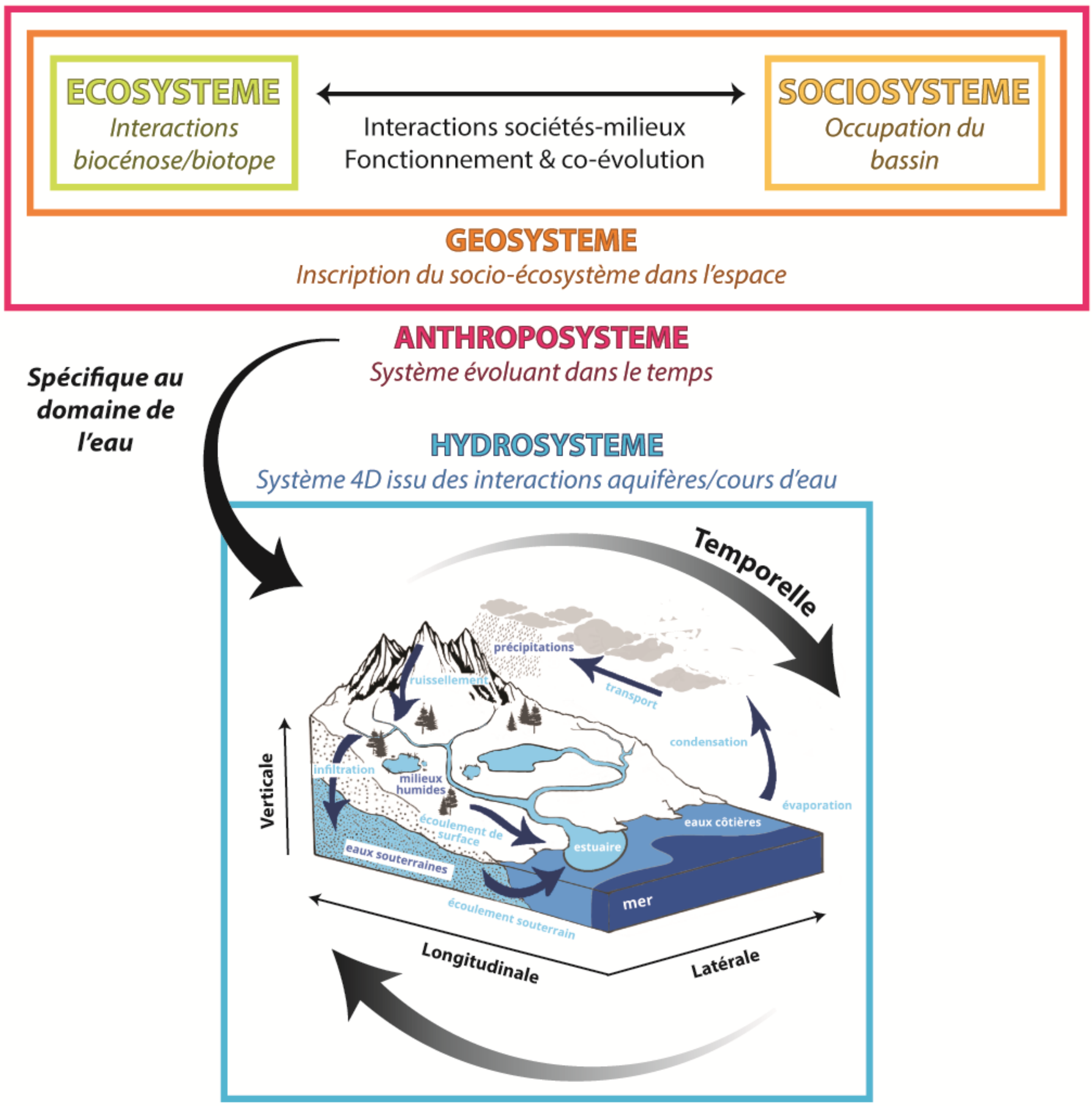

Figure 2 : Organisation et interaction entre les concepts d'écosystème, sociosystème, géosystème, anthroposystème et hydrosystème.

En fournissant un cadre spatial d'organisation de l'habitat, le géographe a permis à l'écologue de comprendre l'organisation et la structuration des peuplements aquatiques. Dans un même temps, l'hydrogéologue a formalisé le rôle des nappes alluviales dans la dynamique des fleuves et introduit la notion de variabilité temporelle et interannuelle dans le fonctionnement du système. De plus en plus, les hydrosystèmes sont appréhendés comme des systèmes interactifs avec, en plus, une dimension temporelle : le système évolue dans le temps. Cette dimension temporelle constitue la quatrième dimension de l'hydrosystème. Elle nécessite une réflexion multidisciplinaire pour dégager les différents scénarii de trajectoire dans le futur (Lévêque, 2011). En plus de considérer le fonctionnement du système comme multidimensionnel et interactif, le concept d'hydrosystème se distingue par sa volonté d'intégrer la question de l'anthropisation. On commence alors à s'interroger sur le poids de l'histoire, les conflits d'usage et les relations entre la santé et l'environnement. A la fin des années 90, on assiste au développement du concept d'anthroposystème, défini ensuite par Lévêque (2013). Il s'agit d'un système hybride et interactif entre deux ensembles co-évolutifs : systèmes naturels (écosystèmes) et systèmes sociaux (socio-écosystèmes), qui s'inscrit dans une échelle temporelle : passé, présent, futur. Avec ce concept, les systèmes naturels ne sont plus considérés comme utilisés et aménagés par l'homme ; c'est l'usage des milieux et des ressources qui modèle les structures sociales et les comportements des sociétés vis-à-vis de leurs territoires.

1.1.2. Importance de l'approche globale

L'étude des hydrosystèmes requiert l'évaluation quantitative et qualitative des ressources en eau souterraine et de surface. Toutes deux constituent des composantes intimement liées, caractérisées par des interactions fortes. Mieux contraindre le fonctionnement de la composante souterraine et ses interactions avec la surface est donc essentiel pour la compréhension globale de l'hydrosystème et des écosystèmes qui le composent. La compréhension du fonctionnement physique d'un hydrosystème passe alors par l'étude des ressources en eau disponibles (prévision des cycles hydrologiques, précipitations, recharge…) et de leur mobilité dans le milieu terrestre (hydraulique, écoulement dans la zone non saturée et saturée du sol, ruissellement…). Les interactions les plus significatives incluent i) la recharge des aquifères par l'infiltration d'eau de surface, ii) le soutien du débit de base des cours d'eau par les eaux souterraines, iii) la décharge des eaux souterraines comme émergence naturelle (sources, systèmes de grottes…) et iv) l'écoulement des eaux souterraines vers les milieux humides (tourbières, marais…) et/ou provenant des réservoirs (lacs, étangs et lagunes). A grande échelle, les échanges hydrologiques entre les eaux souterraines et de surface sont principalement régis par les propriétés hydrauliques du milieu (porosité, conductivité hydraulique, transmissivité, coefficient d'emmagasinement), la géométrie du système et le contexte climatique.

1.1.3. Forçages naturels et anthropiques

Les hydrosystèmes sont soumis à la double influence des forçages naturels et anthropiques (Ormerod et al., 2010; Tockner et al., 2010). Les forçages naturels sont principalement climatiques (vent, température, régime des précipitations…), géomorphologiques ou tectoniques. Cependant, leur expression par l'occurrence de phénomènes extrêmes (inondations, tempêtes, mouvements de terrain…) influence les comportements humains. Les mesures prises pour lutter contre ces catastrophes naturelles peuvent alors avoir un impact sur la qualité de l'eau et sur la biodiversité des milieux (barrages, digues, brise-lames…). Les activités humaines influencent l'hydrosystème via différents aspects et à différentes échelles (Heathwaite, 2010; Smol, 2010). A l'échelle locale, les contraintes exercées sont principalement de l'ordre technique, économique et social, en lien avec l'occupation des sols. Il s'agit alors d'évaluer l'impact sur le milieu des constructions humaines (habitat, industrie, extraction des ressources, barrages…), des prélèvements et rejets effectués (eaux usées domestiques, agricoles, industrielles) et des politiques de gestion de l'eau, des conflits d'usage… A l'impact du développement local des activités humaines sur l'hydrosystème s'ajoute, depuis plusieurs années maintenant, l'évaluation de l'impact lié aux activités humaines à l'échelle globale. Les perturbations, causées notamment par l'émission de gaz à effet de serre sur le cycle hydrologique, le réchauffement climatique (et la montée consécutive du niveau marin) ou encore la modification des chemins d'écoulement sont autant de nouveaux forçages qui influent sur les hydrosystèmes à l'échelle locale. Cette influence humaine peut même parfois prendre le pas sur les moteurs naturels (Tockner et al., 2010).

1.2. Écosystèmes tributaires des eaux souterraines

Les eaux souterraines jouent un rôle important dans les hydrosystèmes en alimentant en eau les écosystèmes terrestres et aquatiques, assurant ainsi leur stabilité et la diversité des habitats (Marmonier et al., 2019). Ces écosystèmes constituent alors des écosystèmes tributaires des eaux souterraines, expression dérivée de l'anglais « groundwater dependent ecosystems » (GDEs). La dépendance aux eaux souterraines fait référence à la relation qui existe entre un aquifère et une zone humide, et à la mesure dans laquelle la zone humide reçoit ou perd de l'eau à partir de ou vers l'aquifère (Figure 3).

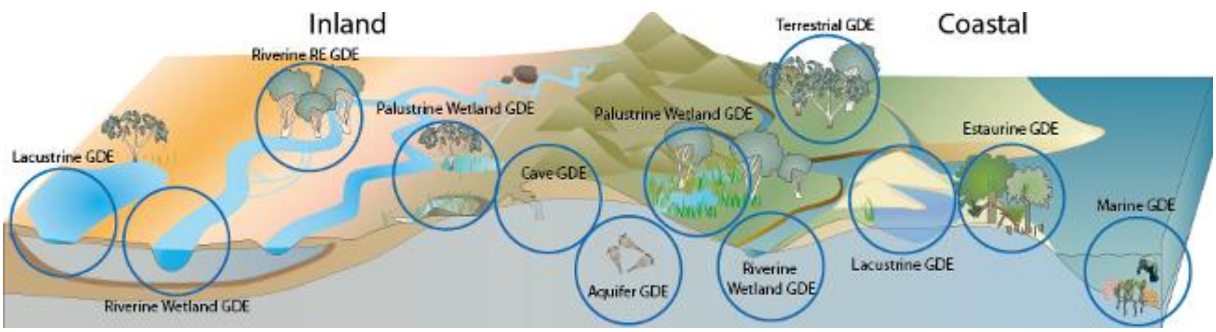

Figure 3 : Diversité des écosystèmes terrestres et aquatiques tributaires directement ou indirectement des eaux souterraines (Queensland Government, 2020)

Les GDEs sont définis comme "*des écosystèmes qui ont besoin d'un accès permanent ou intermittent aux eaux souterraines pour satisfaire tout ou partie de leurs besoins en eau afin de maintenir leurs communautés végétales et animales, leurs processus écologiques et leurs services écosystémiques* " (Richardson et al., 2011). Ils peuvent donc être reliés aux eaux souterraines de façon éphémère (en lien avec l'occurrence des précipitations ou de phénomènes de ruissellement exceptionnels), intermittente (alternance de périodes humides et sèches), saisonnière (alternance saisonnière de périodes humides et sèches) ou permanente. Les écosystèmes des aquifères sont évidemment intrinsèquement tributaires des eaux souterraines. Les GDEs peuvent être classés en trois catégories suivant leur type de dépendance aux eaux souterraines (Eamus et al., 2006):

- les GDEs qui utilisent **l'eau souterraine après sa décharge en surface**, cela comprend généralement tous les milieux humides alimentés par les eaux souterraines (riverains, palustres, lacustres, estuariens, marins) ;

- les GDEs qui bénéficient de **l'eau souterraine de subsurface** pour satisfaire une partie ou la totalité de ses besoins en eau, cela inclut notamment la végétation terrestre ;
- les GDEs **souterrains** : écosystèmes aquifères et cavernicoles.

Les GDEs assurent un large éventail de fonctions et de processus biologiques et physiques qui aident à la régulation et au maintien d'un paysage fonctionnel, tout en prodiguant des avantages à la société. Les bénéfices que l'Homme tire du fonctionnement des écosystèmes sont alors désignés par le terme de « services écosystémiques » (Dufour et al., 2016; Plant et al., 2016). Le concept de « services écosystémiques » tente d'exprimer cette relation complexe entre les communautés humaines, leur environnement et les êtres vivants non humains auxquels elles sont liées (Sartre et al., 2014). Les services écosystémiques incluent quatre types de services : les services d'approvisionnement (pêche, matériaux de construction…), les services de soutien (habitats, aires de reproduction, de nutrition, de migration ou de refuge), les services de régulation (stockage de carbone, régulation du climat, la protection contre les inondations et l'érosion...) et les services culturels (tourisme, loisirs, bénéfices esthétiques…) (Blanchart et al., 2017).

1.3. Spécificités des hydrosystèmes côtiers

1.3.1. Hydrodynamique des zones côtières

En conditions naturelles, sans pompage, l'eau souterraine douce s'écoule des zones de recharge vers les zones exutoires, ou plus couramment appelées « zones de décharge ». En contexte littoral, l'eau souterraine douce continentale rencontre l'eau salée marine dans une zone de transition appelée « biseau salé ». La décharge totale de l'eau souterraine (SGD, pour « *submarine groundwater discharge* ») correspond à l'ensemble des flux d'eau à travers le plancher océanique. La SGD se compose de deux processus distincts : i) la décharge d'eau douce continentale issue d'un aquifère côtier vers la mer (FSGD pour « *fresh submarine groundwater discharge* ») et l'eau de mer recyclée au sein de l'aquifère (RSGD pour « *recirculated submarine groundwater discharge*) (Figure 4). La décharge d'eau douce souterraine continentale est induite par le gradient hydraulique. La recirculation d'eau marine quant à elle est induite par plusieurs mécanismes océaniques (marée, phénomène de convection, houle…). La décharge des eaux souterraines peut provenir d'aquifères libres (non-confinés) ou captifs (confinés). Elle est spatialement diffuse et discontinue.

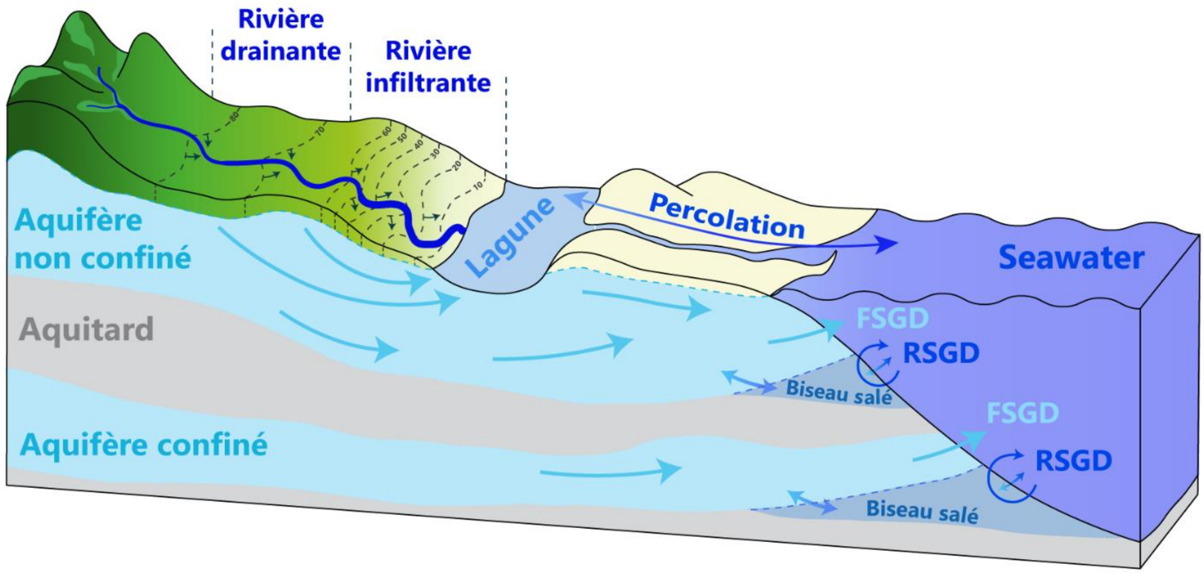

Figure 4 : Coupe schématique d'une zone côtière montrant les interactions entre les eaux de surface (douces fluviales, saumâtres lagunaires et salées marines), entre les eaux de surface et les eaux souterraines ainsi que les processus d'écoulement et de décharge des eaux souterraines (FSGD, RSGD), modifiée d'après Erostate et al. (2020). FSGD représente la décharge d'eau souterraine continentale douce induite par le gradient hydraulique de l'aquifère ; RSGD représente la décharge d'eau souterraine marine recyclée dans l'aquifère induite par les forces physiques telles que la marée et la convection.

1.3.2. Le cas particulier des lagunes côtières

Les littoraux, parce qu'ils constituent des zones de contact entre différents types d'écosystèmes, jouent un rôle essentiel dans la dynamique de la biodiversité. Ils hébergent des organismes appartenant à chacun des deux systèmes contigus (terrestre et océanique) ainsi que des espèces spécifiques à ces environnements de transition (Gibert et al., 1994).

Les lagunes côtières et leurs zones humides, regroupées ici sous le terme de « GDEs côtiers », se distinguent des autres environnements côtiers par leur importance à la fois environnementale, sociale et économique (Newton et al., 2014; Wit et al., 2017). Les GDEs côtiers comptent parmi les écosystèmes les plus productifs et les plus utilisés au monde.

Ils constituent des habitats importants pour le soutien de la biodiversité, et notamment des habitats vitaux pour les bivalves, les crustacés, les poissons et les oiseaux (Basset et al., 2013). Ils jouent également le rôle de nurserie, de refuge contre la prédation ou encore d'aire d'alimentation pour les espèces estuariennes, marines et terrestres (Heck and Thoman, 1984; Harris et al., 2004). Ils abritent une large partie de la population humaine, qui dépend directement de ces écosystèmes (Willaert, 2014) et fournissent non seulement des moyens de subsistance mais aussi de nombreux avantages pour la santé et le bien-être humain (Newton et al., 2014). Ils prodiguent une large diversité de services écosystémiques tels que l'alimentation (principalement poissons et crustacés), les loisirs et l'écotourisme, mais aussi l'équilibre hydrologique, la régulation du climat, la protection contre les inondations et la protection des côtes, la purification des eaux ou encore la production d'oxygène... (Newton et al., 2018). Les retombées monétaires de ces services écosystémiques représentent souvent une contribution majeure à l'économie régionale ou nationale d'un pays (Newton et al., 2018). Les services culturels et d'approvisionnement sont les plus rentables. A titre d'exemple, les services culturels rendus par la lagune de Venise (Italie) rapportent plus de 12 millions d'euros/km^2, soit plus de 500 millions d'euros/an (Newton et al., 2018). Les services d'approvisionnement assurés par la lagune de Cigu (Taïwan) et l'Étang de Thau (France) rapportent respectivement 750 et 450 millions d'euros/an (Newton et al., 2018).

1.3.3. Importance des eaux souterraines pour les GDEs côtiers

Au cours des dernières décennies, diverses études ont démontré l'importance des eaux souterraines pour les écosystèmes côtiers. Elles contrôlent notamment l'évolution, la pérennité et la résilience de ces écosystèmes par i) la quantité, la localisation, la fréquence et la durée de l'approvisionnement en eau douce (Jolly et al., 2008; Rodríguez-Rodríguez et al., 2008; Bertrand et al., 2012, 2014) et par ii) leurs caractéristiques chimiques (Burnett et al., 2006; Moore, 2010), telles que la qualité de l'eau (Ganguli et al., 2012), la salinité (Menció et al., 2017), les concentrations en nutriments, notamment en azote (Szymczycha et al., 2012; Ji et al., 2013; Rodellas et al., 2015, 2018; Hugman et al., 2017) et la température (Brown et al., 2007; Richardson et al., 2011). Les eaux souterraines peuvent par conséquent représenter une menace potentielle, en agissant comme un vecteur important de polluants vers les zones côtières (Ganguli et al., 2012; Sánchez-Martos et al., 2014; Hugman et al., 2017).

L'importance des eaux souterraines est encore exacerbée dans les régions soumises à un climat semi-aride à aride, en cas d'épisodes de sécheresse prolongés ou en situation de stress hydrique. Les eaux souterraines permettent alors de compenser les apports réduits ou inexistants en eau de surface (Eamus et al., 2016) et d'assurer le maintien des écosystèmes. Sous les effets du changement climatique, affectant notamment les températures (Bille et al., 2009; Hallegatte et al., 2009) et le cycle hydrologique (IPCC, 2014), les eaux souterraines sont amenées à devenir une composante clé du maintien de la biodiversité globale des hydrosystèmes côtiers (Datry et al., 2007, 2012).

1.4. GDEs côtiers en contexte méditerranéen : caractérisation, challenges et stratégies de gestion

Malgré leur importance, les GDEs côtiers constituent l'un des écosystèmes les plus menacés au niveau mondial. Plus de 50% des zones humides ont disparu au cours du 20ème siècle dans certaines régions d'Europe (Millennium Ecosystem Assessment, 2005). Au sein du bassin méditerranéen, les données collectées suggèrent une perte probable de 50% des zones humides (Perennou et al., 2012). Sur la même période, les pertes relatives aux zones humides côtières à l'échelle mondiale sont estimées entre 64% et 71% (Gardner et al., 2015).

Face à ce constat alarmant, cette section propose une synthèse sans précédent sur les problématiques de gestion des GDEs côtiers (lagunes côtières et zones humides environnantes) en contexte méditerranéen. Sur ces littoraux, l'importante hausse démographique est couplée au développement croissant des activités humaines (urbanisation, agriculture et industrie). L'augmentation des volumes d'eau extraits des aquifères côtiers induit une diminution de la ressource disponible pour les besoins humains et environnementaux et perturbe l'équilibre souterrain entre eau douce continentale et eau salée marine. De plus, les contaminations liées aux activités développées en amont sur le bassin versant peuvent s'infiltrer et dégrader la qualité de la ressource en eau souterraine. Celle-ci devient alors un important vecteur potentiel de contaminants et de nutriments, menaçant directement l'équilibre des écosystèmes côtiers. Les prélèvements d'eau, l'assèchement, la pollution, la destruction des habitats ou la surexploitation constituent les principales causes de leur dégradation (Millennium Ecosystem Assessment, 2005). A l'ensemble des perturbations causées par les activités humaines s'ajoutent les effets présagés du changement climatique avec notamment la modification attendue de la recharge des aquifères, du stockage et de la qualité des eaux souterraines. Ces modifications devraient être plus prononcées dans les régions arides et surtout dans le bassin méditerranéen, considéré comme un point chaud du changement climatique. L'importance des eaux souterraines pour l'approvisionnement en eau douce des GDEs côtiers est ainsi soulignée, tout comme celle d'instaurer des politiques de gestion durables et adaptées à ces milieux atypiques.

Devant l'absence flagrante de politiques de gestion appropriées, cette évaluation critique de la situation actuelle des GDEs en contexte méditerranéen, portée par une approche transdisciplinaire, permet de mettre en évidence les principaux dysfonctionnements et défis relatifs à la gestion des GDEs. Les prises de conscience internationales concernant les enjeux environnementaux (Sommet de la Terre de Stockolm en 1972, rapport Bruntland sur le développement durable en 1987, Sommet de la Terre à Rio en 1992) ont posé les bases des stratégies et des politiques de gestion des ressources en eau avec le développement du concept de Gestion Intégrée des Ressources en Eau (GIRE) et de Gestion Intégrée des Zones Côtières (GIZC). Ces deux concepts prônent la mise en valeur et la gestion coordonnée des ressources en eau, des terres et des ressources connexes, afin de maximiser le bien-être économique et social qui en résulte de manière équitable, sans compromettre la durabilité des écosystèmes essentiels. Pourtant, la mise en œuvre d'une gestion intégrée et collaborative fait souvent défaut. Les GDEs côtiers ne bénéficient pas d'un statut particulier qui tienne compte de leur complexité, notamment à cause du manque de connaissances quant à leur fonctionnement global.

La préservation des GDE côtiers est subordonnée à la stabilité dans le temps des réserves d'eau douce (eau souterraine et de surface) en quantité et qualité suffisantes. Cependant, chaque GDEs côtier est un cas unique et la détermination des besoins qualitatifs et quantitatifs est difficile. La complexité des interactions entre les diverses masses d'eau (souterraine, de surface, de transition et marine) exige une compréhension du système dans son ensemble. La stratégie de gestion doit considérer le plan d'eau lagunaire, la zone humide environnante et les eaux souterraines comme un ensemble indissociable de vases communicants dont la nature des échanges évolue dans le temps et l'espace. L'importance des eaux souterraines dans le fonctionnement des GDEs côtiers est de plus en plus reconnue et les études scientifiques visant à mieux comprendre leur rôle se développent. Ces avancées devraient permettre de définir des échelles de gestion plus appropriées, prenant en compte l'ensemble des masses d'eau qui influencent le fonctionnement des GDEs côtiers.

A l'heure actuelle, d'importantes lacunes résident encore dans la définition, la législation et la gestion des GDEs côtiers. L'absence d'une définition appropriée faisant consensus chez les juristes et scientifiques ne facilite pas l'établissement d'une politique de gestion durable. Certaines régions méditerranéennes ont commencé à inclure la protection des GDEs dans leurs politiques de gestion de l'eau cependant, aucune considération particulière n'est donnée aux GDEs côtiers. La mauvaise coordination entre gestion des eaux souterraines et gestion des eaux côtières affecte gravement le continuum physique, écologique et social des bassins hydrographiques et de leurs zones côtières. De plus, le manque d'implication, d'appropriation et de collaboration entre gestionnaires et usagers de l'eau, à l'échelle locale, nationale et internationale nuit à l'élaboration et à l'instauration de politiques de gestion adaptées. Pour cela, un investissement des usagers de l'eau et des gestionnaires locaux est clairement un prérequis sans lequel tout effort déployé n'aurait que de faible chance de s'avérer fructueux.

Dans le contexte mondial de pressions anthropiques sans précédent, de crises hydro-alimentaires et de changements climatiques, la prise en compte des GDEs côtiers représente un enjeu majeur pour le développement socio-économique et environnemental de nombreuses zones côtières. Les stratégies de gestion intégrée des hydrosystèmes côtiers doivent maintenant évoluer pour considérer sur un même pied d'égalité les besoins environnementaux et les contraintes socio-économiques. De cette façon, les hydrosystèmes côtiers pourraient réellement bénéficier de conditions environnementales permettant d'assurer leur durabilité.

Ce travail a fait l'objet de la rédaction d'un article de review présenté ci-après, publié dans la revue « *Water Research* » en janvier 2020.

Groundwater dependent ecosystems in coastal Mediterranean regions: Characterization, challenges and management for their protection

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Review

Groundwater dependent ecosystems in coastal Mediterranean regions: Characterization, challenges and management for their protection

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A B S T R A C T

Coastal lagoons deliver a wide range of valuable ecosystem goods and services. These ecosystems, that are often maintained by direct or indirect groundwater supplies, are collectively known as groundwater dependent ecosystems (GDEs). The importance of groundwater supplies is greatly exacerbated in coastal Mediterranean regions where the lack of surface water and the over-development of anthropogenic activities critically threaten the sustainability of coastal GDEs and associated ecosystem services.

Yet, coastal GDEs do not benefit from a legal or managerial recognition to take into account their specificity. Particular attention should be paid to the characterization of environmental and ecological water requirements. The hydrogeological knowledge about the management and behavior of coastal aquifers and GDEs must be strengthened. These investigations must be supplemented by a stronger assessment of potential contaminations to develop local land-uses and human activities according to the groundwater vulnerability. The quantitative management of water resources must also be better supervised and/or more constrained in order to ensure the water needs necessary to maintain coastal GDEs.

The transdisciplinary approach between hydrogeology, hydrology, social sciences and law is essential to fully understand the socio-economic and environmental complexity of coastal GDEs. Priority must now be given to the development of an appropriate definition of coastal GDEs, based on a consensus between scientists and lawyers. It is a necessary first step to develop and implement specific protective legislation and to define an appropriate management scale. The investment and collaboration of local water users, stakeholders and decision-makers need to be strengthened through actions to favor exchanges and discussions. All water resources in the coastal areas should be managed collectively and strategically, in order to maximize use efficiency, reduce water use conflicts and avoid over-exploitation. It is important to continue to raise public awareness of coastal aquifers at the regional level and to integrate their specificities into coastal zone management strategies and plans. In the global context of unprecedented anthropogenic pressures, hydro-food crises and climate change, environmental protection and preservation of coastal GDEs represents a major challenge for the sustainable socio-economic and environmental development of Mediterranean coastal zones.

Contents

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1. Introduction

Coastal lagoons cover about 13% of the coastlines from arid to humid environments (Kjerfve, 1994). Being transitional areas from land to sea, the water balance of coastal lagoons is resulting from both terrestrial (fresh groundwater and surface water) and marine water influences. This dual influence allows the development of specific ecosystems that provide a wide range of ecosystem goods and services (Newton et al., 2014, 2018). Over the past few decades, several studies have highlighted the importance of groundwater in maintaining the physico-chemical conditions of these sensitive ecosystems. Coastal lagoons and surrounding wetlands may then constitute “groundwater-dependent ecosystems” (GDEs) (Krogulec, 2016; Menció et al., 2017) and are referred in the document as “coastal GDEs".

The importance of groundwater is further exacerbated in regions suffering from water stress, when surface water is chronically unavailable. Groundwater inputs support or compensate for surface water inputs and play a vital role in maintaining coastal GDEs. This problem is encountered in a majority of coastal regions with an arid or semi-arid Mediterranean climate (Fig. 1) (Köppen, 1936) such as the Mediterranean basin (European Union -EU - and non-EU countries) but also on the southwestern coasts of Australia, Chile and the State of California (United States) and on the southern coast of South Africa. In these regions, referred to throughout this document as “Mediterranean regions”, the lack of surface water is combined with a high anthropogenic pressure (UNEP/MAP, 2012). Population growth proceed together with the development and expansion of human activities, such as urbanization, agriculture, tourism and industrial activities (Lotze et al., 2006). Increasing human water needs often lead to overexploitation of aquifers and/or degradation of groundwater quality, which present a risk both to the well-being of human activities and to the freshwater needs of coastal GDEs.

These degradations are expected to be worsen under the effects of climate change. Climatic disturbance in terms of increasing temperatures (Bille et al., 2009; Hallegatte et al., 2009), global hydrological cycle (IPCC, 2014) and sea level rise (FitzGerald et al., 2008; Carrasco et al., 2016; Benjamin et al., 2017) should greatly affect the groundwater and coastal GDEs. This is true not only for the Mediterranean basin, considered as a Hot Spot of climate change, but also for all the Mediterranean regions.

Since the 1990s and the Rio de Janeiro Earth Summit, the conservation, the maintenance of potentialities and the improvement of the ecological status of the coastal water bodies constitute a major concern. Nowadays, a first statement can be made on the progress and limitations of groundwater management strategies and consideration given to coastal GDEs in coastal Mediterranean regions. To this aim, this review proposes to:

- Expose the specificities of coastal GDEs and the key role of groundwater in their sustainable development
- Highlight the vulnerability of coastal GDEs to the socio-economic development and climate conditions of Mediterranean regions
- Revise the consideration given to GDEs and particularly to coastal GDEs in the management policies of Mediterranean regions and discuss their implication for the sustainability of coastal GDEs.

2. Specificities and importance of coastal GDEs

2.1. The wide diversity and essential functions of coastal GDEs

GDEs are defined as *“ecosystems that require access to groundwater on a permanent or intermittent basis to meet all or some of their water requirement so as to maintain their communities of plants and animals, ecological processes and ecosystem services”* (Richardson et al., 2011). This definition clearly expresses the crucial role of groundwater in the functioning of GDEs. However, the multitude of processes and services grouped under the terms “ecological processes” and “ecosystem services” does not necessarily make it possible to understand all the specificities and complexity inherent to certain types of GDEs, such as coastal GDEs. Table 1 summarizes the morphologic and hydrological characteristics, the hydrological

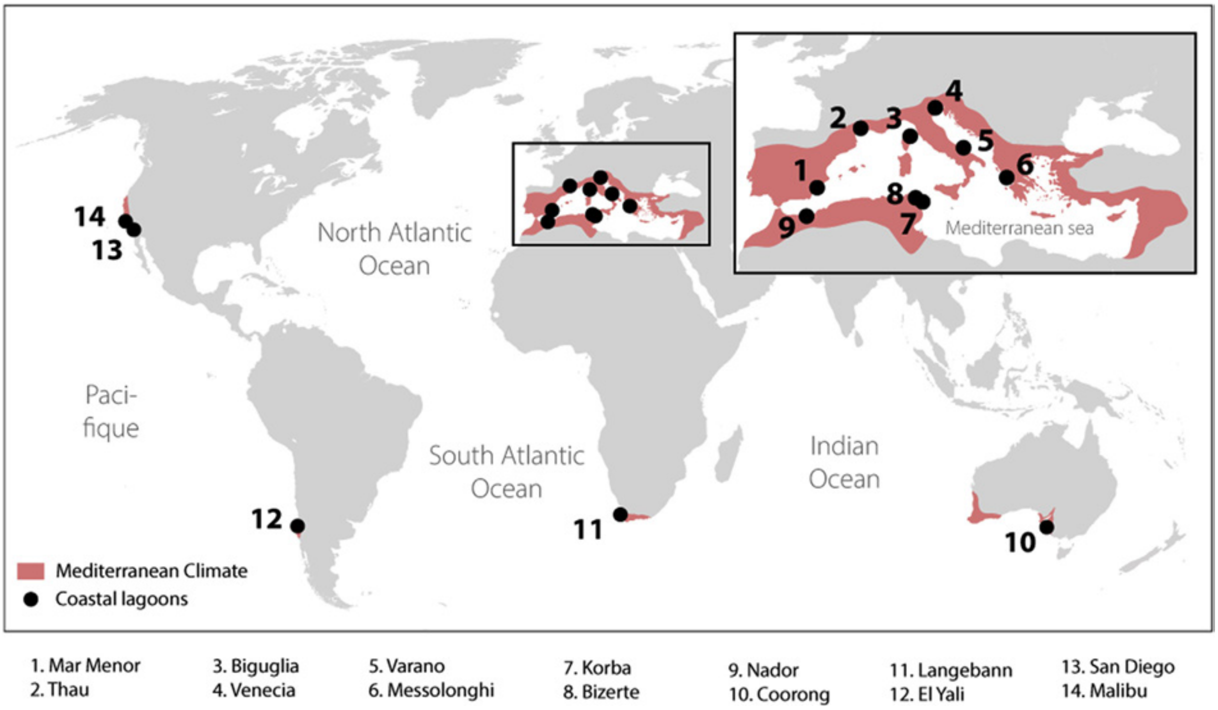

Fig. 1. Coastal regions under Mediterranen climate and location of the 14 coastal lagoons exposed in Table 1.

knowledge and the protection and conservation status of 14 of the most studied lagoons present in Mediterranean regions subject to Mediterranean climate (Fig. 1) (Newton et al., 2018; Pérez-Ruzafa and Marcos, 2008).

The coastal GDEs are distinguished by their diversity, making each of them a special case. This diversity is expressed on several levels. From a morphological point of view, water bodies of coastal GDEs are separated from the sea/ocean by a barrier, connected at least intermittently to the ocean by one or more restricted inlets (Kjerfve, 1994). According to the most widely used classification, these coastal lagoons can be classified into three categories including (i) choked, (ii) restricted and (iii) leaky lagoons Kjerfve (1994). These categories reflect the importance of interactions between coastal lagoons and seawater. Choked lagoon are connected to the sea by a single or few narrow and shallow entrances, resulting in delayed and dampened tidal oscillation or low water exchange with the open sea. Leaky lagoons are connected by many entrances to the adjacent sea and are therefore characterized by almost unimpaired water exchange. The stretch of coastal lagoon can greatly vary, from <0.01 km^2 to more than 10 000 km^2, as is the size of the hydrological watersheds, without an obvious proportionality relationship between the two (Table 1). If the mean depth can also vary, coastal lagoons still remain shallow water environments, generally characterized by shallow mean depth (<2 m) (Table 1).

Although rainfall, pounding of surface flows or flooding are an important source of water for most of coastal GDEs, groundwater plays also a role in many coastal wetlands (Le Maitre et al., 1999). Coastal GDEs can be completely dependent on groundwater discharge, whilst others may have limited dependence, such as only under dry conditions (Howe et al., 2007). Thus, depending on the hydrologic balance, water bodies of coastal GDEs could vary from coastal fresh-water lake to a hypersaline lagoon.

The fauna and flora that make up coastal GDEs are also very diverse. The type of vegetation and wildlife is mainly defined by the salinity of the water and the moisture level of the environment (permanent, semi-permanent or ephemeral wetlands) but also the location and climate. Several thousand plant species grow in coastal wetlands such as reeds, grasses and shrubs (Frieswyk and Zedler, 2007; Lemein et al., 2017; Ramírez and Álvarez, 2017). Hundreds of animal species can also be listed, including fish, reptiles, mammals, frogs and birds. The degree of dependence of wildlife on coastal GDEs ranges from those who need wetlands for part of their life cycle to those who are totally dependent on them.

The environmental importance of coastal GDEs is greatly recognized for most of them, as evidenced by the establishment of various protection or conservation status (Table 1). Because of their relatively low flushing rates, the important availability of nutrients allows high rates of primary production (phytoplankton and aquatic plants) thereby supporting high rate of secondary production (fisheries nurseries) compared to other aquatic ecosystems (Nixon, 1995). Coastal GDEs contribute to the overall productivity of coastal waters by supporting a variety of habitats, including salt marshes, seagrasses or mangroves. These habitats host specific and sensitive ecosystems and provide a rich support for biodiversity, including vital habitats for many fish, shellfish and bivalves (Basset et al., 2013). They constitute also refuge from predation, nursery and feeding habitats for estuarine, marines and terrestrial species (Heck and Thoman, 1984; Harris et al., 2004). Many coastal GDEs support a variety of migratory water bird and shore bird species. Some birds depend on coastal GDEs almost totally for breeding, nesting, feeding, or shelter during their annual cycles. The main migratory birds utilizing the coastal GDEs are ducks, shorebirds, gulls, terns and flamingos.

2.2. Ecosystem services and coastal GDEs

Coastal GDEs harbor a large part of the human population that depends directly on these ecosystems and provide not only livelihoods but also numerous benefits to human health and welfare (Newton et al., 2014, 2018). Coastal GDEs have therefore a socio-economic interest which makes them complex social-ecological

Table 1
Morphological and hydrological characteristics, protection and conservation status and level of knowledge on hydrosystems' behavior and groundwater dependence for 14 of the most studied coastal lagoons under Mediterranean climate according to data available in scientific literature. Lack of available information is symbolized by a "?".

	Lagoons	Countries	Characteristics				Conservation and protection status	Hydrosystem behavior and groundwater dependence		References
			Surface (km^2)	Mean depth (m)	Main aquifer formation(s)	Hydrological watershed (km^2)		Strongly suspected	Demonstrated	
1	Mar menor	Spain, South-East	135	4.5	5 aquifers: Detrital deposits Sandston Limestone Sandy limestone and conglomerate Marble	1200	Ramsar site Special bird habitat Regional park Site of Community importance Specially protected area of Mediterranean importance		Studied and relatively well known	De Pascalis et al. (2012); Baudron et al. (2014); Velasco et al. (2018); Alcolea et al. (2019)
2	Thau	France, South-Est	75	4	Karstified limestone	280	Special bird habitats Natura 2000 Water Framework Directive site		Studied but lack of data to understand the global behavior	Tournoud et al. (2006); Fleury et al. (2007); Stieglitz et al. (2013); Loiseau et al. (2014); La Jeunesse et al. (2016)
3	Biguglia	France, Corsica island	14	1.2	Detrital deposits	182	Ramsar site Nature Reserve Special bird habitats Natura 2000 Water Framework Directive site		Studied and relatively well known	Lafabrie et al. (2013); Erostate et al. (2018); Jaunat et al. (2018); Erostate et al. (2019); Leruste et al. (2019)
4	Venice	Italy, North-East	550	1.5	Detrital deposits	1800	Ramsar site Natura 2000 Special bird habitat		Largely studied and relatively well known	Ravera (2000); Ferrarin et al. (2008); Rapaglia et al. (2010); Da Lio et al. (2013); Mayer et al. (2014)
5	Varano	Italy, South-East	65	3.5	2 main aquifers: Detrital deposits	300		Under-documented		Ferrarin et al. (2010); Roselli et al. (2013); Fabbrocini et al. (2017)
6	Messolonghi central lagoon	Greece, North-West	80	0.8	2 main aquifers: Limestone and breccia Detrital deposits	1979	Ramsar site National Park Important Bird Area Natura 2000	Under-documented		Alexakis (2011); Karageorgis et al. (2012); Stamatis et al. (2013)
7	Korba	Tunisia, plain of Cap Bon	3.1	1	Detrital deposits	27	Ramsar site Important Bird Area	Under-documented		Kouzana et al. (2010); Zghibi et al. (2013); Slama and Bouhlila (2017)
8	Bizerta	Tunisia	128	8	Detrital deposits	380	Ramsar site UNESCO-MAB Reserve	Under-documented		Bouzourra et al. (2015); PNUE-PAM, UNESCO-PHI, 2017
9	Nador	Marocco, Nord-Est	115	5	2 mains aquifers: Detrital deposits	?	Ramsar site Nature Reserve Site of biological and ecological interest	Groundwater contribution known but under-studied		Maanan et al. (2015); Mohamed et al. (2017); Aknaf et al. (2018)
10	Coorong	Australia, South-East	140	1.8	Limestone Sands	6	Ramsar site National Park		Studied and well known	Haese et al. (2008); Richardson et al. (2011); Leterme et al. (2015)
11	Langebaan	South Africa	40	3	Detrital deposits and calcrete	?	Ramsar site National Parks	Under-documented		Flemming (1977)
12	El Yali	Chile	115	0.5	Detrital deposits	?	Ramsar site National reserve	Groundwater contribution known but under-documented		Dussaillant et al. (2009); Vidal-Abarca et al. (2011)
13	San Diego	California (U.S.A)	42	5	Detrital deposits	146	National Wildlife Refuge	Under-documented		Delgadillo-Hinojosa et al. (2008)
14	Malibu	California (U.S.A)	0.05	?	Detrital deposits	280		Groundwater contribution known but under-documented		Dimova et al. (2017); Hoover et al. (2017)

systems (Newton et al., 2014; Wit et al., 2017). Since the 1970s, and more particularly in the 2000s, the concept of "ecosystem services" has attempted to express the complex relationship between human communities, their environment and the non-human living beings to which they are linked (Sartre et al., 2014). The "ecosystem services" can be defined as the full range of benefits that humans derive from the functioning of ecosystems. Ecosystem services include 4 major types of services (Blanchart et al., 2017):

- Provisioning services: correspond to direct products provided or produced by ecosystems such as water, food, construction materials,
- Regulating services: include benefits from regulation of ecosystem processes such as carbon storage, climate regulation, flood and erosion protection,
- Cultural services: include nonmaterial benefits from ecosystems such as recreation, aesthetic or educational benefits,
- Supporting services: are related to necessary factors for producing ecosystem services (photosynthesis, nutrient cycle, refuge areas …).

Ecosystem services are linked to the ecological structure and functions of the environment. In coastal GDEs, many ecosystem services are derived or supported by the presence of groundwater inflow because of its role in regulating the hydrology of wetlands and lagoons (UNEP-MAP/UNESCO-IHP, 2015). One of the main ecosystem services provided by coastal GDEs is related to provisioning services (livestock, fishing, aquaculture) (UNEP-MAP/UNESCO-IHP, 2015). Coastal GDEs are highly productive and food provisioning can often be key for regional economy (Newton et al., 2014). For example, the Ria Formosa in Portugal provided up to 90% of the national production of clams (Newton et al., 2003). Coastal GDEs also have a very important place in the hydrological cycle. They contribute to water flow regulation and control and therefore help to flood protection. They also participate to water retention, quality (salinity regulation) and purification. Finally, cultural services, *e.g.* cultural heritage, tourism or aesthetics are also very profitable for several coastal GDEs. In some specific case, such as the Venice lagoon (Italy), cultural services can exceed 5.10^8 euros/year (Newton et al., 2018).

The various protection and/or conservation status applied to coastal GDEs (Table 1) does not necessarily involve a high level of knowledge of the hydrosystems' behavior. For a large majority, the role and the dependence on groundwater is largely under studied, even if it is suspected (Table 1). Very few coastal lagoons have a sufficient level of knowledge to understand their level of dependence to groundwater (Table 1) and then developed sustainable methods/policies to ensure their conservation. Moreover, even in the case of good knowledge of hydrological functioning and establishment of a conservation/protection status, it does not seem to guarantee the good state of these environments (Leterme et al., 2015, Leruste et al., 2019). The lack of hydrological knowledge then appears to be as much a problem as the lack of specific protection status adapted to the particular cases of the GDES.

2.3. Understanding the dependence on groundwater supplies

Under natural conditions, without pumping, fresh groundwater flows from recharge to discharge areas (Fig. 2). Local groundwater flow is mostly near the surface and over short distances, i.e. from a higher elevation recharge area to an adjacent discharge area. In this case, the discharge of the aquifer (Fig. 2) occurs as diffuse outflow, as for coastal GDEs. Coastal GDEs are thus relying on the surface expression of groundwater (Richardson et al., 2011). On a larger scale, over long distances, groundwater flow is preferentially at greater depths and fresh groundwater meets salt marine water at depth in the transition zone. The discharge of groundwater is composed by two processes: i) the discharge of fresh groundwater (fresh submarine groundwater discharge, FSGD) toward the sea and the discharge of saline groundwater (recirculated submarine groundwater discharge, RSGD) (Fig. 2). Groundwater supplies to coastal GDEs can originate from one or several aquifer formations of variable nature and extension (Table 1). This dependence on groundwater can be variable, ranging from partial and infrequent dependence (seasonal or episodic) to total, continual dependence (Hatton and Evans, 1998).

Groundwater and surface water are the most often characterized by strong interactions (Fig. 2). These interactions result in groundwater discharge to the river (groundwater discharge, Fig. 2) or, conversely, in aquifer recharge through river and lake water infiltration (Fig. 2). Rivers and streams that flow all year (perennially flowing) are often groundwater dependent because a significant proportion of their daily flow is supported by the groundwater flow discharging into the river course (Acuña et al., 2005; Bonada and Resh, 2013; Datry et al., 2014). Groundwater is particularly important in arid and semi-arid regions and in case of extended dry periods, during which evaporation markedly exceeds precipitation and surface water is scarce or even disappeared (Eamus et al., 2006). Both groundwater and surface water flow toward the lagoon, which constitute the last collector of the watershed (Fig. 2). The discharge of groundwater toward coastal GDEs can be either directly into the wetland or indirectly via the river (Fig. 2).

For a long time, groundwater studies in coastal areas focused mainly on seawater intrusion impacting coastal aquifers. The groundwater has only recently been recognized as important contributors to hydrological and biogeochemical budgets of coastal environments such as coastal GDEs (Table 1) (Johannes, 1980; Burnett et al., 2001, 2006; Slomp and Van Cappellen, 2004; Moore, 2006, 2010; Rodellas et al., 2015; Luo and Jiao, 2016; Malta et al., 2017; Correa et al., 2019; David et al., 2019). The presence of groundwater drives the evolution, persistence and resilience of coastal GDEs and their ecosystems on at least two aspects including i) physical characteristics, such as the quantity, location, timing, frequency and duration of groundwater supply (Jolly et al., 2008; Rodríguez-Rodríguez et al., 2008; Bertrand et al., 2012, 2014) and ii) chemical characteristics (Burnett et al., 2006; Moore, 2010), such as water quality (Ganguli et al., 2012), salinity (Menció et al., 2017), nutrient concentrations (Szymczycha et al., 2012; Ji et al., 2013; Rodellas et al., 2015; Hugman et al., 2017) and temperature (Brown et al., 2007; Richardson et al., 2011). Although recognized as essential, the characterization of coastal hydrosystems' behavior still remains under studied in many cases (Table 1) due to the important monitoring and financial resources required to improve their understanding.

2.4. Groundwater dependence monitoring

The "Groundwater dependence" clearly expresses that the prolonged absence of groundwater as well as its quality degradation have a negative impact on the growth, health, composition, structure and function of the ecosystem. Potential threats to groundwater inflow toward the coastal GDEs can be assessed through the study of the groundwater flow paths, the spatial and temporal variability of groundwater discharge and surface/ground water interactions (Kløve et al., 2011). Yet, the groundwater dependence of coastal GDEs remains still difficult to characterize. This difficulty is exacerbated by the thinness of the unsaturated zone, *i.e.* the thickness of the soil between the soil surface and the top of the saturated zone, which allows important mixing between surface and ground waters. Differentiating and quantifying the

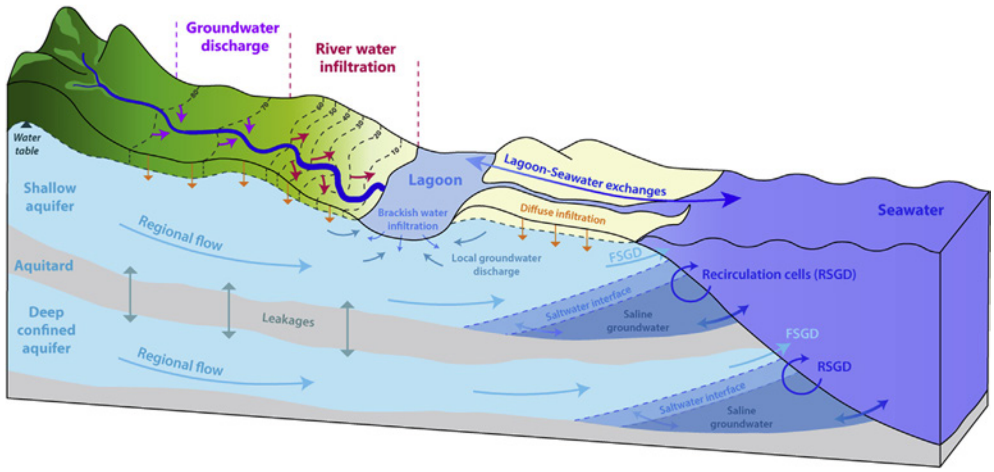

Fig. 2. Conceptual diagram of the hydrogeological behavior of coastal hydrosystems including a costal GDE. On a large scale, the discharge of groundwater is composed by two processes: i) the discharge of fresh groundwater toward the sea (fresh submarine groundwater discharge, FSGD) and ii) the discharge of saline groundwater,i.e. the discharge of a mixture of fresh water and seawater after recirculation through the transition zone (recirculated submarine groundwater discharge, RSGD).

contribution of these end-members is highly complex. A wide range of methodologies have been developed to improve the understanding of coastal GDEs (Sophocleous, 2002; Kalbus et al., 2006; Howe et al., 2007). First of all, the monitoring of groundwater levels and the establishment of piezometric map are often the first steps to highlight the groundwater dependence of coastal GDEs (Sena and Teresa Condesso de Melo, 2012). Then, in the particular case of coastal GDEs, the two main approaches commonly used to assess surface/ground water interaction are i) temperature, geochemical and isotopic tracers (Mudge et al., 2008; Santos et al., 2008; Schubert et al., 2011; Sánchez-Martos et al., 2014; Duque et al., 2016; Sadat-Noori et al., 2016; Dimova et al., 2017) and ii) numerical modeling (De Pascalis et al., 2009; Martínez-Alvarez et al., 2011; Sena and Condesso de Melo, 2012; Read et al., 2014; Menció et al., 2017). Less common approaches, such as geophysical method can also be carried out to obtain information on the spatial scales and dynamics of the fresh water–seawater interface, the rates of coastal groundwater exchange and the total fresh water discharge (Dimova et al., 2012).

3. Dominant human and climatic stressors on groundwater and consequences for coastal GDEs in Mediterranean regions

Although essential, coastal GDEs are one of the most threatened ecosystems in the world. Human activities are exerting increasing pressure on these sensitive systems or on the resources on which they depend, such as groundwater. Water withdrawal, drying, pollution, habitat destruction or overexploitation constitute the main causes of their degradation (Millennium Ecosystem Assessment, 2005). More than 50% of wetlands have disappeared during the 20th century in some regions of Australia and Europe (Millennium Ecosystem Assessment, 2005). Only in the Mediterranean basin, national or sub-national datasets suggest a probable loss of 50% of its wetlands (Perennou et al., 2012). In the specific case of coastal wetlands, global losses are estimated at between 64% and 71% during the 20th century (Gardner et al., 2015).

The characteristic overdevelopment of coastal Mediterranean regions has already led, for several decades, to a significant pressure on groundwater resources. The growing drinking, industrial or agricultural water requirements tend to the overexploitation of the coastal aquifers. Coastal aquifers are threatened by both horizontal exchanges with seawater and vertical infiltrations of pollutants. The development of human activities often constitutes an important source of pollutants and groundwater can constitute an important vector of pollution towards the coastal GDEs (Moore, 2006).

3.1. *The harmful human overdevelopment of coastal Mediterranean regions*

The strong and increasing urbanization as well as fast growing demography represent the two main pressures. For example, in Australia, more than 85% of the population is living within 50 km of the sea. The population density of Australian's coastal areas increased by 14% between 2001 and 2009, from 3.75 hab/km^2 to 4.27 hab/km^2 (Fig. 3a). A very important difference is observed for

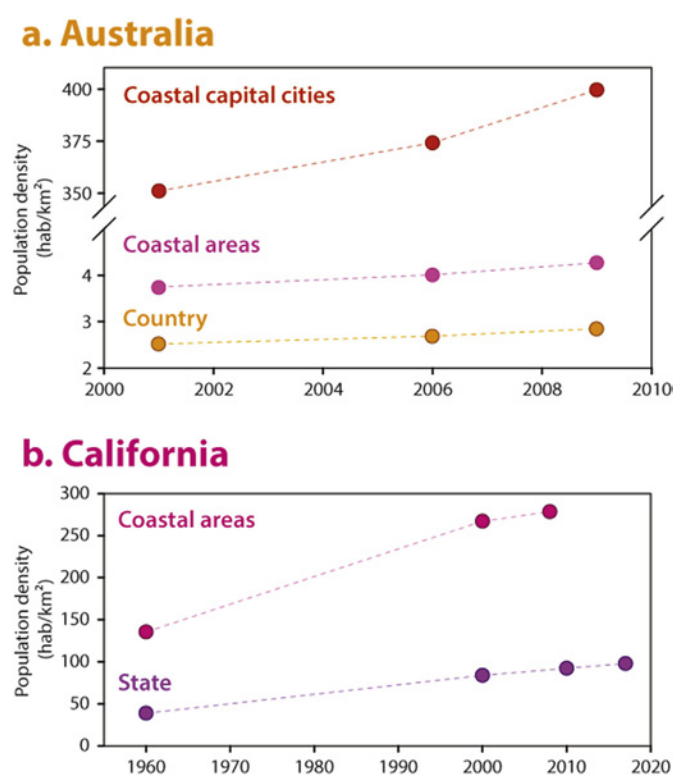

Fig. 3. Population density trends in Australia (a) and California (b).

the urban, coastal population. The population density measured in coastal capital cities is 94 times higher than the average population density of coastal areas (Fig. 3a).

In the Mediterranean basin, the coastal population grew from 95 million in 1979 to 143 million in 2000 and could reach 174 million by 2025 (UNEP/MAP, 2012) (Table 2). In the Mediterranean basin' population, France is the 3rd most populated country (after Turkey and Egypt) (UNEP/MAP, 2012) and allows for a good observation of the attractiveness of the Mediterranean coastline (Insee/SOeS, 2009). Indeed, among the 3 French coasts (Mediterranean, Atlantic and Channel coasts), the Mediterranean coast is clearly distinguished by a rapid population growth (Fig. 4) (Insee/SOeS, 2009). Between 1960 and 2010, the French Mediterranean coast recorded the highest population increase with 56%, although it is the least extensive coastline (Fig. 4). The highest growth of population rate is recorded in the Mediterranean island of Corsica, with an annual increase of 1.3% between 2006 and 2010. The coastal municipalities accounting for 80% of the Corsican population and 30% of the urbanization is concentrated within 1 km of the shoreline (SDAGE, 2015).

In USA, California tops the coastal populations chart. Currently, of the total population of 39.6 million in California, 69% is living in coastal areas (U.S. Census Bureau, 2019) and 95% is living in urban areas. Coastal population density is 3 times higher than the state' population density (Fig. 3b). In less than 60 years, coastal population density went up by a factor of 2.5, from 135.6 hab/km^2 in 1960 to 278.4 hab/km^2 in 2017 (U.S. Census Bureau, 2019) (Fig. 3b). In the major coastal cities, such as San Francisco and Los Angeles, population density exceeds several thousand inhabitants per km^2. In 2018, population density was 7003 hab/km^2 and 3230 hab/km^2 respectively.

This demographic growth is accompanied by a very fast development of urban infrastructure. In the Mediterranean basin, the urbanization increased from 54% in 1970 to 66% in 2010 (Table 2) and the urban coastal population could increase by 33 million between 2000 and 2025 (UNEP/MAP, 2012). The South and the East Mediterranean countries (Non-EU countries) are urbanizing more rapidly than the rest of the world. These that were essentially rural countries, with average urbanization of 41% in 1970, will become urban countries, with 66% urbanization by 2025 (UNEP/MAP, 2012). This tendency is also observed in Australia. Peri-urban and rural cadastral parcels are progressively replaced by urban areas leading to an increased artificialization of coastal areas (Clark and Johnston, 2017).

3.2. Perturbations induced by groundwater degradation

3.2.1. Reduction of groundwater inputs and coastal GDEs dewatering

The modification of fresh groundwater flowing to the lagoons disrupts the fragile balance of the coastal GDEs' ecosystems. As surface water is limited and increasingly affected by pollution and eutrophication, the exploitation of groundwater from coastal aquifers as a source of freshwater has become more intense (Bocanegra et al., 2013; Liu et al., 2017). The number of groundwater abstraction infrastructures have drastically increased. This process

Table 2
Demographic trends and rate of urbanization in the Mediterranean basin.

Mediterranean basin	1970	2000	2010	2025
Whole population (millions)	276	412	466	529
Coastal population (millions)	95	143	–	174
Urbanization rate (%)	54	–	66	–

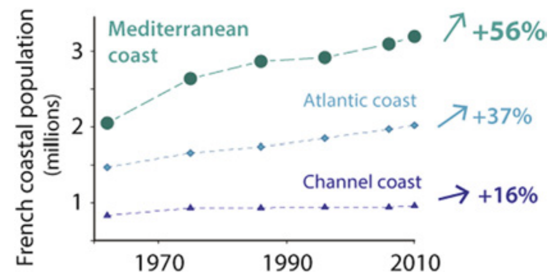

Fig. 4. Demographic trends on the three French coasts.

is the one most frequently exacerbated by unsuitable water resource management plans and/or poor control of water extraction facilities. Unregulated but also illegal pumping draws a high and unreasoned amount of water which is uncountable in the water management policies and leads to groundwater depletion and reduce river, spring and wetland flows. The progressive lowering of the groundwater level reduces or removes the connections between the aquifer and the coastal GDEs. As a result, aquatic vegetation in these transitional wetlands is gradually being replaced by terrestrial vegetation. This process leads to the drying, reduction and disappearance of coastal GDEs. In the worst case, changes in the structure and the functioning of the ecosystem (Balasuriya, 2018; Pérez-Ruzafa et al., 2019) results in a partial or total loss of ecosystem services provided by coastal GDEs.

Anthropogenic activities require a growing demand for space for agricultural production, housing or industrial land use. The land gain can be achieved by the conversion of natural lands or by partially or totally draining wetlands (El-Asmar et al., 2013). The construction of artificial drainage network in order to control the humidity is an old and relatively common practice (Gerakis and Kalburtji, 1998; Avramidis et al., 2014). These practices are highly constraining for the hydrosystems. They drastically alter the natural flow of surface groundwater and greatly affect the coastal GDEs, which are relying on the surface expression of groundwater.

Changes in land use can have a significant impact on aquifer recharge processes and thus on fresh groundwater supplies to coastal GDEs. Infiltration is increasing with the proportion of bare soil and evapotranspiration's patterns are conditioned by the type and the stages of crops development. Soil compaction by urbanization or intensive agriculture may reduce the infiltration and enhance the surface runoff (van den Akker and Soane, 2005; Gregory et al., 2006; Nawaz et al., 2013). In addition, the urban pavement of the shore (El-Asmar et al., 2013) makes the soil impermeable and drastically reduces infiltration and recharge into the aquifer. 40% of the 46 000 km of Mediterranean coast were already artificialized in 2000 and it is expected to exceed 50% by 2025 (AViTeM, 2018).

If groundwater extraction is clearly the main threat in coastal Mediterranean regions, it is important to underline that increasing groundwater flow is also problematic. Some activities, such as irrigation, terracing, land-clearing or managed artificial recharge of aquifers, can appreciably increase the permeability of upper soils and then lead to the increase of the aquifer recharge (Baudron et al., 2014). In urban areas, tap water leaks can also constitute a significant source of groundwater recharge (Minnig et al., 2018; Vystavna et al., 2019). The flow of fresh water to the coastal GDEs can therefore be significantly increased. The physical and chemical disturbances can disturb and modify bio-community structure of the coastal GDEs.

3.2.2. The role of groundwater as a vector of pollution

Coastal GDEs often represent the last collector of water and their quality degradation results, and reflects human activities over the watershed. Anthropogenic activities such as the demographic, economic, industrial and commercial development often introduce new potential contamination sources (Appelo and Postma, 2005) which infiltrate towards the aquifer.

In the coastal Mediterranean regions, the main problem is related to the sewage inputs. The fast growing of urbanization is not always accompanied by the development of sewage infrastructures that results in less efficient treatment of urban wastewater and sewer leaks (Michael et al., 2013). In the Mediterranean basin, almost 40% of coastal settlements with more than 2000 inhabitants do not have any wastewater treatment plant (UNEP/MAP, 2012). This problem is especially exacerbated on the southern Mediterranean basin due to the rapid growth of many coastal cities and towns. In addition, coastal Mediterranean regions are privileged tourism destinations (UNEP/MAP, 2012). The touristic flow picks lead to higher rates of sewage inputs in urban sewerage networks that are often aged and failing. Wastewater and associated pollutants from domestic and industrial sources consequently infiltrate towards the aquifer or through the interaction between groundwater and river water (McCance et al., 2018; Erostate et al., 2019; Koelmans et al., 2019; Vystavna et al., 2019). Nitrogen pollutants, phosphorus, but also organic compounds and heavy metals are the most frequent contaminant affecting the groundwater resources (Wakida and Lerner, 2005; Petrie et al., 2015; Xu et al., 2019). The second main source of groundwater quality degradation is the agricultural activity. The excess of nutrients from fertilizers (nitrogen and phosphorus), pesticides, emerging compounds and, less frequently, pathogenic microorganisms related to agricultural activities contribute to the degradation of both ground and surface water quality (Symonds et al., 2018; Xin et al., 2019).

Once infiltrated, the pollutants follow the groundwater flow and can migrate to coastal GDEs (Rapaglia, 2005; Knee and Paytan, 2011; Jimenez-Martinez et al., 2016; David et al., 2019). According to the temporal dynamic of the aquifer, groundwater can represent a direct short and/or long term vector of pollution for coastal GDEs. Groundwater with short residence times (a few years) into the aquifer will rapidly flow towards the lagoons, carrying pollutants along its way. In case of groundwater with long residence time (several decades) and if no remediation process occurs, pollutants can be accumulated into the aquifer for several decades. The currently observed groundwater contamination can therefore be the result of the legacy of pollution related to human activities previously developed over the watershed (Erostate et al., 2018). This groundwater archiving capacity allows the storage of pollutants that will reach the coastal GDEs in the future.

Once the pollutants are in the coastal GDEs, prolonged groundwater residence times favor the accumulation of pollutants in water but also in aquatic organisms. The progressive accumulation of pollutants, especially heavy metals, along the food chain can pose serious human health issues and greatly impact economical profit by deteriorating ecosystems services such as aquaculture and fisheries. The most frequent impact of exceed in nutrients, sediments and organic maters is the eutrophication which can lead to important degradation or loss of seagrass beds, community structure and biodiversity (National Research Council, 2000; Pasqualini et al., 2017). More than 400 coastal areas have been identified worldwide as experiencing some form of eutrophication (Selman et al., 2008).

3.2.3. Impacts of climate change on aquifer recharge and implications for coastal GDEs

Important changes regarding the aquifer recharge in terms of timing, duration and magnitude (McCallum et al., 2010; Hiscock et al., 2012; Taylor et al., 2013) as well as the storage and the quality of groundwater are expected in a context of climate variability. These modifications will be more pronounced in arid regions and especially in the Mediterranean basin, considered as a Hot Spot of climate change (IPCC, 2014). By the middle to the end of the century, the southern European regions as well as Australia are expected to suffer from increasing arid conditions with longer and more frequent droughts (Stigter et al., 2014) due to the increase in the temperature (Ducci and Tranfaglia, 2008; McCallum et al., 2010), in evapotranspiration (Hiscock et al., 2012), modification of seasonal patterns of precipitation (Polemio and Casarano, 2008; Stigter et al., 2009; Barron et al., 2011) and of average effective infiltration (Ducci and Tranfaglia, 2008). An amplification in the frequency and intensity of drought is also expected in the southern Mediterranean basin, such as in Morocco (Stigter et al., 2014).

The results of predictive models to assess the impact of the climate change on aquifer recharge are often highly variable. The main tendency highlights a decrease in the groundwater recharge in Mediterranean regions, leading to a significant loss of groundwater resources (IPCC, 2007; Barron et al., 2011). In the Mediterranean basin, the decrease of the recharge can reach 30% to up to 80% (Ducci and Tranfaglia, 2008; Döll, 2009; Moseki, 2017). Modification in coastal aquifer recharge as well as the expected sea level rise (Hertig and Jacobeit, 2008; Somot et al., 2008; Mastrandrea and Luers, 2012) can lead to the inland migration of the mixing zone between fresh and saline water.

Climate change will exacerbate existing pressures rather than bring a new set of threats. With the water requirements that are projected to increase under a drier climate, severe water shortages can occur. The outflow into the coastal GDEs can be strongly reduced by the end of the century which could accelerate their drying up. Groundwater degradation by salinization could also greatly affect the physico-chemical conditions and thus the ecosystem balance of the GDEs lagoons. In response to these treats, a decrease in groundwater abstraction and an appropriate management appear as the principal way to ensure the preservation and sustainability of coastal GDEs (Candela et al., 2009; Stigter et al., 2014).

There may be exceptions to this general trend at the local level. In some cases, the modification of rainfall patterns and/or land uses modification can favor the recharge of the aquifer and improve the groundwater quality (Cartwright and Simmonds, 2008; Crosbie et al., 2010; Santoni et al., 2018). For example, in the Murray-Darling Basin in Australia, the clearing of the native vegetation is likely to favor the infiltration and increase the recharge of 5% for future climate around 2030 (Crosbie et al., 2010). If land-clearing could favor the recharge, the strong alteration of the hydrological cycle by vegetation cutting also has strong negative aspects which should be underlined. Among others things, land-clearing can increase runoff and streamflow, favor soil erosion, massive drainage of natural nutrients and salinization of soils and waters (Koivusalo et al., 2006; Cowie et al., 2007; Peña-Arancibia et al., 2012; Kaushal et al., 2018; Cheng and Yu, 2019). The consequences of these practices are often irreversible. Yet, for watersheds severely degraded by salinization, this increase in recharge could help the dilution and potentially improve quality of groundwater (Cartwright and Simmonds, 2008).

The existence of local specificities shows the importance of establishing adaptive case-by-case water management strategies. Water resource management requires the definition of appropriate management scale which makes it possible to manage the hydrosystem as a whole, taking into account the complexity of interactions between water bodies but also between humans and their environment.

4. Management strategies and current considerations for coastal GDEs

4.1. From international environmental awareness to Integrated Water Resource Management

The definition and establishment of water resources management strategies and policies result from an awareness of environmental issues initiated in the 1970s, with in particular the Stockholm Earth Summit in 1972 (Fig. 5a). This ecological awakening then continued in the 1980s with a collective awareness of the existence of pollution and harmful disruption on a global scale. It is in this context that the Bruntland Report define for the first time in 1987 the concept of "sustainable development": *"The sustainable development is development that meets the needs of the present without compromising the ability of future generations to meet their own needs".* This report requires the management of water resources as a common heritage and lays the foundations for integrated natural resource management. Only 5 years later, the Rio Earth Summit marked a turning point in the sustainable management of water resources with the "rediscovery" of the concept of Integrated Water Resource Management (IWRM) (Petit, 2006) and Integrated Coastal Zone Management (ICZM) (Deboudt, 2005).

These two concepts, which appeared in the 1970s (Deboudt, 2005; Petit, 2006), were then highlighted in the 1990s through the media coverage of the Rio Earth Summit and became a key concept in the 2000s thanks to the launch of the concept of sustainable development on the international political scene. In 2000, the Global Water Partnership, an international network created to advance governance and management of water resources, published its first's report on IWRM and clearly define the concept as a "process which promotes the coordinated development and management of water, land and related resources, in order to maximize the resultant economic and social welfare in an equitable manner without compromising the sustainability of vital ecosystems" (GWP, 2000). The IWRM was and remains widely promoted by many international organizations or donor agencies (Rahaman and Varis, 2005; Biswas, 2008), as a strategic approach to water management (Meublat and Le Lourd, 2001). The Johannesburg Earth Summit in 2002 even recommended its implementation in all countries by 2005. This summit also insists on the establishment of ICZM. Sharing the same precepts as IWRM, ICZM is nevertheless committed to taking into account the specific risks associated with water on the coast (Morel et al., 2004). ICZM is developing rapidly, particularly in Europe, thanks to its institutionalization and recommendation of the Council and the European Parliament in 2002 (Ghézali, 2009). Although coastal GDEs are in theory elements in their own right in integrated management strategies, they are still too often forgotten and do not benefit from legal or managerial recognition to take their specificity into account (Cizel, 2017).

4.2. Integrated groundwater management without specific regards for coastal GDEs

Since the 2000s, we have seen an acceleration of sustainable resource management measures at the global, regional and

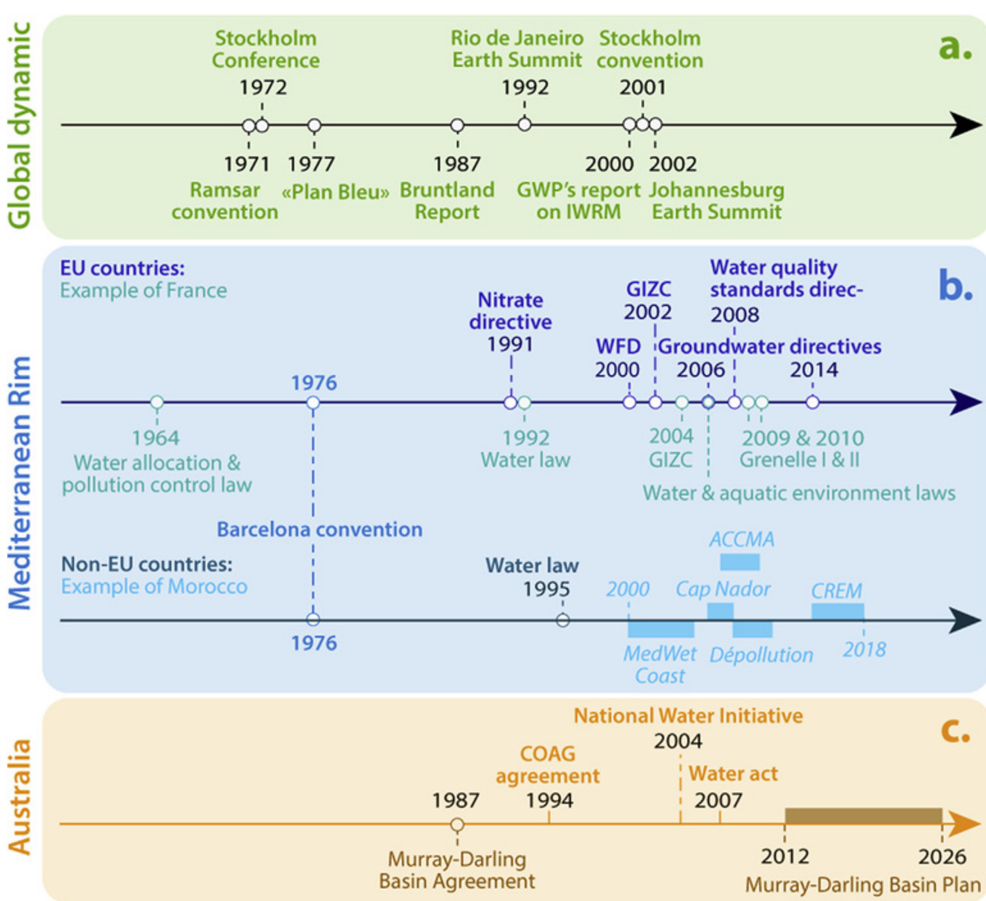

Fig. 5. Main world events that have guided the establishment of sustainable water resources management (a) and their translation into local laws and measures in the case of France (EU country), Morocco (Non-EU country) (b) and Australia (c).

national levels (Fig. 5a). GDEs have been partially propagated in water management policies developed over the past two decades, that recognize a link between groundwater and surface water. Some countries or group of countries particularly vulnerable to shortage of water and repeated severe droughts *e.g.* Australia, countries of the EU, the United-States (California) and South Africa, have yet incorporated specific reference to general GDEs into the legislation. Even if the protection of GDEs is included under water management policies, the implementation of an appropriate management policy is often lacking (Rohde et al., 2017).

Countries of the EU and Australia are the first to have included GDEs in their legislative framework (Rohde et al., 2017). The French model of water management by Water Agencies (created by the law of 1964) and the Australian model, derived from the experience of the Murray Darling Basin (Murray Darling Basin Authority created in 1987) are often considered as a reference model in terms of river basin management (GWP/RIOB, 2009; Brun and Lasserre, 2018). Legislative framework and groundwater managerial strategies set up by the EU and Australia however have shortcomings that undermine their effectiveness in protecting the resource (Fig. 6).

Australia provides the most comprehensive groundwater governance (Ross, 2016). As early as 1994, the agreement of the Council of Australian Governments (COAG) (Fig. 5c) required the development of a comprehensive system of water allocations and rights to ensure better, more sustainable water management. The water reform program initiated by the COAG agreements was then updated in 2004 by developing a new National Water Initiative (NWI) (Fig. 5c). The NWI - currently signed by all states and territories - has been recognized as the national blueprint for water sector reform to improve the state of industry and provide long-term environmental benefits (Willett, 2009). The annually adjustable water entitlements and related water market provide a great flexibility and a better adaptability to the state of the resource (Ross, 2016). However, monitoring of groundwater quality is limited (except for drinking water) and is often carried out on a short-term basis without consistent national program (Geoscience Australia, 2010). In Europe, on the other hand, both the quantitative and qualitative aspects benefit theoretically from an equivalent level of attention. The legislative framework implemented by the Water Framework Directive of 2000 (WFD) (Fig. 5b) provides thus the most comprehensive groundwater protection (European Commission, 2008; Ross, 2016). Member states are required to preserved the groundwater quantity and quality based on threshold values established to prevent any significant diminution of the ecological or chemical quality of surface water nor in any significant damage to terrestrial ecosystems which depend directly on the groundwater body (European Directive, 2000/60/CE). The degree of freedom given to the member states to define groundwater and GDEs management plans and the wide disparity between them can yet reduce the enforcement of EU recommendations (Liefferink et al., 2011). While some countries are considered as models for their efficiency in water management, such as France, Spain or Germany (Rahaman and Varis, 2005) (Fig. 5b and c), others are experiencing significant delays in the transposition of the EU recommendations (Ghiotti, 2011). In EU frameworks, an important point of divergence is the concept of “water bodies” that supports the WFD. This concept requires precise identification, delimitation and definition. However, the scientific knowledge is often incomplete or inaccurate and fails to provide the appropriate level of precision (Bartout, 2015). The lack of knowledge represents a significant bias for the definition of priority actions and the implementation of effective public policies

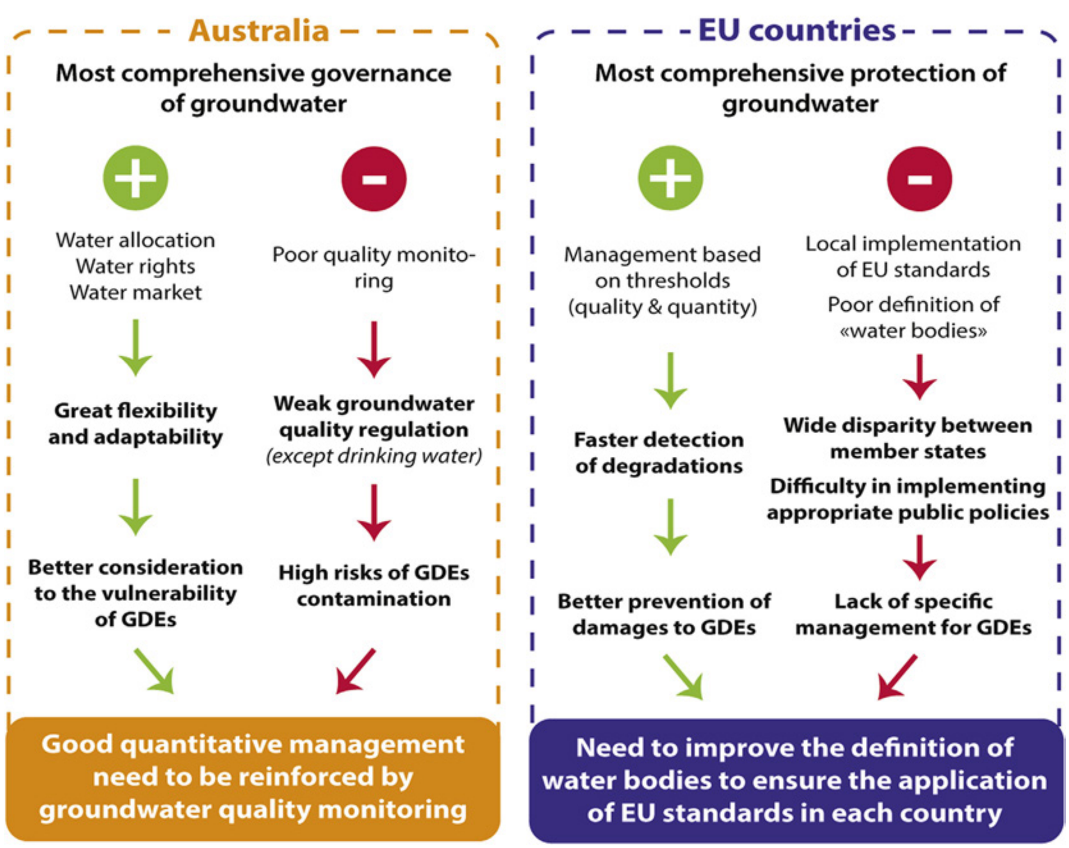

Fig. 6. Comparison of the strengths and weaknesses of management strategies in Australia and European Union.

to achieve the good qualitative and quantitative status set by the European recommendations (Maillet, 2015).

These two management models, one based on strong qualitative regulation of the resource (Australia) and the other on the monitoring of threshold values (EU), lead to significant disparities in GDEs management. In Australia, management decisions are based on an ongoing monitoring and research which help to establish an adaptive GDEs management (Richardson et al., 2011; Rohde et al., 2017). The great adaptability of annual water allocation allows a better consideration to the vulnerability of GDEs, particularly in a case of severe drought. However, the poor water quality monitoring exposes lagoons to high risks of undetected contamination. Efforts made for the qualitative management of the water resource clearly need to be completed and reinforced by an improvement of groundwater quality management to ensure the preservation of GDEs (Ross, 2016). In EU, monitoring threshold values allows a better understanding and thus, a better prevention of qualitative and quantitative degradation risks for GDEs. The groundwater allocation is often included in river basin plans of member states but the adaptability of water withdrawals, particularly in the event of drought, can lack reactivity and damage the GDEs (Sommer et al., 2013; Stein et al., 2016). To really benefit from the European directives, particular attention must be paid to their concrete application in all member countries. In addition, the concept of "water bodies" must be better defined in order to enable the implementation of truly effective public policies.

In the particular case of coastal lagoons, considered by the WFD as "transitional water bodies", the lack of knowledge and data in the early 2000s has triggered the development of monitoring networks implementation. Indeed, the monitoring programs developed for freshwater ecosystems are not relevant for coastal GDEs. These transition environments are subject to many influences that induce a large variation in physical parameters, including salinity. The consideration of biological indicators and the evaluation of shifts in the species presence on coastal ecosystems has emerged as a valid strategy to characterize ecological status (Delpech et al., 2010; Pérez-Domínguez et al., 2012). This approach, followed in the same way by several EU countries, has led to the creation of indicators validated by the EU to improve the assessment of the status of transitional water bodies in the North-East Atlantic (Le Pape et al., 2015). For the Mediterranean region, this work has yet to be completed. Currently, only Greece, Italy and France have developed classification tools, but further developments are still needed to properly assess the ecological status of coastal lagoons (Le Pape et al., 2015).

Even if the groundwater resource management plans help to manage GDEs, specifics on GDEs management are often lacking (Rohde et al., 2017). Coastal GDEs form part of a continuum between continental and marine ecosystems and share common characteristics, species and ecological functions (Pérez-Ruzafa et al., 2010). Inland and coastal waters must be managed as a whole and coordination at river basin and coastal sea levels is required (Pérez-Ruzafa and Marcos, 2008). The IWRM is generally focused on the inland watersheds but likely neglects coastal specificity. Conversely, ICZM focuses exceptionally on coastal areas. However, the coastal area rarely extends to the entire watershed, which influences the quality and quantity of water resources that reach the coast. The link between IWRM and ICZM appears essential to respect the physical, ecological and social continuum of watersheds and their coastal zones.

4.3. Limitations of the project-based approach

The IWRM does not automatically lead to the sustainability of resource uses, although it is a prerequisite (Aubin, 2007). The project-based approach, often applied in environmental protection, makes it difficult to develop a coherent policy. Encouraged by cooperation projects, several countries have tried to initiate the IWRM (Garnaud and Rochette, 2012). This is particularly the case in non-EU countries, such as Morocco and Algeria (Vecchio and Barone, 2018). The coastal GDEs of Nador (Morocco) (Fig. 5b) constitutes a representative example (Garnaud and Rochette, 2012).

Since the 1970s, coastal development has been announced as a priority by the Moroccan government, but there is no national public policy for coastal areas. The growing development exerts a strong pressure on the coastal GDEs, classified as RAMSAR site (Nakhli, 2010). The Nador lagoon is thus the subject of a succession of projects (Fig. 5b) whose objective is to establish a sustainable management of this area (Garnaud and Rochette, 2012). To be "sustainable", resource management must yet be both based on previous actions and forward-looking. Most often, projects follow one another, without taking into account previous results. The standardized procedures proposed by donors do not sufficiently take into account the specificities of the territories. The multiplicity of projects is often counterproductive and compromises the effectiveness of this environmental development assistance. The succession of projects without convincing results ends up reducing the mobilization of local actors and users. This generally too short-term approach limits the involvement and appropriation of target actors. This problem of appropriation is in addition to the problem of the limited funding period, which threatens the sustainability of the actions undertaken (Garnaud and Rochette, 2012). By the end, Morocco's commitment to Integrated Coastal Zone Management (advocated by the - too short - Cap Nador project, from 2006 to 2008) finally found little support in these international collaborations (Garnaud and Rochette, 2012).

5. Better global understanding for a better management of GDEs

Due to their complexity, the development of management strategies adapted to coastal GDEs is particularly complex because it requires a strong transdisciplinary approach. Scientists in the technical sciences (at least hydrology, ecology, hydrogeology, oceanography) need to develop collaborative approach between them but also with social and legal scientists. Although difficult and slow to implement, this transdisciplinary approach has two major advantages. Firstly, it allows scientists to question their own discipline, in particular by putting into perspective the relevance of their own concepts and methods. Then, the development and construction of common methods and concepts results from a shared reflection. These new concepts are thus more relevant because they come from a collaboration work and not from the interweaving of specificities borrowed from each discipline.

5.1. Improving the understanding of GDEs

The improvement of GDEs' management inevitably involves an increasing knowledge of their hydrogeological and ecological condition and processes (IAH, 2016). This information is the most often unavailable and gaps at the intersection of groundwater hydrology and ecology do not facilitate the study of GDEs (Tomlinson, 2011). These gaps are even more important in the case of coastal GDEs which require collaboration between terrestrial hydrology and marine sciences - two epistemic communities that are not necessarily, or very rarely, used to working together. In addition, the implementation of the necessary monitoring systems to improve the understanding of GDEs is often financially and technically expensive and/or difficult to implement (Bowmer, 2003; Roll and Halden, 2016). Improving the management of coastal GDEs inevitably requires the management and understanding of hydraulic processes throughout the water cycle (fresh and salt water).

To overcome the lack of knowledge about GDEs, EU countries and Australian Government and the scientific community have been working together to establish practical guides. These “GDE practical guides” can in theory assist state agencies in the identification and management of GDEs for water management plans (Clifton, 2007; Richardson et al., 2011; Hinsby et al., 2015). They offer a range of methods for determining ecosystem reliance to groundwater and help water managers conducting the necessary technical investigations and monitoring protocols to define ecological water requirements for GDEs. In practice, these often complex guides seek data keys to understand all types of systems but each GDE is an individual case, having specific characteristics and behavior that prohibit any generalization of diagnoses and solutions. The identification of appropriate study tools requires significant scientific support and the evaluation and monitoring of the relevance of the tools used is yet another debate.

Generally, the improving of knowledge depends on the strategic and economic interest of GDEs, assessed by the costs and benefits related to their protection (Millennium Ecosystem Assessment, 2005). The “ecosystem services approach” of the United Nations Millennium Ecosystem Assessment Project thus recommend to complete the technical approach of GDEs by a relevant assessment of the GDEs’ valuation and relationship between ecosystems and human well-being. While the evaluation of ecosystem services tends to highlight man’s dependence on his environment, this economist approach to nature raises two concerns. Firstly, this new way of thinking about nature conservation places nature at the service of mankind (Dufour et al., 2016). GDEs are then considered as providers of valuable goods and services. The diversity and complexity of the relationship between humans and nature cannot be summarized as a monetary evaluation exercise (Sartre et al., 2014). Moreover, human societies had already understood the importance of coastal GDEs and how to benefit from them well before the concept of “ecosystem services” was adopted. Secondly, the economic assessment of GDEs requires a clear definition of the benefits of these ecosystem services including direct (fish and plant production, water storage and purification …) and indirect values (cultural, aesthetic, social reasons …) to the human population (IAH, 2016). Estimating the economic values of ecosystem services is far from easy. Recreation and tourism are the most easily quantifiable services, firstly because the direct revenue they generate are easily quantifiable but also because they receive special attention due to the attractiveness of coastal GDEs (Rolfe and Dyack, 2011; Clara et al., 2018). On the other hand, essential services such as protection against erosion, climate regulation or pollution control are neglected, largely underestimated and/or under-studied due to the lack of available data (Barbier et al., 2011).

5.2. Determining the appropriate management scale

The watershed is considered as the most environmentally and politically relevant management unit. This watershed-based approach can contribute to reinforce the lack of consideration given to “hidden” groundwater resources, while they are essential to establish an integrated management of GDEs. An appropriate management scale is a necessary first-step for the sustainable management of supporting aquifers and of the coastal GDEs (Vieillard-Coffre, 2001; Bertrand et al., 2014).

Firstly, surface and ground water are not constrained by the same geological boundaries. The hydrogeological and hydrological watershed do not necessarily (or rarely) overlap (Affeltranger and Lasserre, 2003). The extension of an aquifer and the drained groundwater can extend well out of the boundaries defined by the hydrological basin. Human activities developed outside the hydrological basin can impact qualitatively and/or quantitatively the groundwater resources flowing within the basin and/or hydraulically connected. A significant water supply-demand gap can therefore be induced. A broader consideration of a “water-supply area” would allow a better assessment of the water resources actually available. This approach would ensure a better allocation of water between human and ecosystem needs.

Surface and groundwater have very different flow dynamics. Groundwater flow takes on average several years, even centuries, compared to a few days or a few weeks for river water (Fetter, 2018). The capacity of recharge and renew is much longer. Their inertial behavior supports their capacity to accumulate the pollutants and to record the degradation caused by human activities over several decades (section 3.2.2.). The positive or negative effects of the land use planning made over the hydrological basin can take several decades or even centuries before being noticeable on groundwater quality and quantity (Boulton, 2005). The notion of sustainability preached by IWRM can then be strongly questioned if the groundwater dynamics are not enough understood and/or not considered by management strategies.

The existing hydraulic exchanges between the different water bodies and the vertical linkages are not always fully appreciated (Boulton, 2000). Part of the problem relates to the difficulties of assessing groundwater volumes, recharge rates and sources but also to the low recognition of the linkages between groundwater and many surface water ecosystems (Boulton, 2005). The qualitative and quantitative status of a water body has an impact - positive or negative - on all the water bodies connected. It is then important to understand the existing relationships between the aquifer and all the other water bodies, which means neighboring aquifers, fresh surface water and brackish surface water.

More and more water resources managers are becoming familiar with the necessity of considering large spatial areas to establish a relevant water management (Boulton, 2005). Even if their perceptions of hydrologic interactions are often restricted to lateral and longitudinal flows (Pringle, 2003), the importance of vertical connectivity is slowly being appreciated (Boulton, 2000). A greater consideration of the ecological processes that support the proper functioning of the GDEs is being given. The study of the “proper functioning areas” of GDEs would define the extension of the surrounding area that supports the ecological processes that ensure the sustainability and resilience of the wetland (Chambaud and Simonnot, 2018). It would take into account all the factors that contribute to the functioning of the GDE, *i.e.* water qualitative and quantitative supply, but also animal species for which all or part of the life cycle occurs near the GDE and the connectivity of the GDE with other biodiversity reservoirs, animal and plant populations.

5.3. Partnership, appropriation and relevant definition of coastal GDEs

The efforts required to establish effective multi-scale governance are not often sufficient to ensure the sustainable management of groundwater and GDEs (Molle et al., 2007) (Fig. 7). Several shortcomings already mentioned above, partially explain these difficulties (Fig. 7). The development of regional guidelines based on too approximate or minimalist knowledge of GDEs, inevitably leads to inconsistencies in management strategies at the local level. Coastal GDEs often suffer from incomplete, inappropriate or even contradictory definitions. Scientific definitions are sometimes in conflict with legal definitions and make the recognition and conservation of these environments more complex (Cizel and Groupe d’histoire des zones humides 2010; Cizel, 2017). Coastal GDEs are often recognized and grouped into the large family of wetlands. A

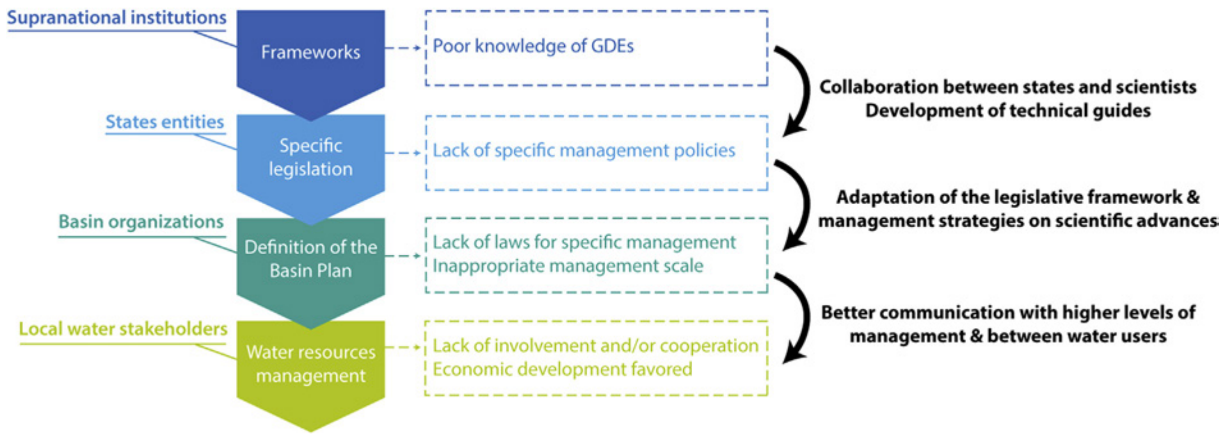

Fig. 7. Conceptual diagram showing the institutions and their roles in water resources management, highlighting major gaps and the points to be improved between two hierarchical levels.

simplification that does not take into account their specificity, consisting of a wetland, a water body and an aquifer, all hydraulically connected, which must be recognized and managed as an inseparable whole. Improving the definition of coastal GDEs is essential both to better understand and to delimit them, but also to develop and to apply specific and appropriate protective legislative acts.

While the advancement of scientific knowledge and its better consideration at the regional level could be a way to improve the management of GDEs, a large part of the solution also seems to come from the local level. At the local scale, collaboration between water stakeholders for integrated resource management can be complicated (Mostert, 2003; Chanya et al., 2014). The initial appropriation by state entities (Water Agencies or Basin Organizations) of the recommendations formulated by regional and national institutions often appears insufficient for the local implementation of adapted and sustainable management strategies (Fig. 7). A real appropriation of existing regulations on coastal GDEs by all local stakeholders, decision-makers and actors in the territory appears essential for the preparation of relevant planning or development documents and the implementation of appropriate action programs. The elements required to define the challenges and perspectives related to GDEs must not be a local adaptation of regional recommendations but rather a collective elaboration by all the actors concerned. Efforts must be made to develop a framework for effective public participation at six levels: information, education, consultation, involvement, collaboration and capacity building (Das et al., 2019).

Coastal aquifers are particularly vulnerable to water users conflicts (Zepeda Quintana et al., 2018). All water users want to be able to benefit from the quality and quantity of water resources they need. No user can be abandoned in favor of another, nor can the need for environmental waters. Environmental water needs cannot be forgotten and must be taken into account in management strategies. Sustainable water management thus requires water demand management, which must be achieved through agreements and collaboration at an appropriate scale. The establishment of a strong collaborative processes appears as the only way to guarantee the essential groundwater supply to coastal GDEs and their sustainability (Boulton, 2005). The management of coastal GDEs must take into account its hydrological basin as well as its territorial water management unit and all territorial units important for its management, *i.e.* tourist unit, geographical unit, air of influence of neighboring cities or migratory bird management (Mermet and Treyer, 2001) …

6. Conclusion

Nowadays, coastal Mediterranean regions suffer from an overdevelopment of anthropogenic activities which strongly impact the groundwater resources and depending coastal GDEs. Although some Mediterranean regions have included the protection of GDEs in their water management policies, the implementation of an appropriate intergraded and collaborative management is often lacking and coastal GDEs do not benefit from a particular status due to their complexity.

The preservation of coastal GDEs is subject to the stability over time of fresh water supplies (ground and surface water) in sufficient quantity and quality. However, the determination of the qualitative and quantitative needs of coastal GDEs is difficult to evaluate and each coastal GDE is a unique case. Particular attention should therefore be paid to the characterization of environmental and ecological water requirements. The hydrogeological knowledge about the management and behavior of coastal aquifers and GDEs must be strengthened. Hydrogeology must be considered as an integral component of the coastal GDEs and not a sub-discipline of hydrology, as is too often the case at present. The inventory and characterization of coastal GDEs must be improved through in-depth systemic approaches. To this end, the coupling of hydrogeochemical and geophysical techniques, which are inexpensive, seem to constitute a relevant strategy. These investigations must be supplemented by the identification and evolution of the sources of contamination present in the catchment areas. In order to better understand the role of groundwater as a vector of pollution, particular attention should be paid to the identification of the main groundwater discharge areas and the assessment of contaminant flows and loads. The systematic mapping of groundwater vulnerability in the coastal areas must be promoted, using methods accounting for both the intrinsic and specific vulnerability of groundwater. This kind of data must help to develop land-uses and human activities according to the groundwater vulnerability. Finally, in the case of effective degradation processes, restoration plans should be considered. A reflection must be carried out for the definition of relevant indicators of the ecological coastal GDEs status. For these environments subject to high variabilities, particularly in terms of salinity, there is a necessity of developing sensitive indicators for monitoring ecological status. Biological indicators seem to be helpful but needs to be further and widely developed.

From a qualitative point of view, the estimation of groundwater withdrawals is often very approximate because of the poor knowledge of the extraction points. It seems essential to carry out an exhaustive inventory of wells and boreholes in the coastal GDE watershed. The implementation of retroactive measures for reporting private wells would also allow a better knowledge of the existing structures, which are currently not recorded. Regularly monitored water quotas for private individuals could also be helpful for the qualitative management of the resource.

At present, the lack of an appropriate definition for coastal GDEs is a huge problem. Lack of discussion and consensus between lawyers and scientists does not facilitate the establishment of management strategies. To be efficient, this definition needs to be the result of a joint reflection between several disciplines. As showed in this synthesis, the transdisciplinary approach between hydrogeology, hydrology, social sciences and law is essential to fully understand the socio-economic and environmental complexity of coastal GDEs. The inventory of coastal GDEs characteristics could help to establish a complete and relevant definition of coastal GDEs. In addition to involve several discipline, thoughts about coastal GDEs definition need to be based on the mobilization of scientist, lawyers but also water users and stakeholders. Information, appropriation and collaboration are clearly strategic, interdependent points to be developed. Local water users and managers must feel concerned by the problems related to coastal GDEs to build appropriate and sustainable management plans. Without this process, all possible efforts can be taken, but their chances of achieving successful results will remain low. The creation of permanent mechanisms such as water user groups or groundwater forums could be useful. These moments of exchange and discussion would also allow managers and decision-makers to better understand the role and benefits of coastal GDEs. Indeed, evaluation of the ecosystem services is essential for valuing the coastal GDEs and decision makers at many levels are unaware of the connection between wetland condition and the provision of wetland services and consequent benefits for people.

All water resources in the coastal areas should be managed collectively and strategically, in order to maximize use efficiency, reduce water use conflicts and avoid over-exploitation. In other words, the management strategy must consider the lagoon water body, the surrounding wetland and groundwater as an inseparable set of communicating vessels whose nature of exchanges is subject to temporal and spatial variations. In the global context of unprecedented anthropogenic pressures, hydro-food crises and climate change, the consideration given to coastal GDEs represents a key issue for the socio-economic and environmental sustainable development of many coastal Mediterranean areas. Integrated water management strategies that consider environmental needs on an equal footing with socio-economic constraints within the coastal hydrosystem need to be improved. The ICZM is the management strategy that most considers water resources in the coastal zone and refers to coastal aquifers as such and specifies a monitoring requirement. However, despite the growing consideration for coastal aquifers, there are still gaps. It is important to continue to raise public awareness of coastal aquifers at the regional level and to integrate their specificities into coastal zone management strategies and plans. Collaboration between states or countries, sharing of knowledge and technology facilitated by the creation of exchange material could also contribute to improving the integration of coastal aquifers into local guidelines and policies.

These practical suggestions could help for improving the management of coastal aquifers and coastal GDEs. In this way, groundwater and coastal water GDEs could really benefit from the optimal environmental conditions required to ensure their sustainability.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Chapitre 2 :

L'hydrosystème de la lagune de Biguglia

Ce chapitre présente le site d'étude de l'hydrosystème de la lagune de Biguglia en abordant notamment le contexte géologique, hydrologique et hydrogéologique ainsi que les pressions anthropiques affectant le bassin versant.

2. L'hydrosystème de la lagune de Biguglia

2.1. Localisation et géomorphologie

Le bassin versant de la lagune de Biguglia est situé au Nord-Ouest de la Corse, au Sud de la ville de Bastia (Figure 5). Il s'étend sur près de 182 km² et présente des reliefs fortement contrastés. Au Nord et à l'Ouest, le bassin versant est délimité par les contreforts schisteux de la Corse alpine, dont l'altitude culmine à 1450 m. Au Sud-Sud-Est du bassin versant se développe la plaine alluviale de la Marana. Le contact avec mer Tyrrhénienne constitue la côte littorale du système. Enfin, le Golu, plus long fleuve de Corse, constitue la limite Sud du bassin versant (Figure 6).

Figure 5: Vue aérienne© du bassin versant de la lagune de Biguglia. Le Nord est localisé à droite de la photo.

D'une superficie de 14,5 km² (11 km de longueur et 2,5 km dans sa plus grande largeur), la lagune de Biguglia est la plus vaste de Corse. Orientée parallèlement à la côte, cette lagune peu profonde (1,2 m en moyenne, avec un maximum de 1,8 m) est isolée de la mer Tyrrhénienne par un cordon littoral (lido de la Marana) d'une largeur inférieure à 1 km. Seul un étroit chenal d'une quinzaine de mètres de largeur, localisé dans sa partie Nord, connecte la lagune à la mer Tyrrhénienne. Les échanges avec le milieu marin sont cependant restreints. Ce grau étroit est souvent comblé par l'accumulation naturelle de sédiments sableux portés par la dérive littorale et se rouvre au gré des tempêtes ou par l'intervention humaine. La lagune constitue par conséquent un système confiné favorisant l'accumulation des éléments et nutriments en provenance du bassin versant, responsables des phénomènes ponctuels d'eutrophisation (Département de la Haute-Corse, 2012 ; Garrido et al, 2016 ; Pasqualini et al, 2017).

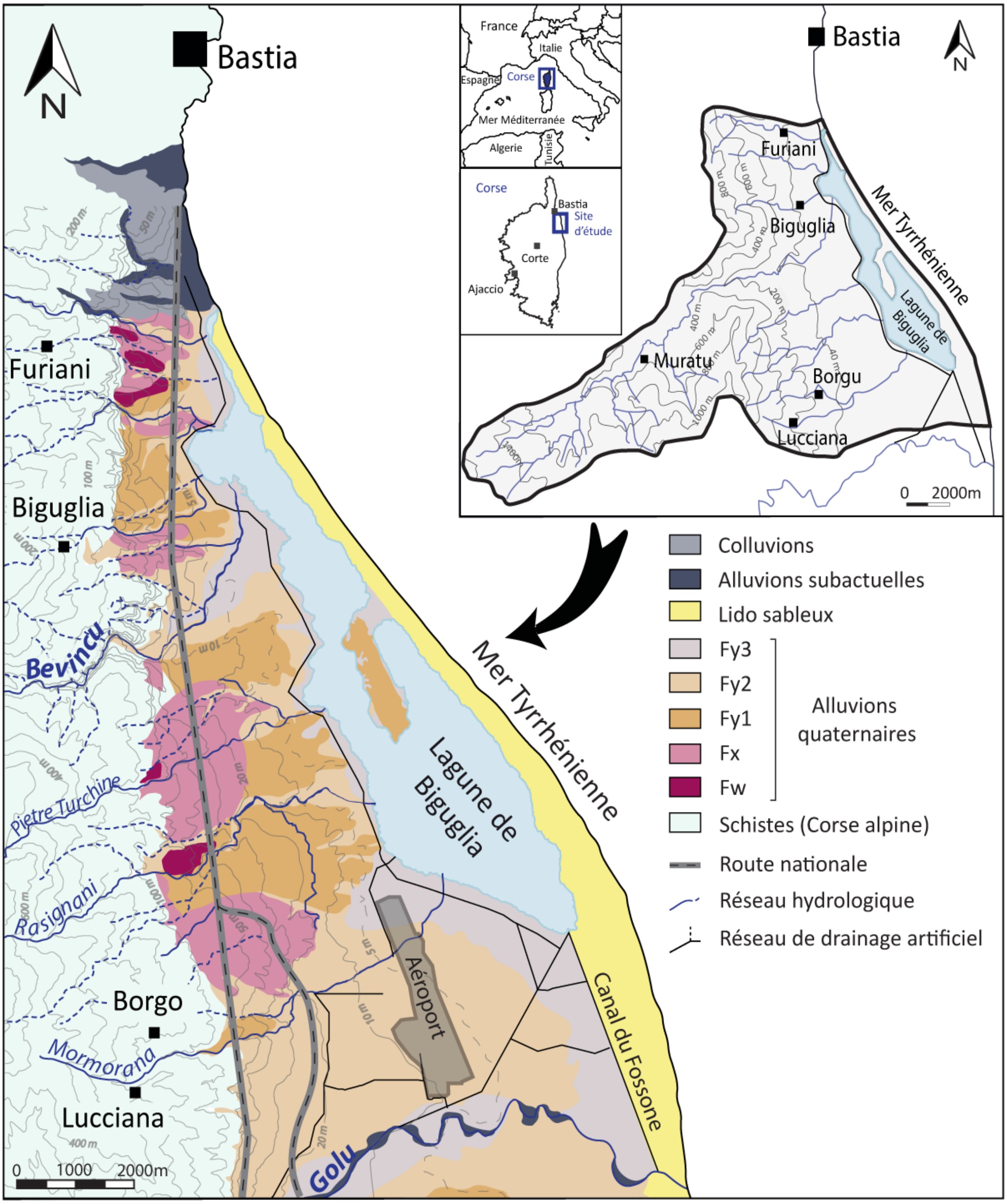

Figure 6: L'hydrosystème de la lagune de Biguglia, modifié d'après Erostate et al. (2018).

Propriété de la Collectivité de Corse depuis 1988, la lagune de Biguglia est classée Réserve Naturelle depuis 1994. Il s'agit d'une Zone d'Intérêt Communautaire pour les Oiseaux (ZICO), Zone de Protection Spéciale (ZPS), Zone Spéciale de Conservation (ZSC) du réseau Natura 2000 (Département de la Haute-Corse, 2013). En raison de son rôle majeur pour les oiseaux migrateurs, cet espace naturel est classé « zone humide d'importance internationale » depuis 1991, conformément aux critères de la convention RAMSAR. Pour limiter les risques de dégradation du milieu, les acteurs locaux et les élus ont initié dès 1994 l'instauration du SAGE de l'Étang de Biguglia (SAGE, 2012). L'élaboration, la révision et le suivi du SAGE sont assurés par la Commission Locale de l'Eau (CLE), créée en 1996. Après un état des lieux plus ou moins exhaustif de l'état actuel et du fonctionnement du bassin versant (Département de la Haute-Corse, 2003), un ensemble de mesures ont été définies pour favoriser l'atteinte du bon état des milieux requis par la DCE (SAGE, 2012). Le SAGE s'attarde notamment sur i) l'importance de la lutte contre les pollutions, surtout diffuses, ii) la gestion durable des ressources en eau et la préservation des équilibres quantitatifs, iii) la préservation ou la restauration de la continuité écologique des différents milieux aquatiques et iv) la sensibilisation des usagers du bassin versant à la qualité patrimoniale, la richesse écologique et la fragilité des milieux naturels et humides associés à l'Étang de Biguglia.

2.2. Contexte géologique

2.2.1. Formations métamorphiques secondaires

L'essentiel de la Corse schisteuse, ou Corse alpine, est formée par les schistes lustrés ophiolitifères et les calcschistes d'âge Jurassique ou Crétacé. Ces schistes, largement allochtones, ont subi plusieurs processus métamorphiques. La zone des schistes lustrés comprend trois ensembles : les schistes lustrés inférieurs et supérieurs, entre lesquels s'intercale un niveau ophiolitique. Ces roches basiques et ultrabasiques se composent de roches ultramafiques (serpentinites et péridotites), de gabbros et de laves basiques. Le bassin du Bevincu est presque entièrement creusé dans le cortège ophiolitique des schistes lustrés (Lahondère, 1981; Lahondère and Lahondère, 1988).

2.2.2. Formations sédimentaires quaternaires

La plaine alluviale de la Marana se compose d'un ensemble de paléo-terrasses quaternaires imbriquées et orientées d'Est en Ouest (Figure 6). Les terrasses alluviales se sont formées au cours des dernières phases glaciaires. Le Wurmien supérieur (stade glaciaire récent) correspond en aval aux alluvions fluviatiles Fy3 des très basses terrasses et les alluvions précédentes correspondraient respectivement au Wurmien moyen (Fy2) et inférieur (Fy1) et aux glaciations antérieures (Fx, Fw). Les altérations de ces alluvions et leur érosion par le Golu et le Bevincu se sont produites pendant les phases interglaciaires (Conchon, 1972).

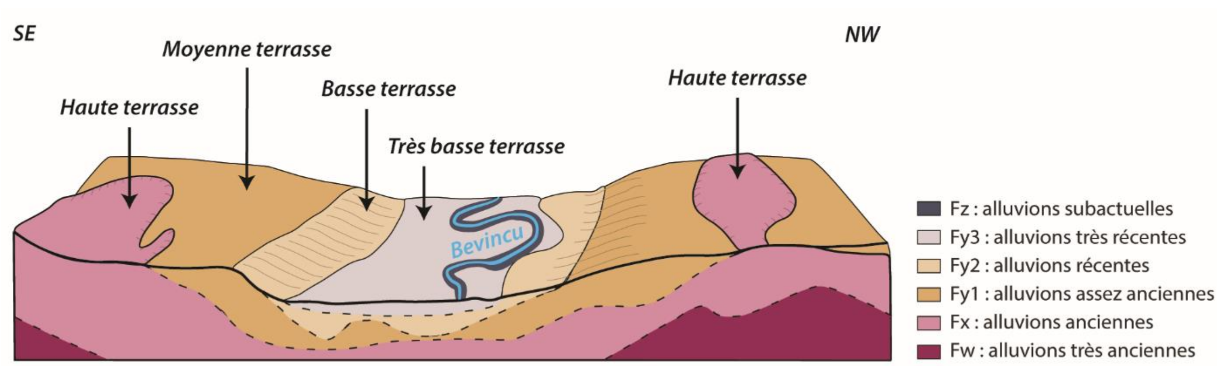

Figure 7: Coupe schématique illustrant l'emboîtement des terrasses alluviales de la Marana au niveau du Bevincu, modifiée d'après Orofino (2010).

Ces dépôts quaternaires (100 à 150 m) sont principalement attribués aux cônes de déjections du Golu et du Bevincu et des torrents temporaires issus de la zone des schistes lustrés. La sédimentation hétérogène se caractérise par une alternance de zones argileuses, conglomératiques ou sableuses, caractéristique de la sédimentation des cônes torrentiels et des dépôts fluviatiles de crues. Quel que soit leur âge, toutes les terrasses sont constituées de graviers, galets ou blocs sans stratification, emballés dans une matrice plus ou moins abondante sablo-limoneuse, parfois argileuse, donnant lieu à des lentilles dépourvues de galets (Conchon, 1972, 1978). Pour l'ensemble des alluvions, les galets sont hétérométriques en amont, mieux triés à l'aval. La granulométrie des galets ne semble pas varier entre la vallée du Bevincu et celle du Golu. La description de l'altération des galets et de la matrice les englobant permet la reconnaissance des dépôts et leur classification en six groupes (Conchon, 1972) :

- **Fw et Fx, les alluvions anciennes** à paléosol rouge-orange, formant les hautes terrasses. Ces alluvions sont principalement composées de galets granitiques et schisteux altérés, entourés d'une matrice fine et imperméable, principalement sablo-argileuse. D'importants bancs et lentilles conglomératiques sont présents au sein de cette série ;
- **Fy1, les alluvions assez anciennes** à paléosol rouge-orange, formant les moyennes terrasses. Ces alluvions présentent une forte proportion de galets de gabbro et de galets de schiste altérés, non-friables, de couleur rouille. La matrice argilo-limoneuse confère une nature imperméable à cet ensemble ;
- **Fy2, les alluvions récentes** à sol brun formant les basses terrasses. Au sein de ces alluvions, les galets sont peu altérés et compris dans une matrice brune sablo-argileuse. Les alluvions Fy2 sont très développées dans le secteur du Golu. Elles sont notamment exploitées pour l'extraction de granulats ;
- **Fy3, les alluvions très récentes** à sol gris formant les très basses terrasses. Dans ce niveau, les galets ne sont pas altérés et la matrice le plus souvent sableuse se caractérise par une teinte grisâtre. En amont, les alluvions Fy3 sont principalement formées de galets et blocs résultants de la sédimentation des cônes torrentiels. En aval, leur nature se distingue nettement entre la vallée du Bevincu et du Golu. Dans le premier cas, on distingue des limons d'inondation gris, bien classés, pouvant atteindre jusqu'à 15 m de puissance. Dans le secteur du Golu, les alluvions sont principalement des sables grossiers, bien triés, pauvres en argiles et en silts ;
- **Fz, les alluvions subactuelles** à sol gris formant le lit majeur des cours d'eau. Il s'agit principalement de sable et limons gris.

2.3. Contexte climatique

La Corse se caractérise par un climat méditerranéen classique avec d'importantes variations saisonnières et interannuelles des températures et des précipitations. Les étés chauds et secs (de 22 à 25 °C en moyenne en juillet et août) sont souvent accompagnés de périodes de sécheresse persistantes. Les hivers sont relativement doux (de 7 à 9 °C en moyenne en janvier) et pluvieux (Rome and Giorgetti, 2007). Les précipitations tombent souvent sous forme d'averses réparties sur 50 à 80 jours seulement, avec deux apports principaux, l'un en fin d'automne (octobre à décembre), l'autre en début d'hiver et de printemps (janvier à mars). Les conditions climatiques sont variées sur l'île, en raison notamment de l'important contraste altitudinal entre les hauts reliefs alpins du centre de l'île et les plaines littorales. Les précipitations sont presque deux fois moins importantes en plaine (de 600 à 770 mm en moyenne) qu'en montagne (1400 mm en moyenne) (Rome and Giorgetti, 2007) et des variations significatives peuvent se faire sentir à l'échelle kilométrique (Bruno et al., 2001). Enfin, les vents locaux (mistral, tramontane, libeccio) sont fréquemment violents, atteignant des forces moyennes de 60 à 80 km/h (Rome and Giorgetti, 2007).

A Bastia, la hauteur moyenne annuelle de précipitation est de 774 mm (Figure 7), avec des cumuls particulièrement forts d'octobre à décembre (supérieur à 85 mm) (Figure 8). La température moyenne annuelle mesurée à la station Météo France de Bastia est de 15,8 °C. La variabilité saisonnière moyenne des pluies et des températures enregistrées montre un régime caractéristique du domaine méditerranéen. Les températures les plus faibles sont naturellement observées de décembre à février et les plus chaudes durant les mois d'été, juillet et août (Figure 8).

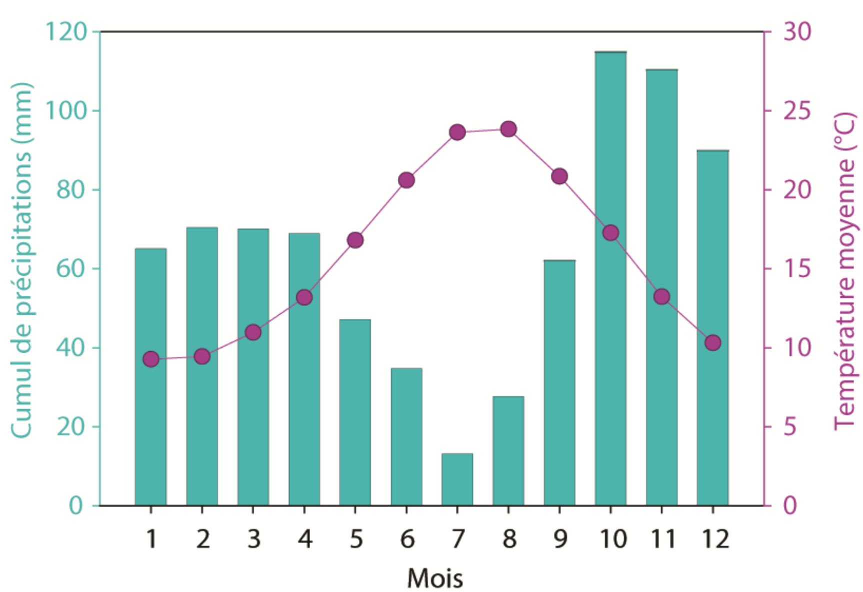

Figure 8 : Diagramme ombrothermique de Bastia construit à partir des normales mesurées à la station Météo France de Bastia (1950-2016)

Les cumuls annuels des précipitations et des températures moyennes montrent une variabilité interannuelle importante au cours du dernier siècle (Figure 8). On constate également une évolution à la hausse des cumuls de précipitations mensuels et des températures moyennes mesurées. Ce phénomène semble particulièrement s'accentuer à partir des années 1980.

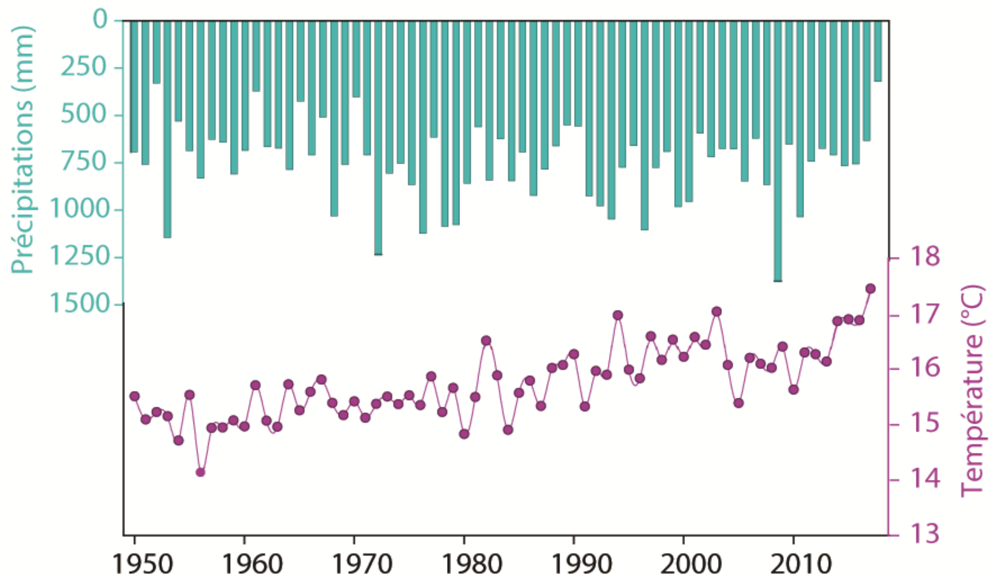

Figure 9: Chronique des cumuls de précipitations à partir des données journalières cumulées annuellement et chronique des températures à partir des données journalières moyennées annuellement, mesurées à la station Météo France de Bastia (1950-2016)

Les précipitations représenteraient un apport en eau douce vers la lagune compris entre 12 Mm³/an (BCEOM 2006) et 14,3 Mm³/an (Frisoni et al., 1992; Mouillot et al., 2000). La quantification de la recharge à l'aquifère reste cependant encore très peu contrainte. Un important travail d'estimation de la recharge a été effectué au cours de ce doctorat. Les résultats sont présentés en détail au chapitre 4.

2.4. Contexte hydrologique

Le bassin versant alimentant la lagune de Biguglia est drainé majoritairement au Nord par le Bevincu. Ce fleuve s'écoule de façon permanente sur près de 24 km (débit moyen de 0,61 m³/s), depuis sa source dans le Monte Reghia di Pozzo, à 1469 m d'altitude. Il contribue directement à l'apport d'eau douce vers la lagune, avec un volume estimé à environ 20 Mm^3/an (Frisoni et al., 1992).

Quatre autres cours d'eau temporaires sont également présents, le Pietre Turchine, le Rasignani, la Mormorana et le San Pancraziu (Figure 4). Leur écoulement est temporaire, en lien direct avec les épisodes pluvieux. Les débits de ces cours d'eau ne sont pas référencés mais l'apport total du bassin versant (incluant le Bevincu) est estimé entre 28 et 58 Mm^3/an (Frisoni et al., 1992; Mouillot 2000).

Le Golu, principal fleuve de Corse, marque la limite Sud du bassin versant. Il ne fait pas partie intégrante du bassin versant mais il est relié à la partie Sud de la lagune via le canal du Fossone (Figure 5). Le Golu prend sa source à 2525 m d'altitude, dans les massifs Hercyniens du centre de l'île. Son débit moyen s'élève à 14,8 m^3/s.

2.5. Contexte hydrogéologique

La plaine alluviale constitue le principal système aquifère de la région en terme de taille. Cet aquifère se développe principalement dans les dépôts alluviaux récents du lit majeur des fleuves (Fy2 et Fy3, Figure 6). La couche d'argile imperméable des alluvions Fy1 constitue le mur de l'aquifère. Les alluvions Fy3, particulièrement limoneuses et imperméables, provoquent localement la mise en charge de la nappe au niveau de l'embouchure du Bevincu (Figure 5).

L'aquifère s'étend sur près de 80 km^2, avec une épaisseur allant de 3 à 40 m. Cet aquifère alluvial libre et peu profond est formé par la combinaison des deux nappes d'accompagnement du Golu et du Bevincu, toutes deux hydrauliquement connectées. Le toit de la nappe est sub-affleurant, entre 1 et 5 m de profondeur. Les dynamiques temporelles des niveaux piézométriques sont par conséquent fortement corrélées aux variations des précipitations, démontrant la rapidité de réponse de la nappe aux évènements pluvieux et sa bonne connexion avec les conditions de surface (Figure 9). Les niveaux piézométriques mesurés au niveau du piézomètre Casatorra (11071X0062 - http://www.ades.eaufrance.fr/) illustrent clairement cette dynamique. Les variations saisonnières et interannuelles des niveaux piézométriques sont relativement faibles, de l'ordre de 1 à 2,5 m.

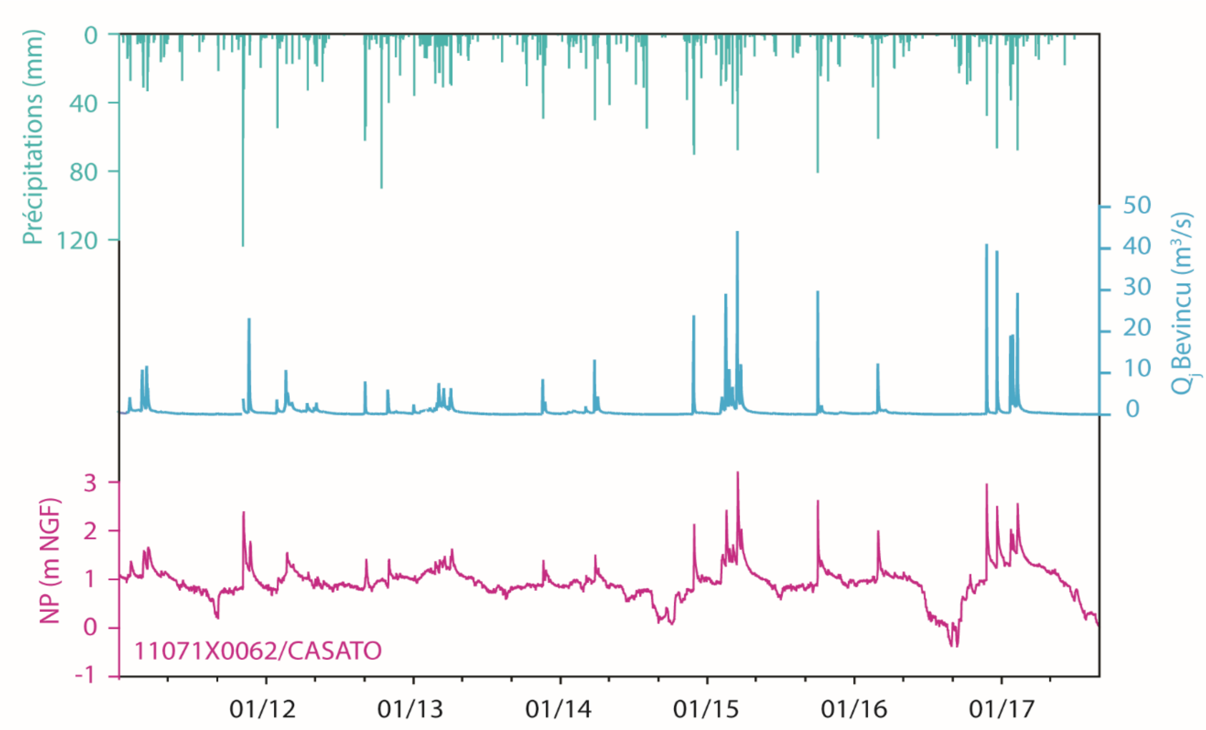

Figure 10 : Chroniques des précipitations journalières, des débits journaliers du Bevincu (Q_j Bevincu) estimés à partir des données limnométriques de la station de Olmeta-di-Tuda et des niveaux piézométriques (NP) mesurés en continu au niveau du piézomètre « Casatorra » (11071X0062 - http://www.ades.eaufrance.fr/).

Le volume du réservoir aquifère est estimé à 40 Mm^3, dont 13 Mm^3 pour les alluvions récentes, avec une conductivité hydraulique estimée à 10^{-3}-10^{-4} m/s (Orofino et al., 2010). La porosité est variable, en lien avec l'hétérogénéité de la sédimentation. En moyenne, elle est estimée à 8% dans les alluvions modernes et descend à 3% dans les dépôts plus anciens. La vitesse de circulation de l'eau dans l'aquifère du Bevincu est estimée entre 1,9 m/j et 3 m/j (DIREN Corse, 2002).

La carte piézométrique montre clairement l'écoulement des eaux souterraines d'Ouest en Est, en direction de la lagune de Biguglia et de la mer (Figure 10). Les gradients hydrauliques sont compris entre 3 et 5 ‰ (Orofino et al., 2010; Garel et al., 2016). Il ne fait aucun doute que l'aquifère contribue de façon significative aux apports en eau douce de la lagune, faisant de cette dernière un écosystème côtier tributaire des eaux souterraines.

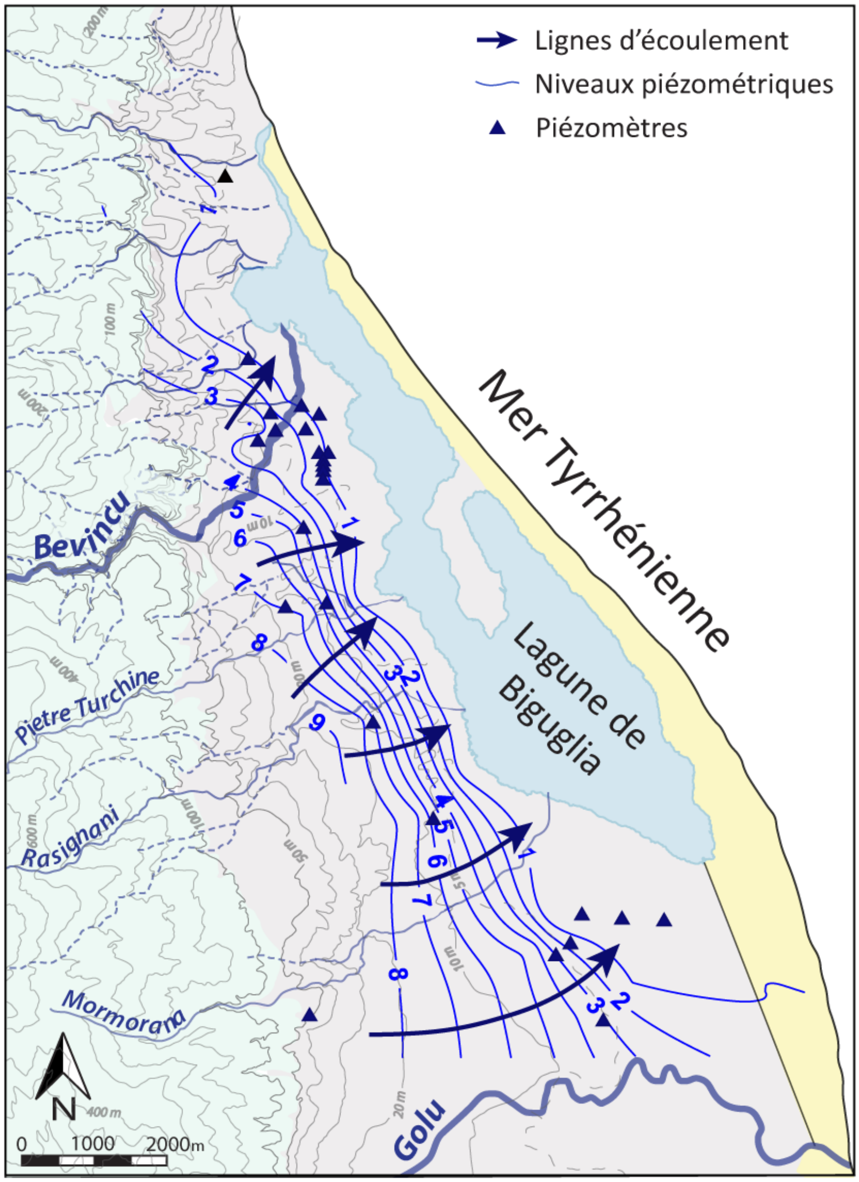

Figure 11: Carte piézométrique de l'aquifère de la Marana basée sur les mesures réalisées en avril 2015, modifiée d'après Erostate et al. (2019).

L'aquifère fournit la quasi-totalité des besoins en eau potable des 72 000 habitants de l'agglomération de Bastia (Orofino et al., 2010). Environ 4 Mm³ sont extraits chaque année, à raison de 1,5 Mm³/an prélevés dans la nappe du Bevincu et 2,5 Mm³/an dans la nappe du Golu (banque nationale de données sur l'eau, http://www.bnpe.eaufrance.fr). La nappe du Golu est également exploitée pour l'alimentation en eau brute des agriculteurs. Enfin, un grand nombre de particulier possède des forages privés non déclarés dont l'utilisation est inconnue de l'administration. Aucune donnée chiffrée n'est donc disponible quant à leur impact sur la ressource.

Des phénomènes d'intrusion saline ont déjà été reportés au niveau du champ captant de la nappe du Bevincu, lors de cas ponctuels de surexploitation de la nappe. Dans la vallée du Golu, la remontée du biseau salé semble principalement localisée au niveau de l'embouchure du Golu et des carrières d'extraction de granulats. L'origine de la salinisation entre eau de mer et eaux lagunaires reste souvent indéterminée.

Malgré l'exploitation assez ancienne, la géométrie des aquifères est encore mal connue (Orofino et al., 2010). Deux niveaux aquifères se superposent dans les alluvions du Bevincu et les relations entre eux (drainage vertical) et avec la lagune sont loin d'être comprises. Jusqu'à présent, les échanges hydrauliques entre l'aquifère et la lagune sont le plus souvent négligés (BCEOM, 2006). Sans véritable preuve scientifique, les eaux souterraines ont été considérées comme les apports en eau les plus faibles parmi l'ensemble des apports participant au fonctionnement hydrologique de la lagune (BCEOM 2006). D'après les estimations du BCEOM, ces apports pourraient représenter entre 5 et 25% des apports totaux annuels d'eau douce. Ces estimations varient d'un facteur 5 en fonction du rôle attribué aux canaux dans le drainage des eaux souterraines.

2.6. Occupations des sols et impacts anthropiques

2.6.1. Réseau de drainage artificiel

L'écoulement naturel de l'ensemble de l'hydrosystème est fortement contraint par les infrastructures hydrauliques construites dès le 19ème siècle. Le réseau de drainage (Figure 12) permet d'assécher la plaine alluviale afin de rendre les terres exploitables en agriculture et de lutter contre la propagation des moustiques (SAGE, 2012). Il est constitué de 5 canaux principaux ceinturant la rive Ouest de la lagune (Figure 12). Depuis le début du 20ème siècle, chaque canal est équipé d'une station de pompage (Le Fort, Petriccia, Quercile, Fornoli et Ghiunchetta) (Figure 13). Les pompages s'activent grâce à un système de flotteur, similaire à celui d'une chasse d'eau. Lorsque l'eau atteint une hauteur fixée, les pompes se déclenchent et permettent alors de réguler le niveau d'eau dans le canal (Figure 14). La hauteur du seuil déclenchant le pompage semble être fixée de manière empirique par les gestionnaires aux alentours de 0,8 à 1 m en dessous du niveau du sol, sur l'ensemble des 5 stations de pompage. L'eau pompée dans les canaux est ensuite directement rejetée dans la lagune. Chaque année, les stations de pompage extraient entre 23 et 39 Mm^3, résultant d'un mélange d'eau lagunaire saumâtre et d'eau douce de surface et souterraine.

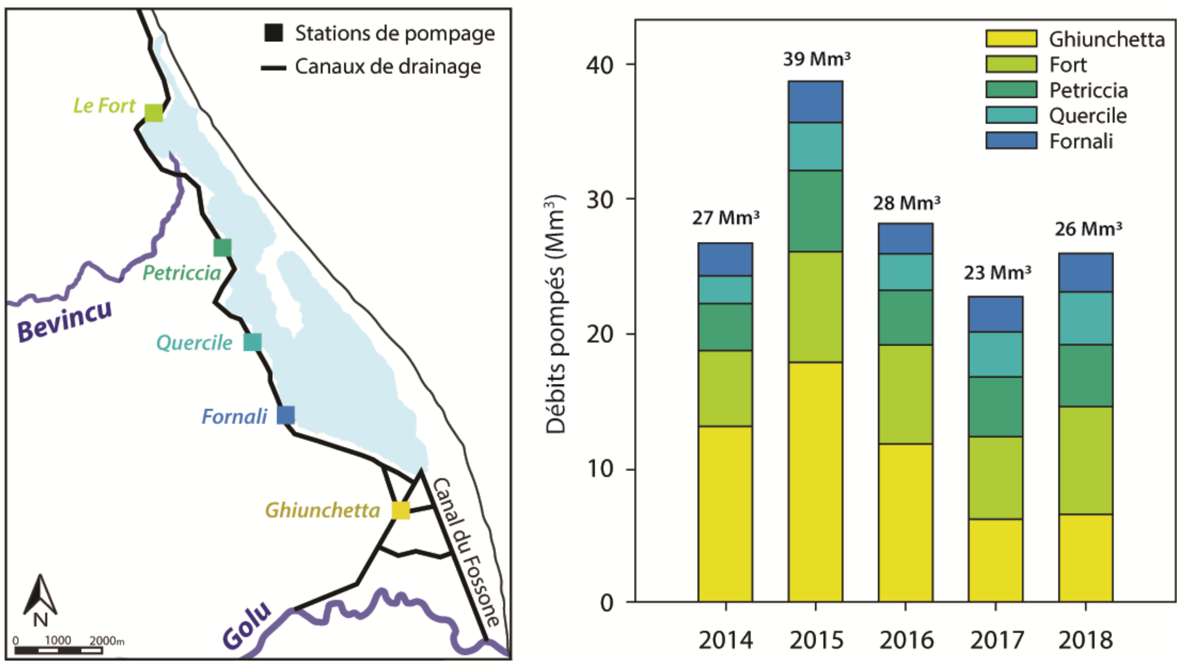

Figure 12: Bilan des débits pompés par les stations de pompage bordant la lagune de Biguglia.

La proportion d'eau souterraine drainée par les stations de pompage varie localement et il est actuellement impossible d'estimer le volume exact d'eau souterraine extrait de l'aquifère alluvial. Il semblerait cependant que l'aquifère soit principalement sollicité au niveau des stations de Quercile et Fornali (Orofino et al., 2010). En période de basses eaux, des phénomènes d'infiltration des eaux de canaux vers l'aquifère peuvent contribuer à sa recharge (Orofino et al., 2010). De par la nature saumâtre des eaux s'écoulant dans les canaux, ces processus représentent un risque localisé de salinisation pour l'aquifère et les terres environnantes. Les relations entre ces canaux et les aquifères qu'ils drainent demeurent actuellement méconnues.

Figure 13: Géométrie d'une station de pompage de la plaine de la Marana : cas de la station du Fortin avec 1) une vue aérienne du réseau de canaux et de la station « Le Fort », 2) l'environnement et le canal à quelques mètres à l'Est de la station de pompage, 3) le bassin de rejet de la station, en cours de pompage, 4) le canal perpendiculaire au bassin de rejet collectant les eaux pompées et 5) la continuité de ce même canal vers la lagune.

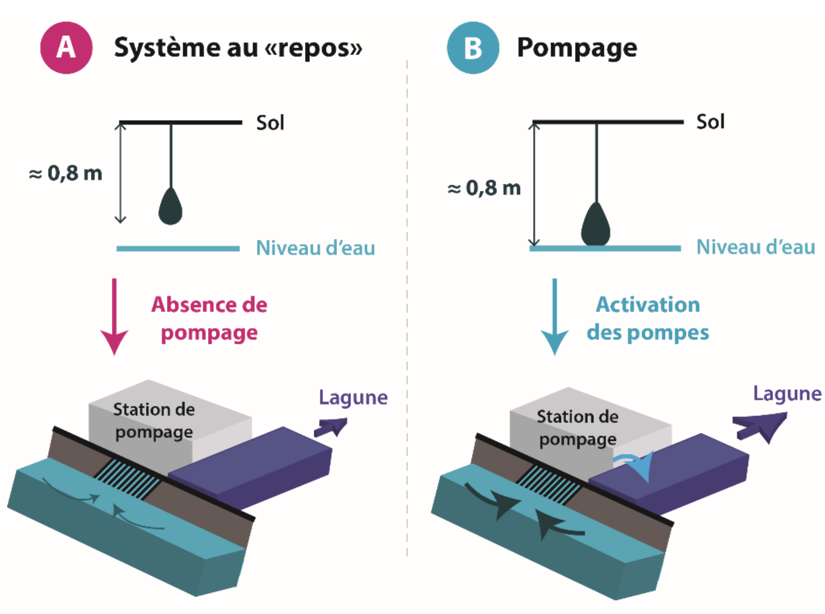

Figure 14: Fonctionnement schématique des stations de pompage de la plaine de la Marana.

2.6.2. Occupation des sols

Aux abords de la lagune, les terrains sont principalement utilisés à des fins agricoles. Cependant, au cours du siècle dernier, les zones urbanisées du bassin versant se sont considérablement développées. Le taux d'urbanisation a particulièrement augmenté depuis le début du 21ème siècle (Fox et al., 2012). Actuellement, la côte de Biguglia est la côte française qui connaît le taux d'artificialisation le plus élevé (Robert et al., 2018).

Des sources multiples de pollution potentielles ont été identifiées sur le bassin versant : effluents domestiques, pollutions agricoles, rejets industriels… Les principaux risques sont attribués aux ouvrages d'assainissement (Département de la Haute-Corse, 2012), en lien avec l'ancienneté des infrastructures et le débordement des stations de relevage en période de fortes précipitations. De plus, l'efficacité du système de traitement des eaux usées est mise à rude épreuve par les fluctuations saisonnières du volume des eaux usées, en lien avec les flux touristiques. La pollution liée aux activités agricoles contemporaines semble occasionner des flux limités de polluants, notamment concernant les matières azotées et phosphorées. Enfin, le développement des infrastructures de transport, concomitant du développement anthropique sur la plaine, est source de déversements ponctuels et/ou diffus d'hydrocarbures et de métaux lourds (Département de la Haute-Corse, 2012).

Les politiques de gestion locales tentent de concilier développement économique, aménagement du territoire et gestion durable de la ressource (SDAGE, 2015). A l'image des politiques de gestion longtemps menées en France, les eaux de surface et les eaux souterraines du bassin versant sont encore abordées de manière trop dissociative. Un manque de connaissance important, et par conséquent de considération, est à signaler quant à l'importance des interactions entre les masses d'eau souterraine, de surface et de transition. L'unicité de la ressource en eau prônée par la Directive Cadre Européenne (DCE - 2000/60/CE) oblige depuis les années 2000 à une évolution des stratégies de gestion. La DCE souligne la nécessité d'une approche de gestion de l'eau dans laquelle les interactions entre masses d'eau souterraine, de surface et écosystèmes jouent un rôle central. Cela implique alors le développement des connaissances sur le fonctionnement global des hydrosystèmes dans l'optique d'élaborer des stratégies de gestion conciliant besoins en eau humains et environnementaux.

Chapitre 3 :

Stratégie d'investigation

Ce chapitre présente succinctement la stratégie d'investigation et les outils utilisés pour mener à bien la compréhension de l'hydrosystème de la lagune de Biguglia. Pour plus de précision, la présentation détaillée des outils et méthodes d'analyse est disponible dans la section « Méthodologie » des différents articles (Chapitres 4 à 6).

3. Stratégie d'investigation

3.1. Définition des points d'échantillonnage

A l'instar de la majorité des nappes alluviales, l'aquifère de la Marana est en liaison hydraulique directe avec les cours d'eau de surface (potentiel hydraulique imposé). Par conséquent, les fluctuations de la nappe, qui sont alors liées au régime des cours d'eau, sont de faibles amplitudes. Elles se reproduisent de manière semblable chaque année, au gré des hautes eaux et des basses eaux saisonnières de la rivière. Ainsi, deux importantes campagnes de terrain ont été réalisées en hautes eaux (avril 2015) et en basses eaux (septembre 2015). Ces campagnes avaient pour but de réaliser une première caractérisation des ressources en eau (chimie, isotopie et hydrodynamique). Elles ont permis d'identifier les secteurs présentant les comportements les plus atypiques et ainsi d'orienter la sélection des points les plus pertinents pour les deux campagnes de prélèvements suivantes (mai 2016 et 2017).

Au total, 59 points de prélèvement ont été identifiés (Figure 15), regroupant 27 puits, 11 forages, 6 piézomètres, 5 points d'échantillonnage dans la rivière Bevincu, 5 points d'échantillonnage dans la lagune de Biguglia, 1 point d'échantillonnage dans un drain et 4 prélèvements d'eaux usées (Tableau 1). Les points de prélèvement des eaux souterraines sont principalement concentrés dans la partie aval du bassin versant, dans les dépôts quaternaires et les contreforts schisteux. Les prélèvements réalisés dans le Bevincu, la lagune de Biguglia et les canaux de drainage ont été réalisés afin de caractériser les interactions nappes-rivière-lagune.

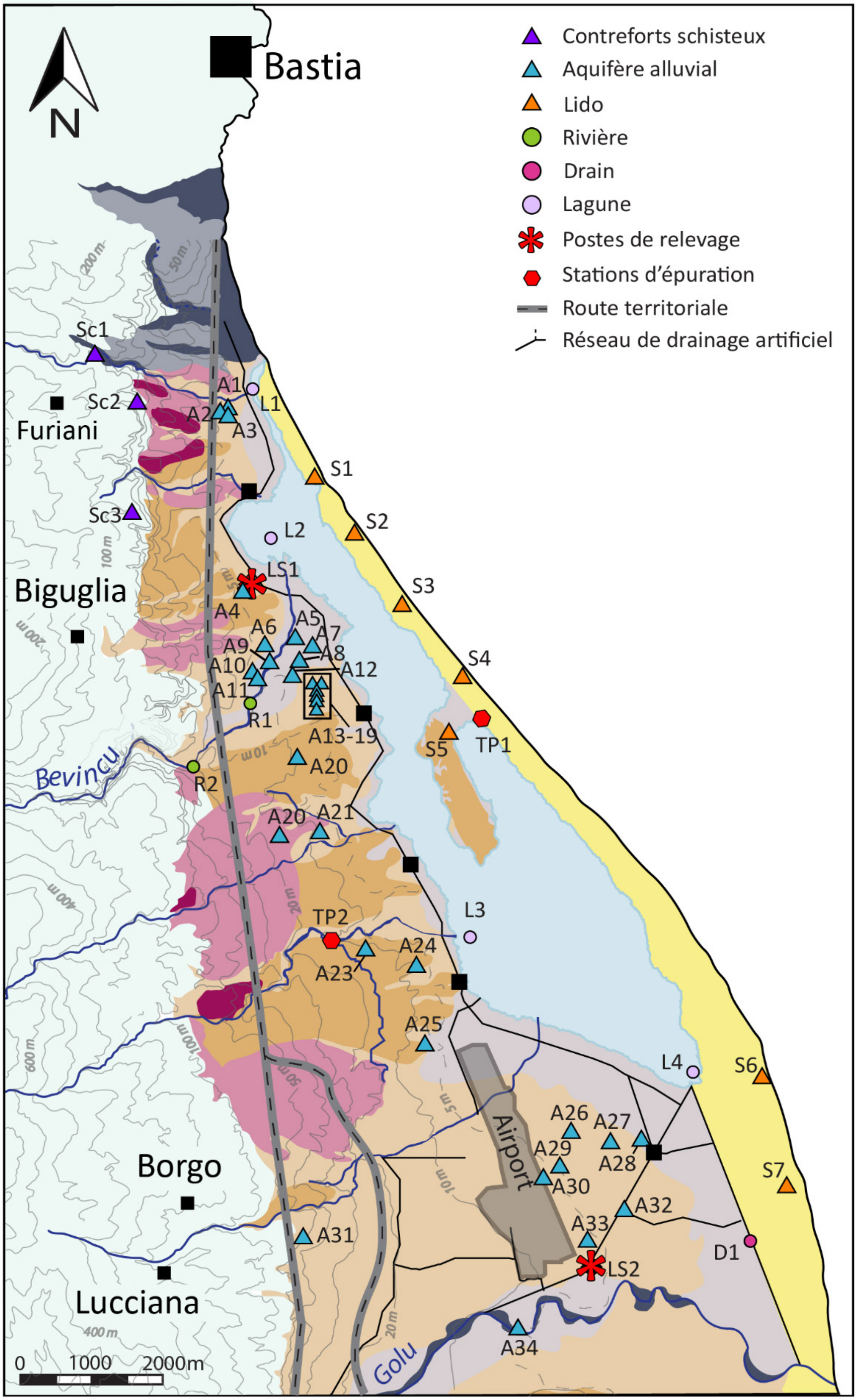

Figure 15 : Localisation de l'ensemble des sites d'échantillonnage de l'hydrosystème de la lagune de Biguglia.

Pour chaque campagne, les mesures physico-chimiques (température, pH, conductivité électrique, oxygène dissous et potentiel redox) ainsi que le dosage *in situ* des bicarbonates par colorimétrie ont été réalisés. Ces données ont été systématiquement complétées par l'analyse des ions majeurs et des isotopes stables de la molécule d'eau. Ensuite, les éléments spécifiques à chaque campagne, définis en fonction des objectifs visés, sont présentés Tableau 1. La campagne de mai 2016 était consacrée à la compréhension de l'origine des flux nitratés et des temps de séjours des eaux souterraines. Un total de 18 points de prélèvements a été analysé pour les signatures isotopique du ^{15}N-NO_3^-, ^{18}O-NO_3^- et du ^{11}B. Parmi ces points, les teneurs en ^{3}H et en CFCs/SF_6 ont été analysées pour 13 et 5 points respectivement. Enfin, la dernière campagne de mai 2017 s'intéressait à l'évaluation de l'impact des fuites d'eaux usées sur la qualité des eaux souterraines et à la caractérisation des processus d'infiltration rapide. 51 composés organiques d'origine domestique ont été analysés sur 18 points de prélèvements, comportant 14 échantillons d'eau naturelle (eaux souterraines et de surface) et 4 échantillons d'eaux usées (stations d'épuration et postes de relevage).

Tableau 1 : Récapitulatif de la nature et de la localisation des points d'échantillonnage ainsi que des campagnes de prélèvements et des paramètres analysés.

Points	**Type**	**Localisation (Lambert 93)**		**Avril 2015**		**Sept 2015**		**Mai 2016**				**Mai 2017**
				^{18}O ^{2}H	**^{3}H**	**^{18}O ^{2}H**	**^{3}H**	**^{11}B**	**$^{15}N-NO_3^-$ $^{18}O-NO_3^-$**	**^{3}H**	**CFCs SF6**	**µpolluants**
Eaux souterraines – Contreforts schisteux												
Sc1	Forage	1226702.5	6195517.8	x		x						
Sc2	Puits	1227207.1	6194282.7	x		x						
Sc3	Forage	1227113.3	6192715.3	x	x	x	x	x	x	x		
Eaux souterraines – Aquifère alluvial												
A1	Forage	1228441.0	6194233.9	x		x		x	x	x	x	
A2	Forage	1228417.7	6194195.0	x	x	x	x					x
A3	Forage	1228534.3	6194111.0	x								
A4	Puits	1228683.9	6191587.2	x		x						
A5	Puits	1229394.4	6191131.5	x		x	x					
A6	Puits	1228962.2	6190855.4	x		x						
A7	Puits	1229624.9	6190946.2	x	x							
A8	Puits	1229383.0	6190670.1	x	x	x	x					
A9	Puits	1228986.8	6190557.3	x		x						
A10	Puits	1228786.8	6190465.1	x	x	x	x					
A11	Puits	1228907.9	6190418.8	x	x	x	x	x	x	x		x
A12	Puits	1229351.8	6190384.1	x		x						
A13	Puits	1229763.0	6190274.8	x		x	x					
A14	Puits	1229674.3	6190263.0	x		x						
A15	Puits	1229711.2	6190174.0	x		x						
A16	Puits	1229708.7	6190114.4	x		x						
A17	Puits	1229714.3	6190067.8	x		x						
A18	Puits	1229725.9	6189979.6	x	x	x	x	x	x	x	x	
A19	Piézomètre	1229094.0	6189984.0					x	x	x	x	
A20	Puits	1229439.3	6189257.1	x	x							
A21	Puits	1229738.9	6188101.9	x		x						
A22	Puits	1229156.1	6188083.4	x	x	x	x	x	x	x		x
A23	Puits	1230473.1	6186524.3	x	x	x	x					
A24	Forage	1231088.1	6186248.4	x	x	x	x	x	x	x	x	x
A25	Puits	1231209.0	6185141.0	x		x		x	x	x		x
A26	Piézomètre	1233242.0	6183809.4	x		x		x	x	x		x
A27	Piézomètre	1233806.5	6183732.8	x		x						
A28	Piézomètre	1234314.6	6183764.6	x		x						
A29	Piézomètre	1233019.8	6183382.9	x		x						
A30	Piézomètre	1232835.4	6183243.4	x		x						
A31	Puits	1229490.7	6182415.9	x	x	x	x	x	x	x		
A32	Puits	1234222.8	6182733.9	x		x						x
A33	Puits	1233534.1	6182301.1	x	x	x	x	x	x	x	x	
A34	Puits	1232652.8	6181163.4	x	x	x	x	x	x	x		

Points	Type	Localisation (Lambert 93)		Avril 2015		Sept 2015		Mai 2016				Mai 2017
				^{18}O ^{2}H	^{3}H	^{18}O ^{2}H	^{3}H	^{11}B	$^{15}N\text{-}NO_3^-$ $^{18}O\text{-}NO_3^-$	^{3}H	CFCs SF6	µpolluants
Eaux souterraines - Lido												
S1	Forage	1229687.0	6193228.8	x		x						x
S2	Forage	1230262.4	6192393.1	x		x						
S3	Forage	1230919.2	6191414.1	x		x		x	x	x		x
S4	Forage	1231824.2	6190419.0	x		x						
S5	Puits	1231472.9	6189659.8	x		x						
S6	Puits	1236000.9	6184646.9	x		x						x
S7	Forage	1236312.3	6183110.9	x	x	x	x					
Rivière												
R1	Rivière	1228766.8	6190080.5	x		x		x	x			x
R2	Rivière	1227832.5	6189124.1	x								
R3	Rivière	1222408.4	6186849.4	x		x						
R4	Rivière	1222692.9	6185483.7	x		x						
R5	Rivière	1217847.1	6184080.1	x		x						
Drain												
D1	Drain	1235844.1	6182621.9	x		x						
Lagune												
L1	Lagune	1228788.2	6194529.8	x		x		x	x			x
L2	Lagune	1229112.5	6192407.6	x		x						x
L3	Lagune	1231845.6	6186731.0	x		x		x	x			x
L4	Lagune	1235023.8	6184716.8	x		x		x	x			
L5	Lagune	1231472.9	6189659.8					x	x			
Eaux usées												
LS1	Poste de relevage	1228776.9	6191671.4									x
LS2	Poste de relevage	1233574.4	6181958.9									x
TP1	Station d'épuration	1232115.9	6189744.6									x
TP2	Station d'épuration	1229916.5	6186596.2									x

3.2. Étude du fonctionnement de l'hydrosystème de Biguglia

3.2.1. Mesure des niveaux piézométriques

Les mesures des niveaux piézométriques permettent de définir l'organisation spatiale des écoulements. Deux campagnes de mesures ont été réalisées en hautes eaux (avril 2015) et en basses eaux (septembre 2015) afin i) d'évaluer les variations saisonnières des niveaux de nappe, ii) de préciser la dynamique de l'aquifère et iii) de démontrer le caractère tributaire de la lagune aux eaux souterraines.

3.2.2. Recharge des eaux souterraines par les précipitations

En comparant la composition isotopique des précipitations et des eaux souterraines, il est alors possible de préciser les conditions de recharge de l'aquifère. Cette approche requiert cependant une bonne caractérisation du signal d'entrée et donc de la signature isotopique des précipitations. Pour cela, les précipitations ont été collectées mensuellement grâce à 2 pluviomètres et analysées ($\delta^{18}O$ et $\delta^{2}H$). Le premier, situé à côté de Bastia (au niveau du Fortin, dans la partie Nord de la lagune), permet de caractériser la signature isotopique des précipitations en plaine (1m NGF). La signature isotopique des pluies de plus haute altitude (contreforts schisteux du bassin versant de Biguglia) est appréhendée grâce au pluviomètre installé à Corte (500m NGF).

3.2.3. Quantification de la recharge

Une incertitude importante demeure quant à la recharge de l'aquifère de la Marana prodiguée par les précipitations. Un important travail consacré à l'estimation du taux de recharge de l'aquifère a été réalisé, grâce notamment à l'usage de la feuille de calculs ESPERE_v1.5 (Lanini et al., 2016).

3.2.4. Caractérisation géochimique et isotopique des eaux souterraines

Les précipitations ne constituant pas l'unique source de recharge pour les aquifères, il est essentiel d'appréhender plus largement l'origine des eaux souterraines de la plaine de la Marana. La proximité des eaux salées marines et saumâtres lagunaires constitue une menace potentielle pour la qualité des eaux souterraines. De plus, la nature des échanges entre l'aquifère alluvial et les contreforts schisteux était encore mal documentée. La caractérisation géochimique (majeurs, traces) et isotopiques ($\delta^{18}O$, $\delta^{2}H$, ^{3}H) des eaux souterraines et de surface a donc été entreprise afin d'identifier les processus d'infiltration (interactions nappes-rivières, nappes-lagune et nappes-mer) et les échanges latéraux entre l'aquifère alluvial et les contreforts schisteux.

Cette caractérisation a également permis d'évaluer l'état qualitatif des eaux souterraines et de surface de l'hydrosystème de Biguglia. L'aquifère de la Marana constitue une ressource stratégique pour l'alimentation en eau potable de la ville de Bastia et des communes alentours, qu'il convient de préserver. Une inconnue quasi totale subsiste encore sur l'origine des fortes teneurs en azote enregistrées dans les eaux lagunaires et les eaux souterraines pourraient constituer un vecteur important de nutriments et de polluants. En raison de l'importante urbanisation développée sur la plaine et de l'état vétuste du réseau d'assainissement évoqué dans les documents de gestion, il semblait important de s'attarder sur la dégradation induite par les eaux usées (éléments traces, ^{15}N-NO_3^-, ^{18}O-NO_3^-, ^{11}B et micropolluants organiques). De plus, les modifications majeures de l'occupation du sol et l'évolution des pratiques agricoles au cours du dernier siècle ont induit un fort remaniement des sols, susceptible d'impacter la qualité des eaux d'infiltration vers l'aquifère.

3.2.5. Interaction eaux-roches et processus de mélange

L'étude des interactions eau-roche permet de préciser les conditions d'écoulement et de quantifier (ou semi-quantifier) les processus de mélange. Les forts contrastes altitudinaux et lithologiques présents sur le bassin versant de la lagune de Biguglia ainsi que la nature des masses d'eau en interaction (fluviales douces et faiblement chargées en ions ; souterraines douces et fortement chargées ; saumâtres/salées très fortement chargées) laissent supposer des compositions géochimiques et isotopiques bien distinctes permettant l'élaboration d'un modèle de mélange robuste.

3.2.6. Temps de séjour

L'estimation des temps de séjour des eaux souterraines permet d'évaluer la capacité de renouvellement de la ressource. La répartition spatiale des âges permet d'identifier les secteurs plus inertiels, avec des temps de résidence plus longs, et ainsi de préciser les conditions d'écoulement au sein de l'aquifère. Les teneurs en ^{3}H ont d'abord été utilisées pour permettre une première estimation relative des temps de séjour des eaux souterraines. Ces données ont ensuite été complétées par l'analyse des CFCs/SF_6 afin de préciser les temps de séjour et d'essayer de proposer une estimation chiffrée de l'âge apparent des eaux au sein de l'aquifère de la Marana.

3.2.7. Flux d'infiltration rapide

Les aquifères alluviaux, s'ils ne sont pas recouverts de sédiments imperméables (nappe libre), sont particulièrement sujets aux flux d'infiltration rapide. La bonne perméabilité des alluvions facilite l'infiltration des eaux de surface vers la zone saturée et augmente par la même occasion la vulnérabilité des eaux souterraines aux pollutions. Les micropolluants organiques, et tout particulièrement les composés peu conservatifs ou labiles, peuvent constituer des traceurs pertinents des flux d'infiltration rapide. Cependant, la complexité de leur comportement et de leur devenir dans l'environnement rend leur exploitation particulièrement complexe. Cette approche permet de tester le potentiel des micropolluants comme traceurs hydrologiques des flux d'infiltration rapide dans le cas des aquifères alluviaux fortement anthropisés.

Chapitre 4 :

Compréhension du fonctionnement global de l'hydrosystème

Ce chapitre présente les avancées réalisées sur la compréhension du fonctionnement de l'hydrosystème de Biguglia grâce à une approche multi-traceurs, couplant traceurs géochimiques et isotopiques et micropolluants organiques. Il précise ainsi i) le fonctionnement de l'aquifère de la Marana, ii) l'état qualitatif de la ressource en eau et sa vulnérabilité face à l'urbanisation croissante et iii) quantifie les relations entre les eaux souterraines et les eaux de surface douces, lagunaires et marines. Enfin, il expose également les avancées méthodologiques réalisées quant à l'usage des micropolluants organiques dans la compréhension des processus hydrologiques.

4. Compréhension du fonctionnement global de l'hydrosystème

4.1. Introduction

Les GDEs côtiers se caractérisent par des fonctionnements complexes résultant d'interactions fortes entre les masses d'eau. L'hydrosystème de Biguglia ne fait pas exception. Le suivi des niveaux piézométriques des eaux souterraines de la Marana confirme sa connexion hydraulique directe avec les cours d'eau de surface, principalement du Bevincu et du Golu ainsi que l'influence des épisodes pluvieux (voir section 2.5). Cependant, les eaux fluviales et les précipitations ne sont pas les seules à pouvoir contribuer à la recharge de l'aquifère. La présence de schistes altérés, potentiellement aquifères, à l'Ouest du bassin versant laisse supposer des apports latéraux. De plus, le réseau de drainage artificiel de la plaine contraint fortement l'hydrodynamique du système. Le rôle des canaux concernant le drainage des eaux souterraines et/ou l'infiltration d'eau vers l'aquifère reste encore largement inconnu. S'il ne fait aucun doute sur la contribution des eaux souterraines à l'alimentation en eau douce de la lagune de Biguglia (voir section 2.5), les processus de déversement restent encore à préciser. En effet, il se pourrait que la décharge naturelle des eaux souterraines dans la lagune soit un processus limité par la nature argileuse du bassin lagunaire, comme ce fût le cas pour d'autres lagunes méditerranéennes (David et al., 2019). Dans ce cas, les stations de pompage pourraient jouer un rôle réellement majeur autant sur les volumes d'eau souterraine extraits que sur le contrôle de la salinité de la lagune de Biguglia.

Les traceurs géochimiques et isotopiques sont des outils largement exploités pour la compréhension des hydrosystèmes et la caractérisation des processus de mélange (Liu and Yamanaka, 2012; Prada et al., 2015; Santoni et al., 2016; Gautam et al., 2018). Cependant, ils atteignent souvent leur limite d'applicabilité dans le cas des processus d'infiltration rapide, d'ordre mensuel à annuel. Pourtant, pour les aquifères en forte connexion avec les eaux de surface, l'infiltration peut être un processus particulièrement rapide, source de recharge mais également de potentielles contaminations, qu'il convient de préciser pour assurer la gestion durable des ressources en eau. Dans ce but, les micropolluants organiques peuvent constituer des outils complémentaires très pertinents. Ces polluants, presque uniquement d'origine humaine, possèdent des propriétés physico-chimiques qui contrôlent leur persistance dans l'environnement. Ainsi, la présence de micropolluants organiques facilement dégradables dans l'environnement peut par exemple permettre d'identifier une infiltration d'eau ou encore une contamination récente ou continue (eaux usées, lixiviats agricoles…). A l'inverse, la présence de micropolluants organiques extrêmement persistants peut, elle, informée sur le comportement inertiel des eaux souterraines, l'accumulation de polluants ou encore l'absence de processus de remédiation. Quantifier et comprendre le comportement des micropolluants organiques peut ainsi apporter des informations essentielles pour la compréhension des processus hydrologiques et renforcer les résultats acquis à l'aide de traceurs plus « conventionnels ».

4.2. Résultats et interprétations

Les résultats et interprétations de ces travaux ont fait l'objet de la rédaction d'un article publié dans "Journal of Hydrology" en novembre 2019, intitulé « Coupling isotope hydrology, geochemical tracers and emerging compounds to evaluate mixing processes and groundwater dependence of a highly anthropized coastal hydrosystem », présenté ci-après. Les principales avancées concernant la compréhension de l'hydrosystème de Biguglia sont également résumées en français, en conclusion de ce chapitre.

Coupling isotope hydrology, geochemical tracers and emerging compounds to evaluate mixing processes and groundwater dependence of a highly anthropized coastal hydrosystem

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Research papers

Coupling isotope hydrology, geochemical tracers and emerging compounds to evaluate mixing processes and groundwater dependence of a highly anthropized coastal hydrosystem

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ABSTRACT

Coastal aquifers can be considered as an important socio-economic and ecological component worldwide. The implementation of sustainable groundwater management is made difficult by complex hydrological behaviors due to many interactions between groundwater and fresh, brackish and salt surface water. This study proposes an original multi-tracer approach including physico-chemical parameters, trace elements, ^{18}O, ^{2}H, ^{3}H of the water molecule and emerging organic compounds (EOCs) in order to improve the understanding of strongly urbanized coastal aquifers. This methodology was applied to a Mediterranean coastal aquifer in northern Corsica (France). Firstly, the isotopic data allow highlighting the complex recharge of groundwater, provided by both allochthonous rainfall from the mountain and autochthonous rainfall on the Marana-Casinca alluvial plain. Secondly, geochemical data coupled with isotopic signatures allow the identification of the different contributors and the quantification of the mixing processes into the aquifer. The residence time of groundwater were estimated with ^{3}H. In addition, to display inertial areas with long residence time, ^{3}H also highlights areas subject to infiltration of recent water. A better understanding of short-time hydrological processes was improved by EOCs. The study of the fate of EOCs in the environment provided a higher level of resolution than geochemical and isotopic tracers alone. Thus, the multi-tracing approach using isotopic and geochemical data, coupled with the study of the fate of EOCs, made possible to specify the conceptual model in highly anthropized hydrosystems, and particularly fast hydrological processes.

1. Introduction

Coastal aquifers ensure a fundamental role for both anthropogenic activities and environmental preservation. They supply a large quantity of freshwater for drinking, agricultural and industrial uses in many countries (Sappa et al., 2015; Comte et al., 2016). They also provide a large part of the fresh water supplies to coastal wetlands, consequently referred as groundwater dependent ecosystems (GDE) (Cartwright and Gilfedder, 2015; Krogulec, 2016; Menció et al., 2017). These sensitive environments play a major ecological role and provide a number of ecosystem services such as food production, recreation areas, coastal and soil protection and climate regulation (de Groot et al., 2002; Rao et al., 2015; Newton et al., 2018; Velasco et al., 2018). With the global change effects, the importance of groundwater supplies should be exacerbated, especially in the Mediterranean area (IPCC, 2014; Sellami et al., 2016; Calvache et al., 2017). The expected modifications in precipitation patterns, aquifer recharge rates and groundwater uses are among the many factors what should greatly impact the groundwater management. In coastal areas, these predicted disruptions are added to an already very precarious fresh water/salt water equilibrium.

Coastal aquifers can be influenced by several fresh and saline water bodies, *i.e.* groundwater, surface water, transitional water and seawater. The resulting mixing processes between these different water bodies then lead to a complex hydrodynamic behavior. The determination and quantification of each end-member participation can be assessed by well-known indicators such as groundwater geochemical composition (Prada et al., 2015; Santoni et al., 2016b; Wang et al., 2016) and isotopic signatures (Liu and Yamanaka, 2012; Gautam et al., 2018). However, other less commonly applied tracers, such as "emerging organic pollutants" (EOCs) can be helpful to determine the

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contribution of particular anthropogenic components (sewage, urban and agricultural runoff) into the aquifers (Lapworth et al., 2012).

EOCs are closely linked to anthropogenic consumption (pharmaceuticals, domestic use or personal care compounds) and its detection in the environment is growing due to the development of analytical tools (Richardson and Ternes, 2018). Among multiple pathways to the environment, the occurrence of EOCs in natural waters is mainly related to urban runoff and wastewaters (Lapworth et al., 2012; Vystavna et al., 2012; Sui et al., 2015). Hence, EOCs are detectable in every anthropized areas impacted by sewage contamination. This is especially true in coastal areas, where the rapid urbanization, in response to a growing demography (Small and Nicholls, 2003), often induces water contamination by septic tank or leakages from sewers (Dougherty et al., 2010; Metcalfe et al., 2011; Del Rosario et al., 2014). The EOCs persistence in natural waters and particularly in groundwater is difficult to constrain, due to their largely under-investigated behavior in environmental media (Lapworth et al., 2012). According to their own physical and chemical properties, some EOCs can be exceptionally persistent whereas other can be easily degradable in the environment (El-Shahawi et al., 2010; Vystavna et al., 2013, Vystavna et al., 2017). To some extent, the persistence of EOCs can provide useful information on groundwater residence time, recharge and mixing processes (Sassine et al., 2015; Zirlewagen et al., 2016). For example, the presence of rapidly degradable EOCs will indicate short residence time and recent anthropogenic influence (Lapworth et al., 2018). In contrast, the presence of persistent EOCs can indicate the archiving and/or the low retention capacities of the aquifer. The known degradation patterns of EOCs can therefore supplement geochemical and isotopic tracers to detail the temporal resolution of hydrological processes. The occurrence of EOCs is also used to evaluate the infiltration of contaminated and non-contaminated surface water into the aquifers and to identify recharge areas (Sassine et al., 2015).

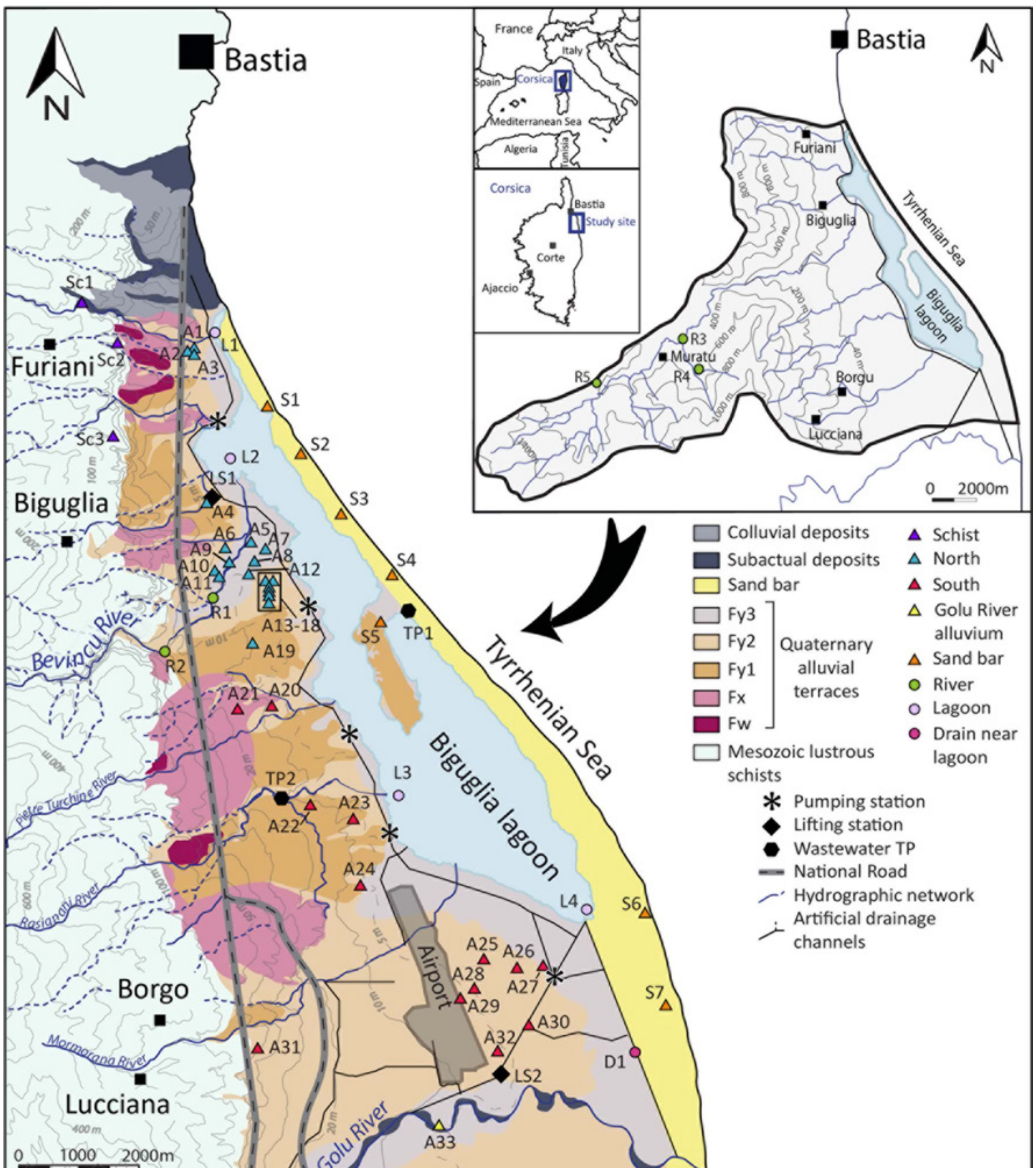

Fig. 1. Location of the study area with simplified presentation of the regional geology and sampling sites.

The aim of this study is to put forward an innovative approach combining geochemical/isotopic data with the fate of emerging compounds in groundwater in order to 1) understand the behavior of anthropized hydrosystems, 2) evaluate the anthropogenic impact and 3) precise groundwater/surface water interaction and rapid infiltration. This study focuses on the Marana-Casinca hydrosystem, located in the North East of Corsica island (France), as a representative example of Mediterranean socio-economic conditions. In this coastal hydrosystem, the sensitive Biguglia coastal lagoon represents the ultimate water collector of a strongly anthropized watershed.

2. Study area

2.1. Geological and hydrological settings

The Biguglia lagoon is located in the northern part of the Corsica Island (western Mediterranean, France), in the South of the densely urbanized city of Bastia (Fig. 1). This shallow brackish coastal lagoon (maximum depth of 1.8 m) is part of the largest wetland in Corsica, with a surface of 14.5 km^2. Located along the coastline, the length of the lagoon is around 11 km with a width of up to 2.5 km in the southern part. The Biguglia lagoon is connected with the Tyrrhenian Sea by a narrow, natural channel at its northern end. Exchange between the lagoon and the sea water is limited (Mouillot et al., 2000; Pasqualini et al., 2017) mainly because of sand accumulations that regularly fill and close the channel. The lagoon has been classified as Natural Reserve since 1994 and recognized as a RAMSAR site in 1991 (Convention on Wetlands of International Importance especially as Waterfowl Habitat, 1971), which emphasizes on the international relevance of this study area.

The region is under a typical Mediterranean climate, implying large seasonal and inter annual variations of temperatures and precipitations. The mean annual temperature and precipitation measured at the Bastia Météo France Station are 15.8 °C and 774 mm respectively (mean calculated for 1957–2016). The Biguglia lagoon watershed is a fairly large area, with around 182 km^2 and bound to the West and the North by high reliefs (until 1450 masl) formed by lustrous schist formations (Fig. 1). The southern part of the watershed is formed by the Marana-Casinca alluvial plain. The watershed is drained by a major river, the Bevincu River, to the North and several smaller non-perennial tributaries only flowing during the rainfall periods. Besides, the Golu River, which originates at 2525 masl in the Hercynian Massifs of inner Corsica, is connected to the lagoon by an artificial channel.

The Marana-Casinca alluvial plain is composed by Quaternary sediment deposits and consists of East to West oriented nested paleo-terraces, eroded by the two main rivers, the Golu River and the Bevincu River. The alluvial terraces were formed during the last glacial phases and were altered during interglacial periods. Regardless of their age, all terraces are composed of gravels, pebbles or blocks, in a sandy-loamy matrix (Conchon, 1972, 1978).

The Marana-Casinca alluvial plain represents the main aquifer system of the region. This shallow alluvial aquifer is constituted by the combination of the two hydraulically connected alluvial aquifers of the Bevincu and the Golu. This aquifer is spread over 80 km^2, with a thickness of 3–40 m and a hydraulic conductivity around 10^{-3}–10^{-4} m/s (Orofino et al., 2010). It is mostly developed in the Middle and Upper Wurmian alluvial deposits (Fy2 and Fy3, Fig. 1) and perched on an impermeable clay-deposit layer formed during the Lower Wurmian (Fy1).

The piezometric map (Fig. 2a), based on piezometric measurements

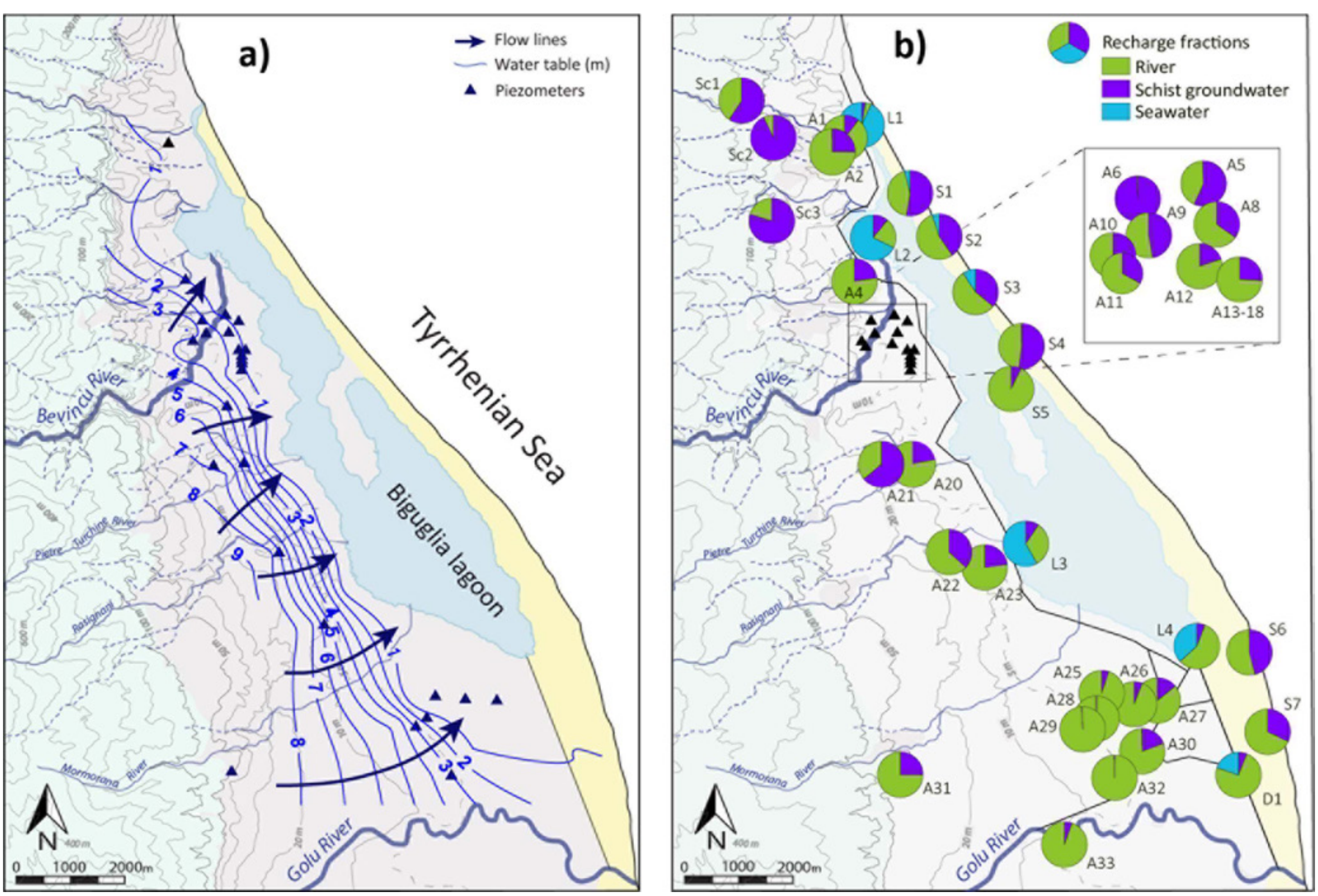

Fig. 2. a) Spatial distribution of aquifer water levels in the study area (April 2015) and b) quantification of mixing processes in the aquifer, between schist, river water and sea water.

realized in April 2015, clearly shows that groundwater flows towards the lagoon with gradients between 3 and 5‰ (Orofino et al., 2010; Garel et al., 2016). Thus, the alluvial aquifer is a significant source of freshwater for the lagoon, making the latter an important coastal GDE.

2.2. Anthropogenic impact

The surface hydrology and the natural behavior of the whole hydrosystem are strongly controlled by man-made hydraulic infrastructures. In the early 20th century, an artificial drainage network (Fig. 1) was constructed. Channels represent a specified head boundary condition and drain the surface groundwater. They therefore lower the water table and allow to dry up the Marana-Casinca alluvial plain in order to use the land for agricultural activities and mosquito control. Then, five pumping stations, located on the west coast of the lagoon, pump the water in the channels when the levels are too high and reallocate this freshwater directly into the lagoon. The huge water volumes withdrawn by the pumping stations, between 23 Mm^3/y and 39 Mm^3/y result from the mixing of fresh groundwater and salt lagoon water. Consequently, estimating the exact groundwater volume extracted from the alluvial aquifer is currently impossible.

The alluvial aquifer is used for the drinking water supply of Bastia's urban area (almost 72,000 inhabitants). The water demand is increasing simultaneously with the growing urbanization that started in the 1950′s as well as touristic pressure, especially during the summer. The water volumes pumped for the drinking supply represent about 4 Mm^3/y (national water data bank, http://www.bnpe.eaufrance.fr).

Over the last century, urbanized areas on the catchment have considerably developed. The rate of urbanization has particularly increased since the beginning of the 21st century, mainly on the Marana-Casinca alluvial plain (Fox et al., 2012). Currently, the coast of Biguglia is one of the French coasts undergoing the highest rate of artificialization

Table 1
Entire set of analysed EOCs, in non-treated sewage water samples of lifting stations and wastewater TP.

EOCs (ng/L)	LS1	LS2	TP1	TP2
Bisphenol A	2000	1800	2500	1700
Ibuprofen	4100	4000	6000	7600
Caffeine	150,000	150,000	79,000	170,000
Paracetamol	160,000	110,000	18,000	120,000
Cotinine	5700	5260	12,100	9850
Paraxanthine	91,300	46,700	78,000	120,000
Acesulfam	9600	9600	12,000	11,000
Carbamazepine	930	130	410	3500
Erythromycin	210	12	370	< 10
Sulfamethoxazol	95	78	630	780
Iopromid	150	110	50	< 50
Diclofenac	< 20	3000	2600	8600
Iopamidol	61	21,000	5100	320
Atenolol	1800	2600	2800	2300
Ketoprofen	2100	2100	2100	2100
Metoprolol	160	11.3	117	< 10
Peniciline G potassium salt	< 10	< 10	< 10	< 10
Sulfamerazine	< 10	< 10	< 10	< 10
Sulfamethazin	< 10	< 10	< 10	< 10
Sulfapyridin	11	10	150	1100
Trimetoprim	25	23	170	170
Furosemide	2700	1300	2600	2300
Gemfibrozil	350	42	210	72
Hydrochlorothiazide	1300	1000	1400	1900
Naproxene	1600	9300	2800	13,000
Triclocarban	250	300	370	170
Triclosan	530	440	330	540
Chloramphenicol	< 20	< 20	< 20	< 20
Bezafibrate	17	260	150	< 10
Warfarin	< 10	< 10	< 10	< 10
Saccharin	19,000	19,000	23,000	33,000
Gabapentin	5300	4000	5700	5100
Tramadol	2100	1400	2100	2200
Sulfanilamide	< 50	< 50	< 50	< 50
Clarithromycin	230	42	110	< 10
Roxithromycin	130	45	87	75
Azithromycin	1300	3800	2500	160
Carbamazepine 10,11-epoxide	14	< 10	33	43
Carbamazepine 10,11-dihydro-10-hydroxy	74	21	450	100
Carbamazepine 10,11-dihydroxy	< 10	< 10	< 10	< 10
Oxcarbazepine	< 10	< 10	37	< 10
Ibuprofen-2-hydroxy	8800	9500	15,000	20,000
Ibuprofen-carboxy	19,000	20,000	31,000	35,000
Diclofenac-4′-hydroxy	33	120	< 20	470
Naproxene-o-desmethyl	470	4600	560	4200
Venlafaxine	570	180	310	1400
Sertraline	31	< 10	< 10	33
Ranitidin	< 10	< 10	43	25
Iohexol	65	< 50	< 50	< 50
Carbamazepine 2-hydroxy	46	21	100	410
Clofibric acid	35.4	< 10	84.8	23.4

Table 2
Annual rainfall at the Bastia Météo France station, mean annual temperature and aquifer recharge rate estimates from empirical and Choride Mass Balance (CMB) methods from 1957 to 2015. Mean, median and standard deviation are calculated for each year and each method.

Year	Annual rainfall amount (mm)	Mean annual temperature (°C)	Thornthwaite (mm) %		Dingman-Penman (mm) %		Dingman-Hamon (mm) %		Turc (mm) %		Mean	Median	Standard deviation
1957	626		56	9%	77	12%	73	12%	83	13%	72	75	12
1958	641	14.9	172	27%	197	31%	83	13%	88	14%	135	130	58
1959	809	15.1	93	11%	128	16%	127	16%	150	19%	124	127	24
1960	684	15.0	67	10%	102	15%	111	16%	103	15%	96	103	20
1961	372	15.7	16	4%	18	5%	19	5%	16	4%	17	17	2
1962	665	15.1	83	13%	106	16%	112	17%	96	14%	99	101	13
1963	672	15.0	75	11%	96	14%	123	18%	99	15%	98	97	20
1964	785	15.7	100	13%	154	20%	159	20%	137	17%	137	145	27
1965	424	15.3	3	1%	6	1%	20	5%	27	6%	14	13	11
1966	708	15.6	63	9%	87	12%	100	14%	108	15%	90	93	20
1967	509	15.8	60	12%	64	13%	69	14%	45	9%	59	62	10
1968	1031	15.4	158	15%	206	20%	230	22%	245	24%	210	218	38
1969	758	15.2	126	17%	132	17%	152	20%	130	17%	135	131	12
1970	402	15.4	0	0%	8	2%	22	6%	22	5%	13	15	11
1971	708	15.1	58	8%	84	12%	115	16%	111	16%	92	97	26
1972	1234	15.4	270	22%	304	25%	330	27%	341	28%	311	317	32
1973	804	15.5	156	19%	164	20%	193	24%	146	18%	165	160	20
1974	753	15.4	126	17%	166	22%	183	24%	126	17%	150	146	29
1975	866	15.5	34	4%	104	12%	137	16%	171	20%	111	120	58
1976	1123	15.3	151	13%	211	19%	239	21%	288	26%	222	224	57
1977	615	15.9	2	0%	7	1%	16	3%	75	12%	25	12	34
1978	1086	15.2	162	15%	240	22%	287	26%	271	25%	240	256	56
1979	1076	15.7	263	24%	288	27%	301	28%	263	24%	279	275	19
1980	859	14.8	142	17%	178	21%	199	23%	173	20%	173	176	23
1981	560	15.5	11	2%	23	4%	50	9%	60	11%	36	36	23
1982	842	16.5	62	7%	143	17%	171	20%	14	18%	133	149	48
1983	623	15.9	0	0%	5	1%	26	4%	78	12%	27	15	36
1984	845	14.9	33	4%	108	13%	133	16%	167	20%	110	120	57
1985	693	15.6	141	20%	161	23%	162	23%	103	15%	142	151	28
1986	922	15.8	194	21%	200	22%	205	22%	193	21%	198	197	6
1987	784	15.9	71	9%	140	18%	156	20%	135	17%	126	138	38
1988	661	16.0	56	9%	89	13%	85	13%	89	14%	80	87	16
1989	551	16.1	22	4%	34	6%	42	8%	55	10%	38	38	14
1990	557	16.3	13	2%	36	6%	50	9%	56	10%	39	43	19
1991	926	15.3	141	15%	179	19%	196	21%	198	21%	178	187	27
1992	977	16.0	160	16%	188	19%	197	20%	215	22%	190	192	23
1993	1046	15.9	286	27%	312	30%	325	31%	247	24%	292	299	34
1994	773	17.0	32	4%	104	13%	131	17%	124	16%	98	114	45
1995	659	16.0	3	0%	30	4%	53	8%	89	13%	43	41	37
1996	1104	15.8	192	17%	231	21%	269	24%	275	25%	242	250	38
1997	776	16.6	106	14%	129	17%	145	19%	127	16%	127	128	16
1998	690	16.2	11	2%	50	7%	82	12%	99	14%	60	66	39
1999	981	16.5	210	21%	250	25%	257	26%	213	22%	232	231	25
2000	941	16.5	104	11%	144	15%	183	19%	195	21%	157	163	41
2001	594	16.6	32	5%	54	9%	79	13%	66	11%	58	60	20
2002	719	16.4	6	1%	33	5%	57	8%	108	15%	51	45	43
2003	676	17.0	100	15%	123	18%	137	20%	89	13%	112	111	21
2004	678	16.1	66	10%	84	12%	85	13%	95	14%	82	84	12
2005	847	15.4	137	16%	173	20%	191	23%	164	19%	166	168	22
2006	621	16.2	22	4%	51	8%	60	10%	76	12%	52	55	22
2007	865	16.1	142	16%	167	19%	190	22%	166	19%	166	166	19
2008	1375	16.0	402	29%	444	32%	459	33%	404	29%	427	424	28
2009	652	16.4	26	4%	41	6%	68	10%	85	13%	54	54	26
2010	1035	15.6	191	18%	225	22%	245	24%	244	24%	226	234	25
2011	742	16.3	102	14%	135	18%	142	19%	117	16%	124	126	18
2012	673	16.3	0	0%	27	4%	48	7%	92	14%	41	37	39
2013	708	16.1	78	11%	98	14%	124	18%	105	15%	101	99	19
2014	766	16.9	44	6%	61	8%	78	10%	122	16%	104	78	33
2015	756	16.9	127	17%	141	19%	155	21%	118	16%			
Mean	777	15.8	98	11%	128	15%	143	17%	140	17%	127	129	27
Median	753	15.8	78	11%	123	16%	133	18%	118	16%	112	120	24
Standard deviation	200	0.6	83	8%	89	8%	90	7%	79	5%	84	85	14

(Robert et al., 2018). The sewage mains are located on the Marana-Casinca alluvial plain and these old infrastructures induce many leakages which represent the main pollution source to the alluvial aquifer (Erostate et al., 2018). The treatment efficiency of the sanitation system is strained by the large seasonal fluctuation of wastewater volume because of a highly variable amount of tourists. In addition to groundwater, the lagoon water quality is also deteriorated due to eutrophication processes (Département de la Haute-Corse, 2012; Garrido et al., 2016; Pasqualini et al., 2017).

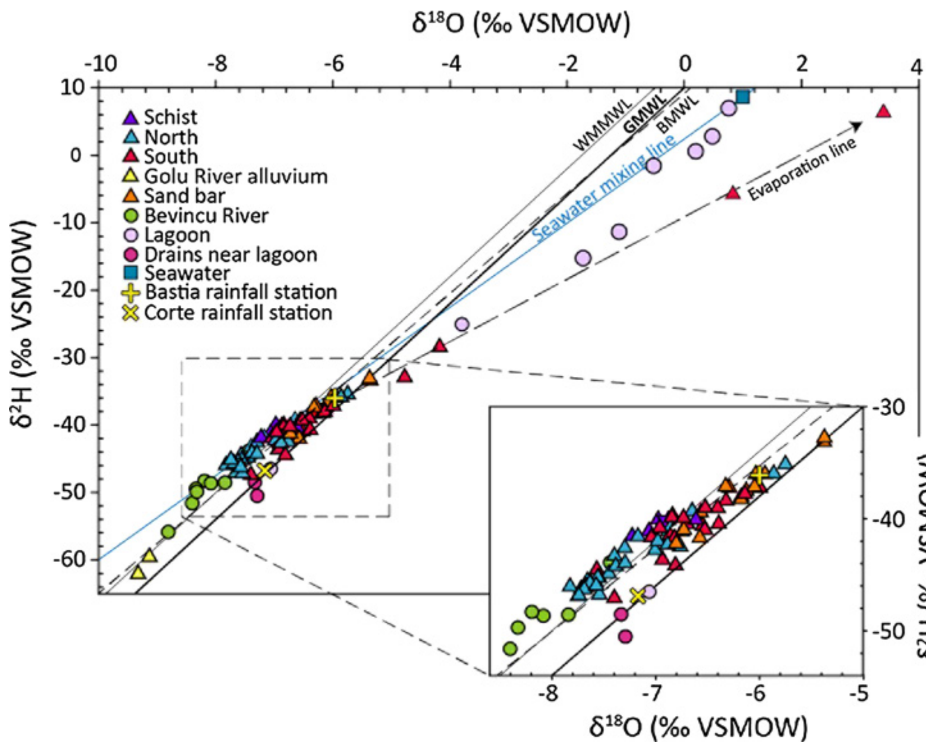

Fig. 3. δ^2H vs. $\delta^{18}O$ for all the sampled points and weighted mean values for rainwater collected at Corte and Bastia. Also shown is the Global Meteoric Water Line (GMWL) from Craig (1961) and the Western Mediterranean Meteoric Water Line (WMMWL) from Celle-Jeanton et al., (2001).

3. Methodology

3.1. Sampling procedure and geochemistry

Two sampling campaigns were carried out during the high (April 2015) and low (September 2015) flow periods. A total of 53 sites distributed all over the Marana-Casinca alluvial plain were collected, as follow: 27 wells, 11 boreholes, 5 piezometers, 3 waters from the Bevincu River, 4 sampling sites from the lagoon and 3 sampling sites in artificial drains near the lagoon.

Depending on the piezometric level and the site's characteristics, groundwater was collected with a Grundfos MP1 or a Comet submersible pump. Some groundwater was directly sampled at the tap, avoiding air contamination. Before collecting water samples, groundwater was purged until stabilization of physico-chemical parameters (electrical conductivity, EC, temperature T, pH and dissolved O_2). Physico-chemical parameters were measured *in situ* using a WTW 3310 Conductivity meter, a WTW 3310 pH meter and a WTW 3310 IDS Oximeter (WTW GmbH, Weilheim, Germany). Alkalinity was determined in the field by volumetric titration, using a digital titrator HACH (Hach Company, Loveland, CO, U.S.A).

Water samples for major cation and anion analysis (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-}) were filtered on site through 0.45 µm nitrocellulose membranes, collected in two pre-cleaned 50 mL polyethylene bottles and stored at 4 °C. Major elements analyses were carried out using a Dionex ICS 1100 chromatograph (Thermo Dionex, Sunnyvale, CA, U.S.A.), at the Hydrogeology Department (CNRS UMR 6134 SPE), University of Corsica, France. The quality of the analysis was checked by calculating the ionic mass balance (the ionic balance error was < 5%). Water samples for trace element analysis were filtered through 0.20 µm nitrocellulose membranes, acidified using ultrapure HNO_3, and collected in 50 mL polyethylene bottles before storage at 4 °C. Trace element analyses were carried out at the AETE technical platform, University of Montpellier, France, using a Q-ICPMS X series II Thermo Fisher (Thermo Fisher Scientific, Bremen, Germany), with an analytical precision better than 8%.

3.2. Stable isotopes of the water molecule

Samples for the analysis of the stable isotopes of the water molecule (δ^2H and $\delta^{18}O$) were collected without head-space in pre-cleaned 20 mL amber glass bottles. $\delta^{18}O$ and δ^2H were determined by a laser-based liquid–vapor stable isotope analyzer DLT-100 (Los Gatos Research, San Jose, CA, U.S.A.) at the Hydrogeology Department (CNRS UMR 6134 SPE), University of Corsica, France. Analyses were realized according to the analytical scheme recommended by the International Atomic Energy Agency (IAEA; Penna et al., 2010). δ^2H and $\delta^{18}O$ are reported in per mil (‰) relative to the VSMOW (Vienna Standard Mean Ocean Water) reference material. The analytical precision was better than 0.6‰ for δ^2H and 0.2‰ for $\delta^{18}O$.

3.3. Tritium

Waters collected for tritium (^3H) analyses were sampled in pre-cleaned 500 mL polyethylene bottles. Tritium analyses were carried out by liquid scintillation counting (Thatcher et al., 1977) after electrolytic enrichment (Kaufman and Libby, 1954) at the Hydrogeology Department of the University of Avignon, France. Tritium concentration is expressed in TU (tritium units).

3.4. Emerging compounds

During an additional field campaign (May 2017), a total of 18 sites were collected: 5 wells, 4 boreholes, 1 piezometer, 1 sampling site from the Bevincu River, 3 sampling sites in the lagoon water, 2 samples of untreated wastewater from water treatment plants and 2 samples from sewage lift stations. Replicated water samples (n = 2) were taken in pre-cleaned 60 mL amber glass vials and immediately frozen at −18 °C. In total, 51 organic compounds (Table 1) were analyzed, including pharmaceuticals and other substances, such as artificial sweeteners, caffeine and so forth (Table 1). Samples were defrosted at max. 30 °C and immediately analyzed. Analysis were done using positive (ESI+) and negative (ESI-) modes of the electrospray at the "Povodi Vltavy" laboratory, Pilsen, Czech Republic. Each sample was centrifuged in headspace vials at about 3500 rpm during 5 min. Formic and acetic acids were used for the preparation of sample solution for ESI+ and ESI− respectively. Deuterized internal standards of d10-carbamazepine, d6-sulfamethoxazole, d3-iopromide and 13C2-erythomycin (ESI+), or d3-ibuprofen, d4-diclofenac, d3-naproxen, d5-chloramphenicol and d3-iopamidol (ESI−) were used for compounds quantification. Samples were directly injected into the A 1200 Ultra High-Performance Liquid Chromatograph (UHPLC) tandem with 6410 Triple Quad Mass Spectrophotometer (MS/MS) with ESI+ and ESI− modes of Agilent Technologies (Agilent, Santa Clara, CA, U.S.A.) following the LC–MS/MS analytical method. For ESI+ and ESI−, the separation was performed on a Zorbax Eclipse XDB-C18 analytical column (100 × 4.6 mm, 3.5 m particle size). For ESI+, mobile phase consisted of methanol and water with 0.1% formic acid and 5 mM ammonium formate as additives. The flow rate was 0.25 mL/min and injection volume was 0.50 mL. For ESI−, mobile phase consisted of methanol and water with 0.05% acetic acid as mobile phase additive. The flow rate was 0.25 mL/min and injection volume was 1 mL.

3.5. Aquifer recharge rate estimation

The alluvial aquifer recharge rates were computed using the ESPERE_v1.5 multiple-method Excel sheet application (Lanini et al., 2016). The empirical methods of Turc and the Soil Water Balance (SWB) method initially proposed by Thornthwaite (1948) and improved by Dingman (2002) were applied. Daily rainfall (mm), temperature (°C) and potential evapotranspiration (PET, mm) provided by Météo-France rainfall station of Bastia for the 01/1957–12/2016 period were used.

The rainfall composition used to calculate the ionic concentrations supplied by rainfall to groundwater was approximated by the rainfall composition measured in Bonifacio (nearest station with data along the East coast of Corsica), from 01/2013 to 12/2014 (Santoni et al., 2018).

3.6. End-member mixing analysis

A ternary mixing model based on geochemical and isotopic composition of groundwater was used to constrain the participation of the different water bodies. Calculation was carried out using Microsoft Excel and also checked using the MIX2 Program (Vázquez-Suñé et al. 2010). Differences between the two approaches were less than ± 10%.

4. Results and discussion

4.1. Aquifer recharge rate estimation

The mean annual alluvial aquifer recharge rate is about 127 mm which corresponds to 16% of the 777 mm of mean annual rainfall (01/1957–12/2016 period, Table 2). This result is consistent with the previously estimated infiltration rates from 9% to 26% for alluvial aquifers in the Mediterranean area (Candela et al., 2009; Ghiglieri et al., 2012; Stigter et al., 2014). The annual recharge rates display an important inter-annual variability (Table 2), consistent with the Western Mediterranean context (Scozzafava and Tallini, 2001; Pulido-Velazquez et al., 2017; Santoni et al., 2018). The rainfall amount, particularly variable from one year to another, induces an extremely variable annual aquifer recharge rate, ranging from 14 to 427 mm/y, which represent 3 to 31% of the total annual rainfall, respectively (Table 2). The values of estimated annual aquifer recharge rate also vary from one method to another. Both Turc and SWB methods improved by Dingman provided similar results with mean inter-annual recharge rates between 128 mm and 143 mm. The initial SWB proposed by Thornwaite seems to underestimate the recharge rate (98 mm). This method, which requires fewer parameters than the SWB method improved by Dingman and does not take into account snowfall, seems less appropriated. Thereby, combining several methods is consequently the best approach to calculate realistic aquifer recharge rate estimations (Scanlon et al., 2006; Santoni et al., 2018).

4.2. Groundwater origin

The relationships between δ^2H and $\delta^{18}O$ in groundwater, river, lagoon and sea water samples are shown in Fig. 3. The isotopic composition of groundwater ranges from −9.33‰ to + 3.38‰ for $\delta^{18}O$ and from −62.5‰ to + 6.4‰ for δ^2H (Table 3 and 4). Most of the groundwater samples are plotted between the Global Meteoric Water Line (GMWL; Craig, 1961) and the Western Mediterranean Meteoric Water Line (WMMWL; Celle-Jeanton et al., 2001). The local meteoric water line for the Bastia precipitation (BMWL: $\delta^2H = 7.42\ \delta^{18}O + 9.15$, $r^2 = 0.95$) was also defined using an ordinary least squares regression (OLSR) (IAEA, 1992) for the 12/2013–04/2017 period. This water line was proposed considering only non-evaporated samples to exclude the effects of sub-cloud evaporation (Froehlich et al., 2008; Santoni et al., 2018). The proposed BMWL is consistent with the WMMWL, the local meteoric water line calculated for Bonifacio, France ($\delta^2H = 7.39\ \delta^{18}O + 6.03$, Santoni et al,. 2018), Montpellier, France ($\delta^2H = 7.4\ \delta^{18}O + 7.3$, Ladouche et al., 2009) and Sicily, Italy ($\delta^2H = 6.75\ \delta^{18}O + 8.2$, Liotta et al. 2013).

Groundwater from the northern part of the study area is plotted on the BMWL, characterized by $\delta^{18}O$ between −7.83‰ and −5.75‰ and by δ^2H between −46.6‰ and −34.8‰ (Fig. 3). These depleted signatures, close to the annual weighted means of isotopes in rainwater collected in Corte, central Corsica (450 masl, −7.3‰ for δ^{18} and −48‰ for δ^2H) suggest a recharge from high-altitude precipitation (Huneau et al., 2015). Thus, groundwater from the northern part of the alluvial aquifer benefits from an allochthonous recharge from the headwater catchment, essentially formed by lustrous schists.

Groundwater from the southern part of the study area and the sand bar is more enriched in ^{18}O and ^2H with values ranging from −7.58‰ to + 3.38‰ for $\delta^{18}O$ and from −46.8‰ to + 6.4‰ for δ^2H (Fig. 3). Isotopic signatures are plotted along the BMWL, close to the annual weighted mean of isotopes in rainwater collected from the Bastia station (1 masl, −6.1‰ f or $\delta^{18}O$ and −36‰ f or δ^2H) (Huneau et al., 2015). The good correlation between the isotopic values of groundwater and the precipitation collected at Bastia rainfall station (Fig. 3) is in favor of a similar meteoric origin and then highlights an autochthonous recharge.

Groundwater from the Golu River alluvium (A33, Fig. 1) and from the Bevincu River (R1 to R5) are more depleted in ^{18}O (from −9.33‰ to −9.13‰ and from −8.81‰ to −7.44‰, respectively) and ^2H (from −62.0‰ to −59.0‰ and from −55.9‰ to −43.9‰, respectively) (Fig. 3). This very depleted signature highlights high-altitude water origin, from the inner Corsica mountainous headwater catchment, where the sources of the rivers are located.

The isotopic composition of groundwater is clearly different from that of the seawater composition (8.6% for δ^2H and 1.1% for $\delta^{18}O$). The groundwater samples A28 (April and September samples), A29 and A30 are plotted along the evaporation line. The three piezometers are located in a quarry for aggregate building material. The extraction leads to a rise in the water table and allows the formation of several small ponds that are subject to evaporation processes. Groundwater is therefore locally influenced by the re-infiltration of this surface water. The isotopic signatures of the lagoon water (Fig. 3) indicate mixing processes with seawater. The enriched isotopic signatures, slightly below the seawater-freshwater mixing line, also highlight the evaporation processes affecting the lagoon water (Gemitzi et al., 2014).

4.3. Groundwater quality characterization

Groundwater temperatures are around 16.7 °C (Tables 3 and 4), close to the mean annual air temperature in Bastia's region (15.8 °C).

Table 3
Field parameters, ionic composition, saturation indices and stable isotopes for April 2015 sampling campaign.

ID	Type	Dept (m)	Localisation (Lambert 93)		T (°C)	pH	EC (µS/cm)	O_2 (mg/L)	HCO_3^- (mg/L)	Cl^- (mg/L)	SO_4^{2-} (mg/L)	NO_3^- (mg/L)
Sc1	B	-	1226702.5	6195517.8	17.7	7.1	891	6.3	332	70.0	46.2	12.0
Sc2	W	70.0	1227207.1	6194282.7	16.6	7.3	904	0.4	467	52.3	42.7	0.0
Sc3	B	90.0	1227113.3	6192715.3	19.4	7.4	837	3.1	421	39.3	36.3	4.5
A1	B	17.5	1228441.0	6194233.9	15.7	7.3	427	8.9	188	26.9	20.2	4.5
A2	B	20.0	1228417.7	6194195.0	17.3	7.1	474	8.5	223	28.8	25.3	11.0
A3	B	25.0	1228534.3	6194111.0	16.7	7.5	483	6.4	200	32.3	16.6	3.8
A4	W	3.9	1228683.9	6191587.2	14.0	6.4	505	2.2	145	55.7	44.2	11.2
A5	W	9.0	1229394.4	6191131.5	16.9	7.2	1285	4.4	215	254.6	65.5	7.1
A6	W	3.3	1228962.2	6190855.4	15.1	7.6	937	0.2	494	70.1	4.2	1.4
A7	W	2.7	1229624.9	6190946.2	13.3	7.2	650	1.5	306	46.3	34.3	2.5
A8	W	4.7	1229383.0	6190670.1	16.5	7.2	467	0.6	234	21.2	24.7	2.7
A9	W	7.1	1228986.8	6190557.3	17.2	6.9	666	2.6	295	32.5	36.6	21.8
A10	W	10.4	1228786.8	6190465.1	16.6	6.7	561	–	250	31.9	26.7	15.0
A11	W	–	1228907.9	6190418.8	16.9	6.9	462	4.0	220	20.3	15.5	9.3
A12	W	–	1229351.8	6190384.1	7.5	6.9	433	0.5	227	19.0	18.4	5.2
A13	W	19.5	1229763.0	6190274.8	16.2	7.3	403	3.0	188	20.9	18.2	6.2
A14	W	15.8	1229674.3	6190263.0	16.0	7.3	391	1.8	194	18.3	17.2	4.0
A15	W	18.8	1229711.2	6190174.0	15.8	7.2	394	6.6	209	18.6	18.7	5.3
A16	W	16.0	1229708.7	6190114.4	15.9	7.2	403	4.4	194	19.3	20.1	6.3
A17	W	13.3	1229714.3	6190067.8	15.9	7.2	405	4.6	206	19.6	19.3	6.2
A18	W	13.7	1229725.9	6189979.6	16.2	7.1	452	4.0	210	22.6	27.0	6.1
A19	W	8.8	1229439.3	6189257.1	15.5	6.9	753	2.0	303	47.1	27.9	39.8
A20	W	7.2	1229738.9	6188101.9	12.9	7.2	283	7.3	143	13.9	10.2	1.2
A21	W	6.2	1229156.1	6188083.4	13.6	6.5	825	0.9	328	31.6	114.4	0.5
A22	W	8.2	1230473.1	6186524.3	16.6	6.9	505	4.1	217	25.2	35.3	20.0
A23	B	23.0	1231088.1	6186248.4	18.0	6.7	394	6.7	195	25.6	8.7	11.0
A24	W	8.6	1231209.0	6185141.0	15.1	7.1	743	5.2	237	47.9	34.9	79.7
A25	P	11.7	1233242.0	6183809.4	15.3	6.5	373	8.1	105	26.2	31.3	52.0
A26	P	11.9	1233806.5	6183732.8	16.6	6.2	395	5.9	118	21.1	28.1	37.3
A27	P	11.4	1234314.6	6183764.6	15.3	6.7	341	0.6	154	29.4	21.2	8.0
A28	P	10.5	1233019.8	6183382.9	15.0	6.2	223	5.5	73	16.4	10.1	4.5
A29	P	12.3	1232835.4	6183243.4	15.4	7.0	929	0.5	86	184.9	32.6	3.0
A30	W	6.0	1229490.7	6182415.9	15.7	7.5	604	6.9	273	28.9	39.0	13.8
A31	W	11.6	1234222.8	6182733.9	13.8	6.7	844	–	207	123.1	50.5	19.2
A32	W	5.8	1233534.1	6182301.1	15.0	6.7	271	3.7	110	20.3	11.4	9.6
A33	W	–	1232652.8	6181163.4	12.3	6.9	243	4.3	118	8.9	8.5	6.4
S1	B	2.2	1229687.0	6193228.8	16.0	7.3	1755	1.2	340	355.5	67.7	71.9
S2	B	1.8	1230262.4	6192393.1	16.1	7.6	3080	7.0	210	798.0	114.8	10.6
S3	B	1.9	1230919.2	6191414.1	17.6	7.1	5860	0.5	189	1787.1	235.5	33.1
S4	B	1.9	1231824.2	6190419.0	16.6	7.1	1073	0.6	310	107.2	60.9	76.2
S5	W	2.4	1231472.9	6189659.8	14.0	7.0	492	9.4	149	54.4	21.8	2.5
S6	W	2.0	1236000.9	6184646.9	19.8	7.1	821	2.5	326	57.4	46.6	23.0
S7	B	5.7	1236312.3	6183110.9	16.5	7.6	523	4.4	216	32.3	44.2	1.4
R1	R	–	1228766.8	6190080.5	14.7	8.6	294	11.3	154	13.6	9.2	0.3
R2	R	–	1227832.5	6189124.1	14.8	8.4	296	10.2	143	13.5	9.1	0.0
R3	R	–	1222408.4	6186849.4	13.0	8.9	273	10.6	143	11.3	7.9	0.0
R4	R	–	1222692.9	6185483.7	10.4	7.5	198	10.4	95	8.9	6.3	0.6
R5	R	–	1217847.1	6184080.1	12.9	7.9	202	9.9	100	8.5	5.6	0.0

(*continued on next page*)

Table 3 (*continued*)

ID	Type	Dept (m)	Localisation (Lambert 93)		T (°C)	pH	EC (μS/cm)	O_2 (mg/L)	HCO_3^- (mg/L)	Cl^- (mg/L)	SO_4^{2-} (mg/L)	NO_3^- (mg/L)
D1	D	–	1235844.1	6182621.9	15.1	8.2	17,520	11	89	6447	916	0
L1	L	–	1228788.2	6194529.8	17.5	8.2	56,100	10	154	24,827	3327	0
L2	L	–	1229112.5	6192407.6	18.1	8.3	48,800	9	161	19,777	2617	0
L3	L	–	1231845.6	6186731.0	19.7	8.1	15,550	7	154	5486	794	0
L4	L	–	1235023.8	6184716.8	14.8	8.0	14,270	5	107	5036	649	0

ID	Ca^{2+} (mg/L)	Na^+ (mg/L)	Mg^{2+} (mg/L)	K^+ (mg/L)	SiO_2 (mg/L)	SI calcite	SI dolomite	SI quartz	SI chalcedony	δ^2H (‰)	$\delta^{18}O$ (‰)	^3H (TU)
Sc1	96.6	44.6	26.1	1.9	15.0	−0.04	−0.39	0.49	0.04	−40.0 ± 0.2	−6.98 ± 0.06	-
Sc2	61.0	66.1	51.4	2.1	31.3	0.06	0.28	0.83	0.37	−40.0 ± 0.2	−6.88 ± 0.02	–
Sc3	45.6	73.7	36.9	3.1	21.9	0.08	0.35	0.63	0.18	−41.0 ± 0.5	−7.06 ± 0.02	1.7 ± 0.1
A1	54.1	17.8	10.2	1.3	10.2	−0.24	−0.99	0.35	−0.11	−43.2 ± 0.1	−7.40 ± 0.01	–
A2	57.7	22.0	11.9	1.4	12.4	−0.38	−1.19	0.41	−0.04	−42.5 ± 0.3	−7.30 ± 0.11	3.1 ± 0.4
A3	60.8	20.0	10.9	1.2	10.1	0.05	−0.40	0.33	−0.12	−41.0 ± 0.5	−7.25 ± 0.12	–
A4	31.0	33.0	20.8	1.2	21.3	−1.60	−3.18	0.70	0.23	−39.2 ± 0.2	−6.65 ± 0.05	–
A5	96.5	71.9	57.1	1.7	17.0	−0.15	−0.28	0.56	0.10	−44.8 ± 0.2	−7.45 ± 0.09	–
A6	48.2	89.6	40.1	1.7	23.5	0.33	0.80	0.72	0.26	−35.8 ± 0.3	−5.86 ± 0.03	–
A7	50.3	20.0	37.4	1.9	16.3	−0.25	−0.45	0.59	0.13	−40.4 ± 0.2	−6.46 ± 0.04	3.3 ± 0.2
A8	51.3	14.1	27.4	0.6	18.6	−0.34	−0.72	0.60	0.14	−45.1 ± 0.2	−7.66 ± 0.03	3.1 ± 0.3
A9	45.0	30.1	40.5	0.5	27.8	−0.55	−0.90	0.76	0.31	−41.3 ± 0.4	−6.92 ± 0.04	–
A10	42.0	24.7	31.2	0.4	27.5	−0.85	−1.60	0.77	0.31	−41.0 ± 0.1	−6.75 ± 0.05	5.2 ± 0.3
A11	39.8	15.4	27.6	0.4	20.0	−0.71	−1.34	0.63	0.17	−44.1 ± 0.2	−7.38 ± 0.03	3.1 ± 0.3
A12	42.0	12.1	25.7	0.6	19.3	−0.87	−1.88	0.76	0.27	−45.1 ± 0.2	−7.56 ± 0.06	–
A13	34.1	13.2	23.0	0.7	17.3	−0.48	−0.90	0.57	0.12	−46.0 ± 0.1	−7.67 ± 0.04	–
A14	35.3	11.3	21.9	0.5	16.7	−0.43	−0.85	0.56	0.10	−45.7 ± 0.2	−7.67 ± 0.05	–
A15	35.8	11.6	21.6	0.5	15.5	−0.47	−0.94	0.53	0.07	−40.4 ± 0.2	−6.87 ± 0.06	–
A16	36.4	12.1	22.1	0.5	15.4	−0.54	−1.08	0.53	0.07	−45.4 ± 0.3	−7.62 ± 0.01	–
A17	36.8	12.1	22.3	0.5	15.6	−0.51	−1.01	0.53	0.07	−46.1 ± 0.1	−7.68 ± 0.04	–
A18	40.2	13.9	25.8	0.6	18.0	−0.53	−1.03	0.59	0.13	−45.2 ± 0.2	−7.55 ± 0.03	3.2 ± 0.3
A19	37.8	29.6	48.8	11.3	40.2	−0.70	−1.06	0.95	0.49	−40.2 ± 0.2	−6.90 ± 0.08	2.8 ± 0.2
A20	29.7	8.8	11.2	0.8	12.1	−0.76	−1.77	0.47	0.00	−44.4 ± 0.3	−7.57 ± 0.04	–
A21	66.8	22.0	49.0	2.8	18.8	−0.82	−1.59	0.65	0.19	−43.5 ± 0.3	−6.93 ± 0.10	3.5 ± 0.4
A22	48.5	23.5	17.1	5.6	22.7	−0.64	−1.51	0.69	0.23	−41.4 ± 0.2	−6.86 ± 0.08	3.8 ± 0.4
A23	27.1	18.9	24.0	1.9	30.8	−1.11	−2.02	0.80	0.35	−45.5 ± 0.2	−7.58 ± 0.01	1.2 ± 0.3
A24	67.8	26.4	25.7	28.0	22.2	−0.35	−0.92	0.70	0.24	−41.6 ± 0.1	−6.82 ± 0.07	–
A25	15.9	14.2	21.4	9.4	17.1	−1.82	−3.29	0.58	0.12	−38.3 ± 0.4	−6.32 ± 0.07	–
A26	11.5	17.1	28.2	3.2	12.4	−2.18	−3.73	0.42	−0.03	−37.1 ± 0.2	−5.99 ± 0.02	–
A27	11.2	16.0	25.5	1.8	18.3	−1.68	−2.78	0.61	0.15	−28.4 ± 0.2	−4.18 ± 0.06	–
A28	8.1	11.2	15.6	0.7	19.7	−2.56	−4.63	0.65	0.19	−37.9 ± 0.1	−6.17 ± 0.06	–
A29	48.9	45.5	43.7	2.7	17.8	−0.95	−1.74	0.60	0.14	−40.9 ± 0.4	−6.52 ± 0.07	–
A30	54.1	28.7	24.5	5.6	17.6	0.04	−0.04	0.59	0.13	−40.4 ± 0.1	−6.70 ± 0.05	3.7 ± 0.3
A31	63.9	37.0	42.4	4.0	18.0	−0.85	−1.70	0.63	0.16	−38.9 ± 0.2	−6.40 ± 0.15	–
A32	20.7	10.3	11.6	3.7	12.2	−1.52	−3.08	0.44	−0.02	−40.5 ± 0.1	−6.55 ± 0.03	3.6 ± 0.3
A33	31.0	7.1	6.9	0.9	8.6	−1.12	−2.74	0.33	−0.14	−62.0 ± 0.5	−9.33 ± 0.08	4.2 ± 0.4
S1	115.8	167.9	44.6	14.0	12.6	0.15	0.11	0.44	−0.02	−46.1 ± 0.1	−7.72 ± 0.04	–
S2	70.5	447.0	70.0	16.5	12.4	−0.02	0.19	0.43	−0.02	−38.1 ± 0.2	−6.17 ± 0.05	–
S3	103.5	1005.7	156.3	54.9	15.1	−0.42	−0.41	0.50	0.05	−35.9 ± 0.1	−5.94 ± 0.07	–
S4	88.7	72.7	35.8	13.1	17.6	−0.16	−0.48	0.58	0.12	−39.3 ± 0.2	−6.56 ± 0.03	–
S5	22.5	52.7	9.0	11.5	20.2	−1.03	−2.28	0.68	0.21	−37.1 ± 0.3	−6.29 ± 0.02	–
S6	66.0	48.2	29.4	7.5	17.2	−0.16	−0.39	0.52	0.07	−37.4 ± 0.2	−6.13 ± 0.01	–
S7	51.6	25.0	13.0	5.4	15.7	0.05	−0.27	0.53	0.07	−35.9 ± 0.1	−6.03 ± 0.05	3.3 ± 0.3
R1	31.7	9.1	12.1	0.6	9.0	0.66	1.11	0.30	−0.17	−48.7 ± 0.2	−8.08 ± 0.03	–
R2	32.7	9.0	11.5	0.6	9.6	0.52	0.80	0.33	−0.14	−49.1 ± 0.3	−8.11 ± 0.03	–
R3	33.7	7.8	9.4	0.5	8.9	0.89	1.42	0.31	−0.16	−51.6 ± 0.2	−8.40 ± 0.01	–

(*continued on next page*)

Table 3 (*continued*)

ID	Ca^{2+} (mg/L)	Na^{+} (mg/L)	Mg^{2+} (mg/L)	K^{+} (mg/L)	SiO_2 (mg/L)	SI calcite	SI dolomite	SI quartz	SI chalcedony	δ^2H (‰)	$\delta^{18}O$ (‰)	^3H (TU)
R4	17.4	6.5	10.1	0.6	12.7	−0.87	−1.86	0.53	0.05	−49.6 ± 0.2	−8.34 ± 0.06	–
R5	20.8	7.3	7.9	0.4	11.3	−0.32	−0.88	0.44	−0.03	−55.9 ± 0.3	−8.81 ± 0.07	–
D1	157	3690	447	135	5	0.24	1.16	0.04	−0.42	−48.5 ± 0.4	−7.33 ± 0.12	–
L1	511	13,622	1632	503	0	0.75	2.31	−0.88	−1.33	6.9 ± 0.2	0.76 ± 0.03	–
L2	430	10,940	1309	403	3	0.87	2.52	−0.21	−0.66	−1.6 ± 0.6	−0.52 ± 0.01	–
L3	168	3130	397	111	0	0.48	1.63	−2.01	−2.45	−25.1 ± 0.3	−3.79 ± 0.03	–
L4	143	2861	361	102	6	0.09	0.80	0.17	−0.29	−46.5 ± 0.4	−7.06 ± 0.09	–

B: boreholes, D: drain near lagoon, L: lagoon, P: piezometer, R: river; W: well

The lowest temperatures and EC are mainly observed in groundwater close to the rivers (the Bevincu River, the Golu River and the Pietre Turchine River).

Groundwater from the schist formations displays alkaline pH (from 7.1 to 7.5) and high EC (mean of 884 µS/cm) (Tables 3 and 4). The Ca^{2+}–HCO_3^- water type tends towards a Na^+-K^+ water type, indicating important interactions with the calcareous-schist formations (Fig. 4).

Groundwater from the northern part is characterized by slightly alkaline pH (mean of 7.2) and relatively high EC mean of 536 µS/cm (Tables 3 and 4). The groundwater mineralization is predominantly controlled by high concentrations in calcium, magnesium and bicarbonate, giving a HCO_3^--Ca^{2+}-Mg^{2+} water type (Fig. 4). Their geochemical compositions are generally plotted between the groundwater compositions measured in schist formations and river waters, suggesting likewise important mixing processes within the alluvial aquifer (Fig. 5). NO_3^- concentrations are generally low (mean < 7 mg/L) (Tables 3 and 4), which indicates a limited anthropogenic impact.

Groundwater from the southern part is slightly acidic (mean pH around 6.9) (Tables 3 and 4). EC is lower (mean of 497 µS/cm) and its geochemical composition is closer to the river water composition (Fig. 5). The higher concentrations in Cl^- and SO_4^{2-} associated with NO_3^- concentrations above the natural baseline (between 5 and 7 mg/L) (Appelo and Postma, 2005; Santoni et al., 2016a,b) indicate an anthropogenic impact (Fig. 4). Finally, groundwater from the sand bar and lagoon waters display the highest EC values (mean of 2355 µS/cm and 35,065 µS/cm respectively) and high Cl^--Na^+-K^+ concentrations (Tables 3 and 4) suggesting seawater mixing influences. S1, S3 and S6, located on the sand bar, display significantly high concentrations for most measured metallic trace elements (MTE), especially Li and B (Table 5) suggesting coastal water influence. Yet, the high concentrations in Mn, Fe, Zn and As up to 220,56 µg/L, 181,88 µg/L, 18,93 µg/L and 19,55 µg/L, respectively also suggest the impact of anthropogenic pollution infiltration (Table 5).

4.4. Residence time and water–rock interaction

Tritium concentrations range from 1.2 to 4.2 TU, with a mean value of 3.1 TU (Tables 3 and 4). The local ^3H content of rain water in about 3.8 TU (Erostate et al., 2018; IAEA, WMO, 2019). Groundwater from schist formations (Sc1) and A23 presents the lowest ^3H concentrations with a mean of 1.9 TU and 1.3 TU respectively. These low concentrations argue in favour of groundwater with relatively long residence time. The sample from the schist formations (Sc1) indicates the long residence times for water resources from the relatively impervious sediments, with minimal connectivity to surface water. For the sample point A23, a previous study has already highlighted that this deep well (23 m) benefits from a low contribution from recently infiltrated water (Erostate et al., 2018), which explains the low ^3H concentration.

The highest ^3H concentrations, 5.2 and 4.2 TU, are measured in A10 and A33, respectively, localized close to the Bevincu River and the Golu River respectively. These high ^3H contents (Tables 3 and 4) and geochemical composition similar to river water composition (Fig. 5) argue in favor of a significant recharge by recently infiltrated water, brought by the river. Finally, groundwater from the southern part displays ^3H concentrations (median value of 3.4 TU) higher than those of groundwater from the northern part (median value of 2.3 TU). Therefore, groundwater from the southern part presents shorter residence times, suggesting a greater contribution of recently infiltrated water to the recharge of the alluvial aquifer.

Chemical composition (Fig. 5) and saturation indices (Tables 3 and 4) reflect the lithological control on groundwater. All groundwater samples are saturated with respect to quartz and/or chalcedony. The water rock interaction processes involving the schist formations as well as the schistous and granitic nature of alluvial deposits favor the dissolution of chalcedony and quartz minerals. Groundwater samples are generally undersaturated with respect to carbonate minerals. Only the

Table 4
Field parameters, ionic composition, saturation indices and stable isotopes for September 2015 sampling campaign.

ID	Type	Depth (m)	Localisation (Lambert 93)		T (°C)	pH	EC (µS/cm)	O_2 (mg/L)	HCO_3^- (mg/L)	Cl^- (mg/L)	SO_4^{2-} (mg/L)	NO_3^- (mg/L)
Sc1	B	–	1226702.5	6195517.8	18.4	7.2	923	4.9	316	81.8	53.5	12.8
Sc2	W	70.0	1227207.1	6194282.7	18.8	7.3	927	2.7	461	52.3	45.8	0.2
Sc3	B	90.0	1227113.3	6192715.3	19.4	7.5	822	0.2	398	39.3	42.8	0.7
A1	B	17.5	1228441.0	6194233.9	15.6	7.2	534	8.0	204	32.1	26.6	10.9
A2	B	20.0	1228417.7	6194195.0	18.1	7.0	518	8.2	210	32.8	31.7	19.5
A3	B	25.0	1228534.3	6194111.0	–	–	–	–	–	–	–	–
A4	W	3.9	1228683.9	6191587.2	17.8	6.7	564	0.2	212	46.1	24.8	0.4
A5	W	9.0	1229394.4	6191131.5	18.6	7.3	681	0.1	327	32.5	27.7	4.4
A6	W	3.3	1228962.2	6190855.4	19.2	7.7	1038	0.1	506	79.9	6.0	0.2
A7	W	2.7	1229624.9	6190946.2	–	–	–	–	–	–	–	–
A8	W	4.7	1229383.0	6190670.1	16.8	7.3	465	0.1	222	21.1	24.5	1.7
A9	W	7.1	1228986.8	6190557.3	18.8	7.4	531	1.6	262	35.1	25.1	5.7
A10	W	10.4	1228786.8	6190465.1	18.6	7.2	584	1.8	248	24.8	20.7	6.9
A11	W	–	1228907.9	6190418.8	19.8	7.2	403	1.5	217	20.5	14.4	5.0
A12	W	–	1229351.8	6190384.1	16.6	7.2	423	0.5	193	20.4	17.8	4.2
A13	W	19.5	1229763.0	6190274.8	16.2	7.6	382	1.9	193	18.5	15.1	1.8
A14	W	15.8	1229674.3	6190263.0	16.5	7.5	387	1.3	175	18.5	15.8	2.0
A15	W	18.8	1229711.2	6190174.0	16.6	7.7	376	2.2	182	18.0	13.2	0.8
A16	W	16.0	1229708.7	6190114.4	16.4	7.6	378	2.8	178	17.4	13.8	1.0
A17	W	13.3	1229714.3	6190067.8	16.3	7.5	381	2.7	181	19.3	14.1	1.2
A18	W	13.7	1229725.9	6189979.6	16.6	7.6	441	5.3	218	23.1	21.1	2.4
A19	W	8.8	1229439.3	6189257.1	–	–	–	–	–	–	–	–
A20	W	7.2	1229738.9	6188101.9	16.6	6.8	373	5.8	212	14.2	12.6	3.9
A21	W	6.2	1229156.1	6188083.4	15.4	6.6	926	0.2	364	33.9	169.8	0.3
A22	W	8.2	1230473.1	6186524.3	17.1	7.4	567	0.3	254	23.0	30.8	4.5
A23	B	23.0	1231088.1	6186248.4	18.2	6.8	393	6.2	162	26.2	14.4	14.6
A24	W	8.6	1231209.0	6185141.0	17.1	6.7	432	–	–	29.2	19.6	34.0
A25	P	11.7	1233242.0	6183809.4	20.1	6.9	397	2.9	127	35.2	23.8	14.4
A26	P	11.9	1233806.5	6183732.8	18.7	6.9	368	3.2	112	22.2	27.6	26.3
A27	P	11.4	1234314.6	6183764.6	22.7	7.4	930	0.7	143	204.4	32.0	5.1
A28	P	10.5	1233019.8	6183382.9	21.9	6.9	436	2.1	96	42.2	80.9	2.4
A29	P	12.3	1232835.4	6183243.4	20.2	6.8	485	0.4	90	47.9	99.0	0.3
A30	W	6.0	1229490.7	6182415.9	17.3	7.4	620	1.7	295	26.3	34.3	16.0
A31	W	11.6	1234222.8	6182733.9	19.9	7.1	381	0.3	123	36.9	15.3	21.7
A32	W	5.8	1233534.1	6182301.1	20.3	6.6	410	4.4	94	37.0	16.0	54.8
A33	W	–	1232652.8	6181163.4	19.2	7.1	224	1.7	104	9.9	7.3	5.0
S1	B	2.2	1229687.0	6193228.8	19.8	7.6	4370	1.5	253	1382.2	186.4	1.1
S2	B	1.8	1230262.4	6192393.1	21.1	7.8	5040	5.9	289	1420.4	203.1	6.9
S3	B	1.9	1230919.2	6191414.1	21.7	7.2	7490	0.1	277	2212.1	300.1	29.7
S4	B	1.9	1231824.2	6190419.0	21.3	7.4	849	1.0	278	86.3	46.5	39.6
S5	W	2.4	1231472.9	6189659.8	17.8	7.1	566	0.4	81	87.0	32.8	14.6
S6	W	2.0	1236000.9	6184646.9	21.1	7.8	503	5.5	216	33.6	24.2	2.1
S7	B	5.7	1236312.3	6183110.9	21.6	7.9	546	4.2	216	35.0	40.2	2.1
R1	R	–	1228766.8	6190080.5	19.4	7.9	360	7.5	181	17.7	11.3	0.2
R2	R	–	1227832.5	6189124.1	–	–	–	–	–	–	–	–
R3	R	–	1222408.4	6186849.4	15.4	8.5	360	10.1	188	15.2	10.3	1.2
R4	R	–	1222692.9	6185483.7	14.2	8.1	304	9.6	161	12.4	8.2	0.6
R5	R	–	1217847.1	6184080.1	15.5	8.0	346	8.6	173	14.7	10.3	0.2

(continued on next page)

Table 4 (*continued*)

ID	Type	Depth (m)	Localisation (Lambert 93)		T (°C)	pH	EC (µS/cm)	O_2 (mg/L)	HCO_3^- (mg/L)	Cl^- (mg/L)	SO_4^{2-} (mg/L)	NO_3^- (mg/L)
D1	D	–	1235844.1	6182621.9	20.9	7.5	7910	5	104	1796	239	1
L1	L	–	1228788.2	6194529.8	16.3	8.0	49,100	–	149	18,585	2460	0
L2	L	–	1229112.5	6192407.6	14.9	7.9	40,400	–	159	9423	1233	0
L3	L	–	1231845.6	6186731.0	15.9	8.0	30,300	–	129	19,576	2546	0
L4	L	–	1235023.8	6184716.8	14.8	7.4	26,000	–	148	10,798	1427	0

ID	Ca^{2+} (mg/L)	Na^+ (mg/L)	Mg^{2+} (mg/L)	K^+ (mg/L)	SiO_2(mg/L)	SI calcite	SI dolomite	SI quartz	SI chalcedony	δ^2H (‰)	$\delta^{18}O$ (‰)	^3H (TU)
Sc1	104.0	50.8	28.2	1.9	14.0	0.06	−0.18	0.45	0.00	−40.2 ± 0.2	−6.91 ± 0.05	–
Sc2	62.8	63.9	54.2	2.1	26.2	0.17	0.55	0.71	0.26	−39.8 ± 0.2	−6.61 ± 0.09	–
Sc3	49.8	75.3	44.8	3.2	21.4	0.17	0.57	0.62	0.17	−41.3 ± 0.1	−7.23 ± 0.08	2.0 ± 0.3
A1	67.8	23.0	13.0	1.4	6.8	−0.26	−1.01	0.18	−0.28	−41.7 ± 0.2	−6.98 ± 0.04	–
A2	63.0	25.0	13.7	1.5	13.1	−0.45	−1.31	0.42	−0.03	−42.1 ± 0.8	−6.89 ± 0.06	3.6 ± 0.4
A3	–	–	–	–	–	–	–	–	–	–	–	–
A4	39.3	34.8	22.2	1.4	20.7	−0.98	−1.95	0.63	0.17	−41.9 ± 0.0	−6.76 ± 0.09	–
A5	52.3	27.9	47.5	1.2	20.4	−0.10	0.03	0.61	0.16	−41.3 ± 0.4	−7.17 ± 0.05	3.2 ± 0.4
A6	55.9	106.4	52.6	1.9	24.3	0.50	1.26	0.67	0.23	−34.8 ± 0.2	−5.75 ± 0.09	–
A7	–	–	–	–	–	–	–	–	–	–	–	–
A8	49.2	13.7	25.5	0.6	14.1	−0.25	−0.56	0.47	0.02	−45.7 ± 0.4	−7.57 ± 0.12	3.5 ± 0.5
A9	42.2	27.4	38.5	0.5	26.9	−0.14	−0.05	0.72	0.27	−40.6 ± 0.1	−6.89 ± 0.05	–
A10	38.0	21.6	37.3	0.3	25.0	−0.34	−0.42	0.70	0.25	−42.5 ± 0.3	−7.01 ± 0.07	5.2 ± 0.3
A11	36.9	13.5	24.9	0.4	18.3	−0.39	−0.68	0.54	0.10	−43.7 ± 0.7	−7.29 ± 0.08	2.8 ± 0.4
A12	39.8	11.9	25.5	0.6	15.3	−0.49	−0.95	0.51	0.06	−46.6 ± 0.6	−7.75 ± 0.05	–
A13	34.9	12.1	22.2	0.5	16.4	−0.15	−0.26	0.55	0.09	−46.6 ± 0.1	−7.74 ± 0.04	3.2 ± 0.4
A14	35.7	11.2	23.7	0.5	16.8	−0.22	−0.38	0.55	0.10	−46.5 ± 0.2	−7.54 ± 0.03	–
A15	34.7	11.2	21.5	0.5	14.7	−0.10	−0.18	0.50	0.04	−46.0 ± 0.1	−7.69 ± 0.06	–
A16	33.6	10.9	21.7	0.4	14.3	−0.15	−0.27	0.49	0.03	−45.5 ± 0.3	−7.63 ± 0.12	–
A17	35.0	11.7	22.4	0.5	15.7	−0.26	−0.49	0.53	0.07	−45.8 ± 0.2	−7.83 ± 0.07	–
A18	41.3	14.1	27.3	0.6	15.2	−0.06	−0.06	0.51	0.05	−46.1 ± 0.4	−7.60 ± 0.09	3.2 ± 0.3
A19	–	–	–	–	–	–	–	–	–	–	–	
A20	44.5	11.1	16.5	1.0	15.1	−0.73	−1.67	0.51	0.05	−40.5 ± 0.5	−6.96 ± 0.12	–
A21	84.8	22.8	68.1	3.4	19.7	−0.63	−1.14	0.64	0.18	−43.9 ± 0.1	−6.81 ± 0.05	3.6 ± 0.5
A22	65.3	24.5	22.7	4.1	20.4	0.02	−0.18	0.63	0.18	−39.4 ± 0.6	−6.84 ± 0.12	2.8 ± 0.2
A23	24.5	20.7	25.2	1.4	29.5	−1.16	−2.04	0.77	0.32	−39.6 ± 0.7	−6.84 ± 0.14	1.5 ± 0.3
A24	27.9	20.6	24.2	5.8	–	–	–	–	–	−39.6 ± 0.7	−6.73 ± 0.04	–
A25	15.2	17.4	29.6	1.4	21.9	−1.34	−2.11	0.62	0.17	−37.5 ± 0.8	−6.14 ± 0.11	–
A26	12.1	16.7	29.6	1.6	23.4	−1.48	−2.30	0.67	0.22	−38.7 ± 0.1	−6.52 ± 0.16	–
A27	57.9	43.9	53.9	2.8	17.8	−0.25	−0.22	0.49	0.05	–32.4 ± 0.2	−4.78 ± 0.10	–
A28	21.4	24.5	24.5	11.5	12.2	−1.29	−2.22	0.33	−0.10	−5.2 ± 0.3	0.83 ± 0.05	–
A29	23.4	32.3	26.3	2.0	13.7	−1.39	−2.44	0.41	−0.04	6.4 ± 0.5	3.38 ± 0.07	–
A30	53.7	32.5	30.5	4.4	15.8	0.02	0.03	0.52	0.06	−41.4 ± 0.1	−7.04 ± 0.05	3.3 ± 0.4
A31	19.4	17.3	21.9	2.2	19.2	−1.02	−1.70	0.56	0.12	−40.2 ± 0.1	−6.39 ± 0.08	–
A32	31.2	14.8	18.7	5.0	14.6	−1.46	−2.85	0.44	−0.01	−46.8 ± 0.3	−7.40 ± 0.04	3.1 ± 0.3
A33	28.1	7.3	6.6	0.8	10.1	−0.93	−2.22	0.29	−0.15	−59.0 ± 0.4	−9.13 ± 0.10	4.2 ± 0.6
S1	95.7	716.7	95.5	31.5	14.5	0.23	0.74	0.45	0.00	−42.2 ± 0.2	−6.80 ± 0.09	–
S2	107.1	787.2	99.5	24.5	17.1	0.51	1.29	0.50	0.05	–32.8 ± 0.1	−5.37 ± 0.03	–
S3	118.8	1125.4	189.8	73.2	18.0	−0.14	0.23	0.52	0.08	–32.5 ± 0.4	−5.38 ± 0.16	–
S4	61.0	71.2	31.5	11.2	19.3	0.04	0.09	0.54	0.10	−40.7 ± 0.6	−6.73 ± 0.10	–
S5	7.1	94.3	6.1	5.4	23.4	−1.69	−3.2	0.68	0.23	−36.8 ± 0.1	−6.32 ± 0.04	–
S6	43.0	31.4	20.7	4.8	16.2	0.29	0.56	0.46	0.02	−41.4 ± 0.6	−6.57 ± 0.07	–
S7	58.1	27.1	16.3	6.3	15.0	0.45	0.66	0.42	−0.02	−36.9 ± 0.3	−6.05 ± 0.05	3.2 ± 0.5
R1	37.2	11.1	17.0	0.5	11.5	0.19	0.31	0.34	−0.10	−43.9 ± 0.4	−7.44 ± 0.11	–
R2	–	–	–	–	–	–	–	–	–	–	–	–
R3	45.2	9.9	13.3	0.6	13.4	0.82	1.33	0.46	0.00	−48.3 ± 0.5	−8.19 ± 0.06	–

(*continued on next page*)

Table 4 (*continued*)

ID	Ca^{2+} (mg/L)	Na^{+} (mg/L)	Mg^{2+} (mg/L)	K^{+} (mg/L)	SiO_2(mg/L)	SI calcite	SI dolomite	SI quartz	SI chalcedony	δ^2H (‰)	$\delta^{18}O$ (‰)	^3H (TU)
R4	30.0	6.8	17.1	0.4	19.6	0.18	0.32	0.65	0.19	−49.5 ± 0.5	−8.34 ± 0.10	–
R5	40.1	10.7	14.1	0.3	12.9	0.25	0.27	0.45	−0.01	−48.6 ± 0.6	−7.84 ± 0.18	–
D1	57	961	127	35	6	−0.53	−0.41	0.07	−0.38	−50.5 ± 0.2	−7.29 ± 0.07	–
L1	363	9880	1278	356	2	0.46	1.75	−0.25	−0.71	0.6 ± 0.1	0.20 ± 0.06	–
L2	207	5066	614	176	8	0.21	1.12	0.29	−0.17	2.8 ± 0.3	0.48 ± 0.02	–
L3	387	10,437	1351	379	2	0.37	1.57	−0.36	−0.82	−11.4 ± 0.2	−1.11 ± 0.07	–
L4	228	5817	711	203	7	−0.25	0.23	0.22	−0.24	−15.3 ± 0.2	−1.73 ± 0.07	–

B: boreholes, D: drain near lagoon, L: lagoon, P: piezometer, R: river; W: well

saturation indices of groundwater from schist formations (Sc1, Sc2 and Sc3) approach saturation with respect to carbonate. Schist groundwater are enriched in HCO_3^-, Ca^{2+}, Na^+ and Mg^{2+} in accordance with the nature of the basement, mainly constituted by metamorphosed carbonates (Lahondère et al., 1994). The hydrolysis of silicate minerals constituting part of the schistous formations could also locally participate to the observed ionic contents. The long residence time of schist groundwater (1.9 TU) is in favor of longer interactions with the rock and allows dissolving more minerals leading to an equilibrium with respect to the carbonate minerals and an alkaline pH.

Groundwater from the northern part can be distinguished from that occurring in the southern part by a marked enrichment in HCO_3^-, Ca^{2+}, Na^+ and Mg^{2+} (Fig. 5) reflecting the predominant influence of schist groundwater in this sector. The depletion in HCO_3^-, Ca^{2+}, Na^+ and Mg^{2+} concentrations from the northern to the southern parts display the progressive decrease in schist groundwater participation. The undersaturation with respect to carbonate minerals also suggests a low availability of carbonate minerals and/or important mixing processes with low mineralized water. The concentrations in Cl^-, Ca^{2+}, Na^+ and Mg^{2+} measured in the lagoon are in favor of a contribution of both freshwater and seawater. The geochemical composition allows the identification of the three water bodies interacting within the alluvial aquifer: schist groundwater, river water and sea water (Fig. 5). Thus, particular consideration must be given to the quantification of mixing processes in the alluvial aquifer.

4.5. End-member characterization and mixing processes

Taking into account the circulations of water in the alluvial system, the chemical composition of groundwater is influenced by four potential end-members:

1. Groundwater from schist formations (Fig. 1): is defined by the mean concentrations in Cl^-, HCO_3^- and mean $\delta^{18}O$ isotopic signatures measured in Sc1, Sc2 and Sc3, during high flow and low flow conditions. This end-member is characterized by high HCO_3^- concentrations (mean of 409.3 mg/L) and moderate Cl^- concentrations (mean of 39.3 mg/L), in agreement with the geogenic control. The relative depletion in $\delta^{18}O$ (-7.1‰) is consistent with the high recharge altitude taking place in the upper part of the watershed.
2. River water: corresponds to the mean values measured at R1 to R5. This end-member is characterized by low concentrations in Cl^- (mean value: 15.7 mg/L) and HCO_3^- (mean value: 167.1 mg/L) and depleted signature in $\delta^{18}O$ (mean value: −8.1‰) explained by the high altitude of the source of the rivers.
3. Seawater: is characterized by a very high concentration in Cl^- (21293.9 mg/L), low concentration in HCO_3^- (154.2 mg/L) and enriched isotopic signature in $\delta^{18}O$ (+1.1‰).
4. Local rainfall: is a non-negligible natural source of recharge for the aquifer (Table 2) but contains very low amounts of ions. Concentrations supplied by rainfall to groundwater can be calculated by using the following equation (Appelo and Postma, 2005):

$$Concentration factor = \left(\frac{P}{Peff}\right) - WM_{ions} \tag{2}$$

where P represents the total volume of annual precipitation, P*eff* refers to the effective rainfall (the total volume of annual precipitation – the total annual evapotranspiration) and WM is the weighted mean of each ionic concentration for the period of measurement. The calculation gives the major ions concentrations of the local supply by rainfall: HCO_3^- = 18.1 mg/L, Cl^- = 12.4 mg/L, Na^+ = 7.4 mg/L, Ca^{2+} = 3.4 mg/L, SO_4^{2-} = 3.5 mg/L, NO_3^- = 2.9 mg/L K^+ = 1.1 mg/L, Mg^{2+} = 1.0 mg/L. The calculated groundwater concentrations due to rainfall are largely lower than those measured in the aquifer.

The mathematical resolution of 3 equations system with 3 unknown values leads to the mean participation percentages of each end-members to the chemical composition of each sampling point (Mohammed et al., 2014). The isotopic tracers are usually affected by fewer processes than chemical elements. In addition, the marked altitudinal contrast on the watershed induces contrasted signatures of the stable isotopes of water molecule. Thus, the isotopic data allow a better discrimination of the end-members and less ambiguous interpretation of mixing processes. The ternary mixing model can be expressed by:

$$\begin{cases} C_{sample}^{Cl^-} = C_{Sc}^{Cl^-} * F_{Sc} + C_{R}^{Cl^-} * F_R + C_{Sw}^{Cl^-} * F_{Sw} \\ C_{sample}^{\delta^{18}O} = \delta^{18}O_{Sc} * F_{Sc} + \delta^{18}O_R * F_R + \delta^{18}O_{Sw} * F_{Sw} \\ C_{sample}^{HCO_3^-} = C_{Sc}^{HCO_3^-} * F_{Sc} + C_{R}^{HCO_3^-} * F_R + C_{Sw}^{HCO_3^-} * F_{Sw} \end{cases} \quad (1)$$

where C^{Cl^-}, $C^{\delta^{18}O}$ and $C^{HCO_3^-}$ correspond to the content in Cl^-, $\delta^{18}O$ and HCO_3^- respectively, for each samples and end-members: Schist groundwater (Sc), River water (R) and Seawater (Sw). F is the relative contribution of each end-member to groundwater chemistry.

The results of mixing ratios calculations make it possible to define and quantify the major recharge sources. However, the mixing model was not sensitive enough to quantify the minor geochemical contribution from local rainfall. The model shows that river water is the major contribution for almost all the groundwater sampled on the watershed (Fig. 2b). The river water contribution clearly increases from the northern (mean value: 26%) to the southern (mean value: 72%) part.

The contribution of schist to groundwater from the northern part appears as greatly variable (Fig. 2b), from 10% in A1 to 99% in A6 (mean of 56%). Boreholes with a depth lower than 11 m (A5, A6, A8, A9 and A10) (Table 3 and 4) display the most important contribution from schist (from 38% to 99%). By contrast, boreholes with a depth of more than 11 m (A1-A3 and A13-A18) display a less important contribution from schist, from 10% to 32% with a mean of 25%. Thus, the alluvial aquifer seems vertically stratified. In the shallow part, groundwater from schistous formations ensures the majority of the recharge. The deepest part of the alluvial aquifer is mainly recharged by river water infiltration. This vertical subdivision could be explained by the thickness and the degree of the alteration of schistous formation. The alteration processes mainly affect the first meters of rock and increase the fissure permeability. The aquifer capacity is therefore more developed in the first meters compared to non-altered bedrock (BRGM, 1989, 1979) and the lateral contribution from schist to groundwater is more important in the shallow part of the alluvial aquifer. This vertical subdivision cannot be clearly proven in the southern part because of the shallow depths of the boreholes. However, regarding the schist contributions of 64% and 23% in the shallowest (A21, 6.2 m) and the deepest (A23, 23.0 m) boreholes, respectively the vertical subdivision highlighted in the northern part seems to exist also in the southern part.

Some sampling points (A27, A30 and A31) are distinguished by individual behavior (Fig. 2b). They display a higher contribution from schist, that is not correlated with the trends observed on the surrounding sampling points. A27 and A30 are localized close to an artificial drainage channel and/or a pumping station. The artificial drainage network allows lowering the shallow groundwater level, where the schist groundwater contribution is predominant. Thus, schist groundwater represents a large part of the volume of water pumped and rejected into the artificial channels. As channels are dug into the ground, without waterproofing system, water infiltration is facilitated, which explains the high contribution from schist groundwater. The infiltration of water from artificial drainage network must also be considered in the alluvial aquifer recharge processes. Reversely, A31 is mostly controlled by local geological influences. Its location at the boundary between schist formations and the alluvial aquifer logically explains the important contribution of schist groundwater to the local recharge.

Groundwater from the sand bar displays an almost equivalent

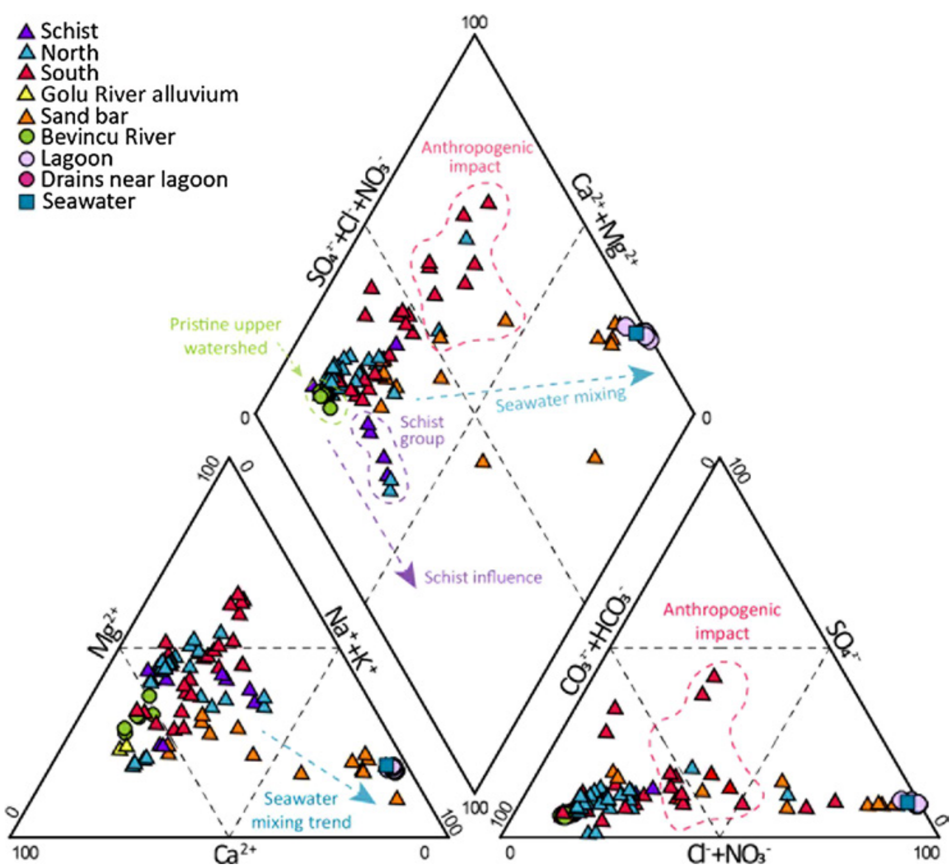

Fig. 4. Piper diagram for all water samples.

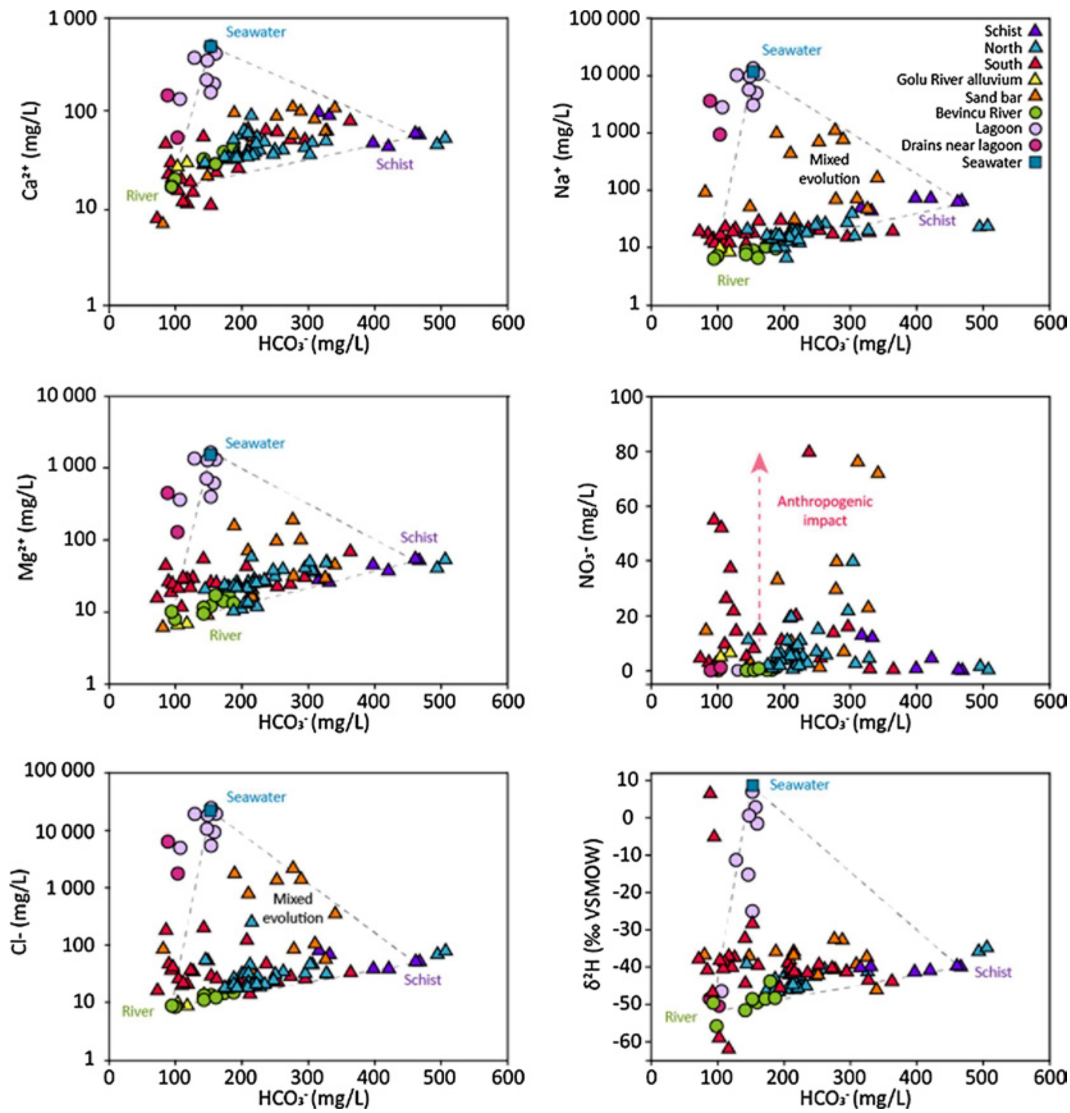

Fig. 5. Plots of the concentrations of Ca^{2+}, Na^{+}, Mg^{2+}, NO_3^{-}, Cl^{-} and isotopic signature in $\delta^{18}O$ with respect to HCO_3^{-}.

contribution of schist and river water (Fig. 2b). This observation highlights the lateral continuity of the alluvial aquifer, flowing under the shallow lagoon. The 3 points located at the North (S1, S2, S3), close to the channel, are the most impacted by seawater mixing, according to the isotopic data.

Finally, the lagoon water results from a mixing between seawater, river water and schist lateral contribution. The seawater contribution is more important close to the channel, directly connected to the sea. The sea water percentage progressively decreases while the freshwater percentage (i.e. river water and schist groundwater, Fig. 2b) increases as the distance to the channel grows. The river water contribution and the lateral contribution from schist groundwater range from 4% to 57% and from 3 to 11%, respectively (Fig. 2b). Among these freshwater supplies, the sampling points L2 and L3 show the most important contribution of groundwater from schist formations (Fig. 2b). These two sampling points are located in the vicinity of pumping stations which pumped the surface groundwater drained into the artificial channels. As groundwater from schistous formations participates mainly in the recharge in the first 10 m of the aquifer, it is preferentially drained. Therefore, the contribution of groundwater from schist formations is more important in these two points. The mixing model does not allow yet to distinguish river water supplies provided by runoff or having passed through the alluvial aquifer. This mixing model confirms the groundwater dependence of the lagoon already highlighted by piezometric data (Fig. 2b) but does not allow the exact quantification of the groundwater supplies.

4.6. Emerging compounds: groundwater vulnerability and tracers of ground-surface connections

4.6.1. Anthropogenic impact and groundwater vulnerability

Fig. 6 displays the spatial distribution of total EOCs concentrations, distributed all over the alluvial aquifer. Among the 51 EOCs analyzed, 44 are detected in significant quantities in sewage from wastewater treatment plan (TP1and TP2, Fig. 1) and sewage lift stations (LS1 and LS2) (Table 1). In the sewage waters, the high EOCs contents are found together with high MTE contents (Table 5). However, only 6 EOCs among the 51 analyzed, namely ibuprofen, paracetamol, caffeine, paraxanthine, cotinine and acesulfam, are detected in the natural waters (Table 5 and Fig. 7).

The set of EOCs detected signals anthropogenic pollution due to domestic uses and not from agricultural or industrial contamination. The important urbanization of the area seems to represent the main threat for the alluvial aquifer. Even if the ECOs concentrations are very high in sewage water (Table 5), their concentrations in natural water are relatively low. Thus, the impact on groundwater seems to be

Table 5
EOCs and TME concentrations in natural and sewage waters.

Sample ID	Caffeine (ng/L)	Paraxanthine (ng/L)	Ibuprofen (ng/L)	Paracetamol (ng/L)	Cotinine (ng/L)	Acesulfam (ng/L)	Li (µg/L)	B (µg/L)	Al (µg/L)	Cr (µg/L)	Mn (µg/L)	Fe (µg/L)	Co (µg/L)	Ni (µg/L)	Cu (µg/L)	Zn (µg/L)	As (µg/L)	Ba (µg/L)
A2	< 100	< 100	28	< 10	< 20	< 50	1.61	27.46	1.01	0.88	0.06	1.53	0.05	0.73	0.92	2.53	0.07	16.14
A11	< 100	< 100	28	< 10	< 20	< 50	1.47	26.04	0.80	3.74	0.03	1.00	0.07	13.96	2.74	1.70	0.58	7.56
A21	< 100	< 100	27	13	< 20	< 50	0.95	50.00	15.64	1.30	1.92	24.64	0.21	59.17	1.80	8.71	0.13	30.47
A23	< 100	< 100	150	< 10	< 20	< 50	3.28	16.88	0.81	6.90	0.08	0.97	0.03	3.80	1.16	1.85	0.06	4.74
A24	< 100	< 100	30	< 10	< 20	< 50	2.31	25.37	3.36	2.88	0.34	3.86	0.05	3.73	1.04	7.46	0.33	13.60
A25	220	< 100	110	34	61	78	0.98	25.35	2.74	3.35	0.30	4.24	0.04	14.41	0.64	0.99	0.16	7.84
A31	< 100	< 100	27	< 10	< 20	< 50	2.29	102.43	2.44	13.72	0.32	1.03	0.03	1.13	16.44	43.72	0.33	12.58
S1	650	653	23	< 10	< 20	120	9.91	126.08	1.82	0.12	65.78	181.88	0.29	9.52	0.79	18.93	19.55	1.27
S3	< 100	< 100	25	< 10	< 20	< 50	10.77	127.09	4.11	0.50	89.08	55.52	0.36	15.88	5.33	10.94	5.56	6.25
S6	2300	2210	33	27	27	110	5.56	35.70	3.04	0.63	220.56	17.15	0.24	3.63	1.81	2.53	4.75	1.34
R1	120	< 100	26	11	< 20	< 50	0.61	15.58	2.46	1.70	8.76	11.79	0.09	3.90	0.91	0.46	0.25	5.74
L1	1800	1920	< 20	48	< 20	< 50	115.38	4377.17	30.81	0.27	24.36	5.54	0.21	8.10	1.36	1.75	1.16	22.02
L2	1200	639	< 20	19	< 20	< 50	110.29	4031.86	31.93	0.34	5.72	0.78	0.11	9.14	1.25	0.80	0.94	26.79
L3	< 100	< 100	< 20	< 10	< 20	< 50	83.84	2996.14	22.54	0.26	46.46	3.34	0.19	10.19	1.75	2.95	0.89	33.58
LS1	150,000	91,300	4100	160,000	5700	9600	10.58	281.38	75.21	1.11	1430.30	219.51	0.86	5.08	1.66	10.37	0.70	8.98
LS2	150,000	46,700	4000	110,000	5260	9600	5.09	143.30	387.17	2.23	36.62	267.49	2.69	6.69	9.00	17.44	0.54	18.87
TP1	79,000	78,000	6000	18,000	12,100	12,000	5.52	177.56	160.94	1.30	35.26	165.44	1.12	5.70	4.52	9.01	0.56	25.91
TP2	170,000	120,000	7600	120,000	9850	11,000	7.18	198.34	151.69	1.15	40.60	270.79	1.36	5.42	2.04	15.97	0.44	16.91

currently limited. It is also true for the MTE, especially B, Al, Mn, Fe, Cu and Zn. Their high concentrations in untreated sewage water are significantly lower in groundwater, diluted by the mixing processes. Groundwater from the sand bar (S1 and S6) seems to be the most impacted, showing high contents in EOCs and MTE (Mn, Fe, Zn and As).

Most of the EOCs detected have similar properties, with notably $LogK_{ow} < 4$, indicating that the compounds are easily degradable in the environment (Buser et al., 1999; Buerge et al., 2003; Benotti and Brownawell, 2009). Even if persistent pharmaceuticals (carbamazepine, diclofenac, ketoprofen…) are detected in wastewater at noticeable concentrations (Table 1), none of them was detected in natural waters (Table 5). Ibuprofen was detected in 100% of samples from ground and surface waters, from 23 ng/L to 150 ng/L (Table 5). The ibuprofen is the only EOCs measured in groundwater from the northern part. In contrast, groundwater from the southern part is impacted by caffeine (220 ng/L), paracetamol (from 13 ng/L to 34 ng/L), cotinine (61.1 ng/L) and acesulfam (78 ng/L) (Fig. 7). The paraxanthine is also detected (from 653 ng/L to 2210 ng/L) in samples from the sand bar.

The omnipresence of ibuprofen brings to light the domestic consumption habits. Ibuprofen, commonly used for various treatments, is a cheap over-the-counter anti-inflammatory drug. Its presence is likely to be related to a significant consumption but also to its persistence in these particular environmental conditions. Indeed, the photodegradation is the dominant attenuation mechanism of this compound in the environment (Yamamoto et al., 2009; Araujo et al., 2014). However, the photodegradation can be a seasonal process and is also very limited within the groundwater, suggesting the high potential of ibuprofen infiltrating the alluvial aquifer and persisting in groundwater. Ibuprofen can therefore be considered as a "pharmaceutical fingerprint" of this study area.

Finally, EOCs provide valuable information about the lagoon's vulnerability (Table 5). Even if the groundwater is known to be influenced by anthropogenic contamination (Erostate et al., 2018) no major contamination can be evidenced from geochemical data of the lagoon's water because of the large dilution caused by mixing processes between ground and surface water. However, EOCs are detectable at very low concentrations (ng/L) and EOCs contents measured in the lagoon (caffeine from 1200 ng/L to 1800 ng/L, paraxanthine from 6390 to 1920 ng/L and paracetamol from 19 ng/L to 48 ng/L) highlight the anthropogenic contamination (Fig. 7). Ibuprofen, detected in all groundwater samples, was not found in the lagoon's water (Fig. 7). As previously demonstrated, groundwater participates to the fresh water supplies to the lagoon (Fig. 2b). The absence of ibuprofen in the lagoon water therefore highlights the occurrence of attenuation processes such as photodegradation and biodegradation. The particularly high concentrations of other EOCs in the lagoon (Fig. 7) displays that, even if they are degradable, the quantity of pollutants is too important and/or the degradation processes are too low to remove the contamination. Regarding the ecotoxicity, the bioaccumulation capacity of EOCs and their impact on living organisms and ecosystems (Gossett et al., 1983; Santos et al., 2010), the occurrence of micro pollutants in a classified Natural Reserve site should be considered as a serious concern.

4.6.2. Ground-surface water interaction

To highlight the anthropogenic impact and the hydrosystem's vulnerability, EOCs can also be used to specify ground-surface water interactions and then, recharge areas of the aquifers (Sassine et al., 2015; Zirlewagen et al., 2016). Labile EOCs (caffeine, paracetamol) were previously identified as good tracers of untreated wastewaters of human origin (Buerge et al., 2003) but WWTPs is not always the main source of labile pharmaceuticals (Vystavna et al., 2013). These organic compounds can enter the water flow with runoff (Bartelt-Hunt et al., 2009) and/or untreated illegal discharges or in the event of an accident on the WWTPs.

In this study, river water displays significant concentrations of ibuprofen (26 ng/L), caffeine (120 ng/L) and paracetamol (11 ng/L),

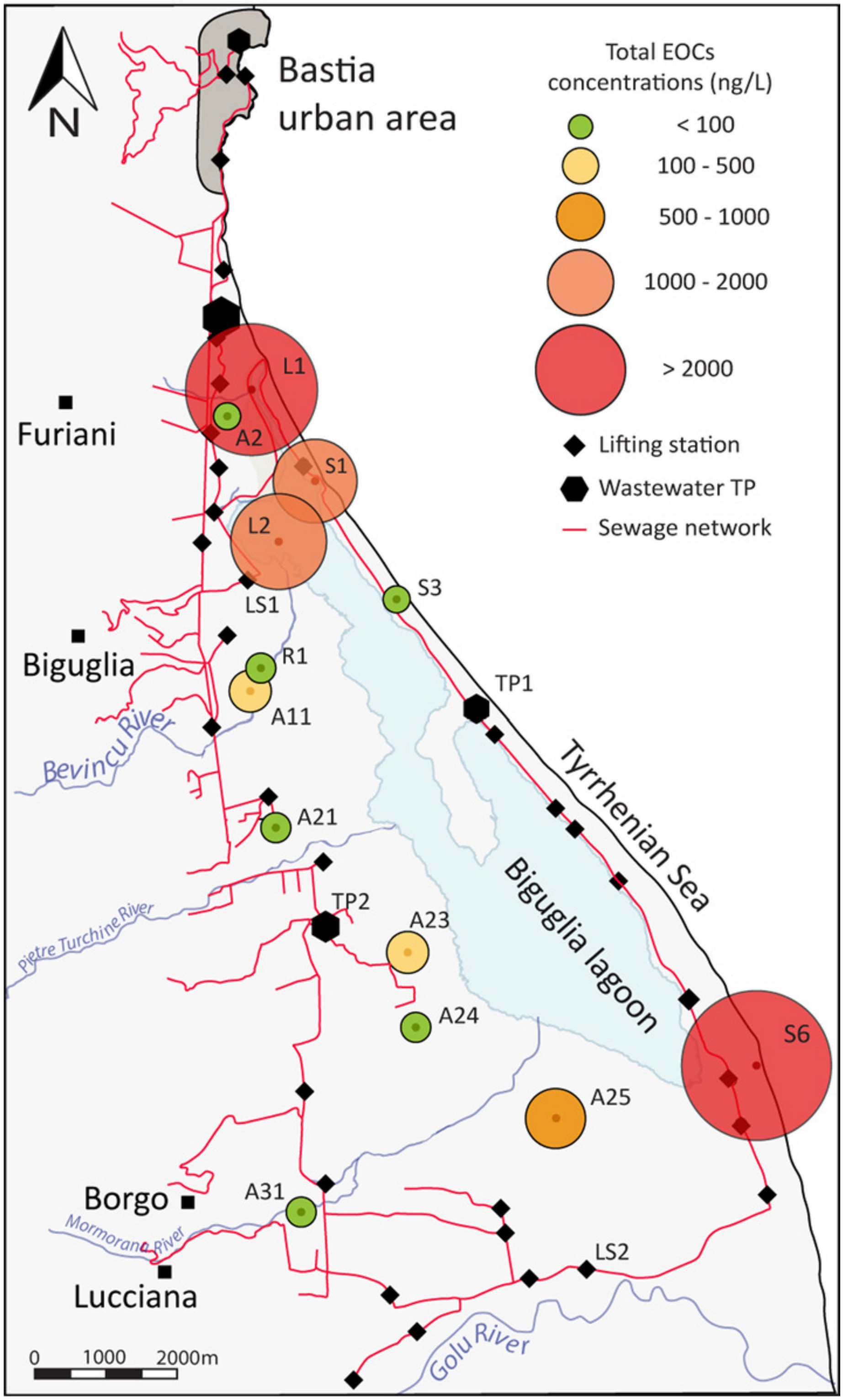

Fig. 6. Spatial distribution of sanitation network, lifting stations (LS) and wastewater treatment plant (TP) and total measured concentrations in EOCs.

suggesting the impact of sewage water from urban runoff and/or overflow of sewage collector (Fig. 7). The sampling points A2, A11, A24 and A31 displays ibuprofen concentrations (from 27 ng/L to 30 ng/L) similar to those measured in the river (26 ng/L) (Table 5). They also benefit from a high river water contribution (67% to 77%) (Fig. 2b). Thus, EOCs constitute a complementary tracers to reinforce the established mixing model and to trace surface water inflow. The absence of caffeine and paracetamol in groundwater is explained by their relatively fast biodegradation during transport through the subsurface (Benotti and Brownawell, 2009). The sampling point A23 is impacted by ibuprofen since it showed the highest concentration (150 ng/L) (Table 5). This elevated concentration can be explained by the longer residence time of groundwater (Tables 3 and 4), allowing the progressive accumulation of ibuprofen into the alluvial aquifer,

The types of EOCs detected also provide qualitative temporal information (Robertson et al., 2013, 2016). The information provided by

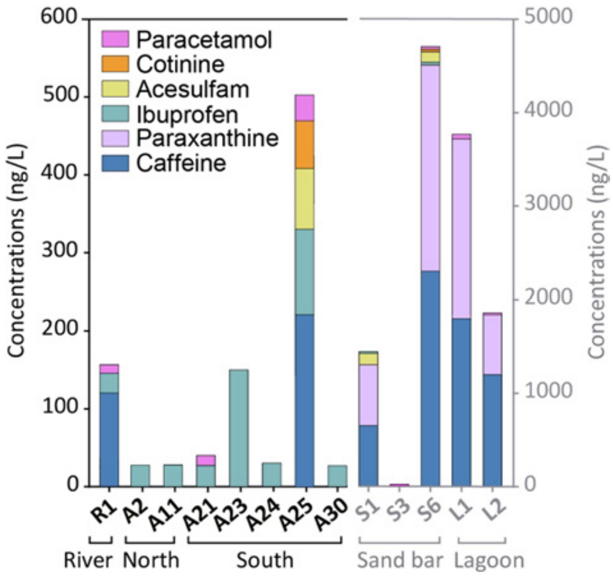

Fig. 7. Concentrations in EOCs in surface, ground and lagoon water.

EOCs is particularly important in the case of non-persistent EOCs (LogKow < 4), with a short half-life time (never exceeding a few months). The presence of highly biodegradable compounds such as caffeine or paracetamol indicates the rapid infiltration of contaminated water into the alluvial aquifer. If caffeine could be of natural origin (Buerge et al., 2003), paracetamol is a compound of purely human origin, well known to be efficiently eliminated by WWTPs (Buser et al., 1999; Sui et al., 2015). Its presence thus argues for an untreated sewage contribution (wastewater and urban runoff infiltration).

The high contents in NO_3^- and EOCs measured at the sampling point A25 (Fig. 5 and Fig. 7) and the mixing model (Fig. 2b) demonstrate the high infiltration potential of surface water towards the groundwater. Paracetamol also allows to highlight the rapid flow at the sampling point A21 (Fig. 7). The low contribution of river water rather suggests a contamination from untreated sewage than seepage from septic tanks or leaks from the sanitary network. The high concentration in several MTE such as Al, Fe, Ni, Ba, Zn and Mn also argues in favor of a contamination from untreated sewage. This observation is confirmed by the isotopic signatures in $\delta^{15}N\text{-}NO_3^-$ and $\delta^{11}B$ previously measured (Erostate et al., 2018). Indeed, the combination of elevated $\delta^{15}N\text{-}NO_3^-$ values with depleted $\delta^{11}B$ signatures suggests systematic contamination by sewage (Vengosh et al., 1994; Widory et al., 2013).

The data on environmental concentration and physical chemical

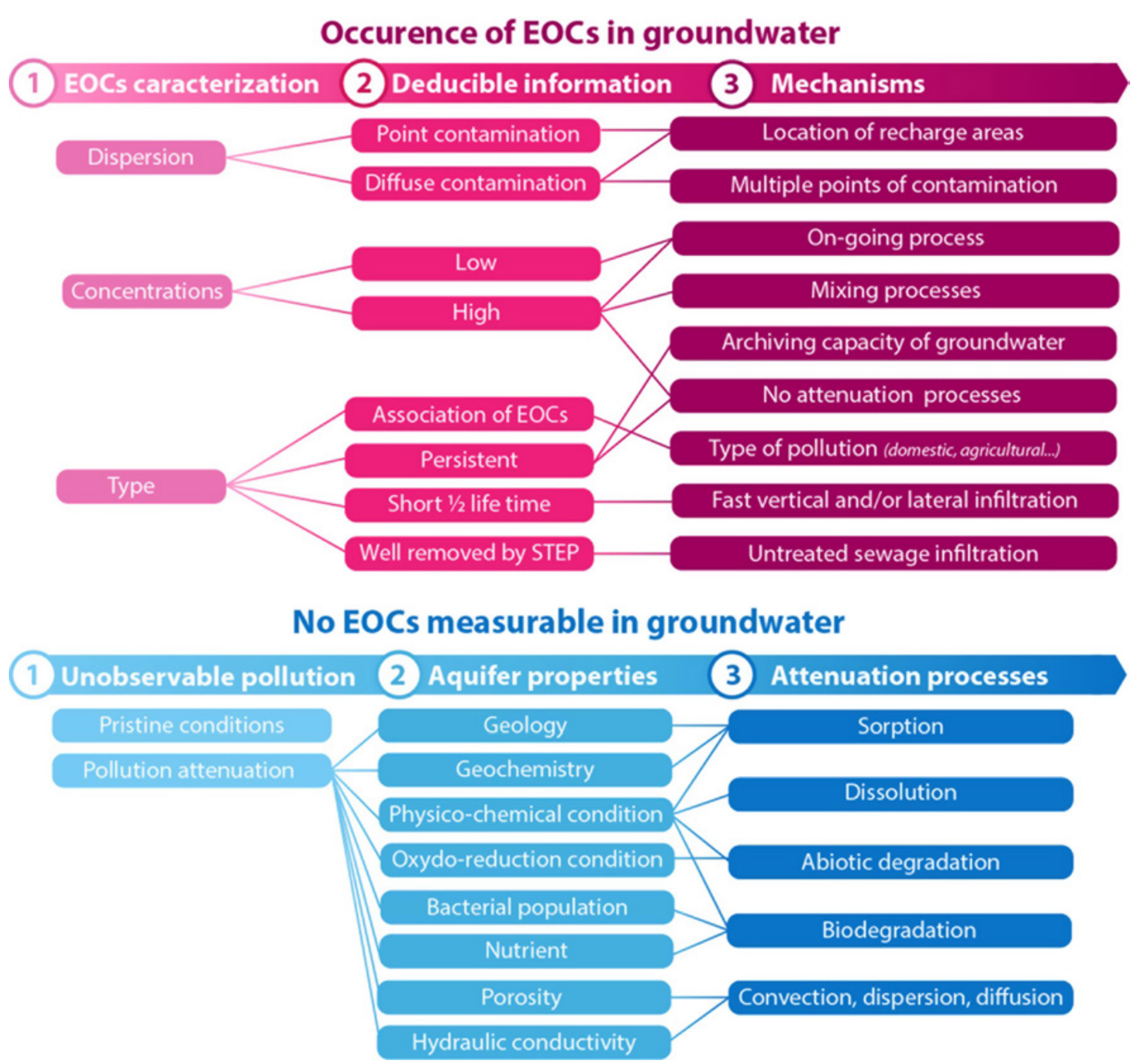

Fig. 8. Mechanisms and processes deduced from the occurrence of EOCs and actions to undertake in case of no EOCs detection in groundwater.

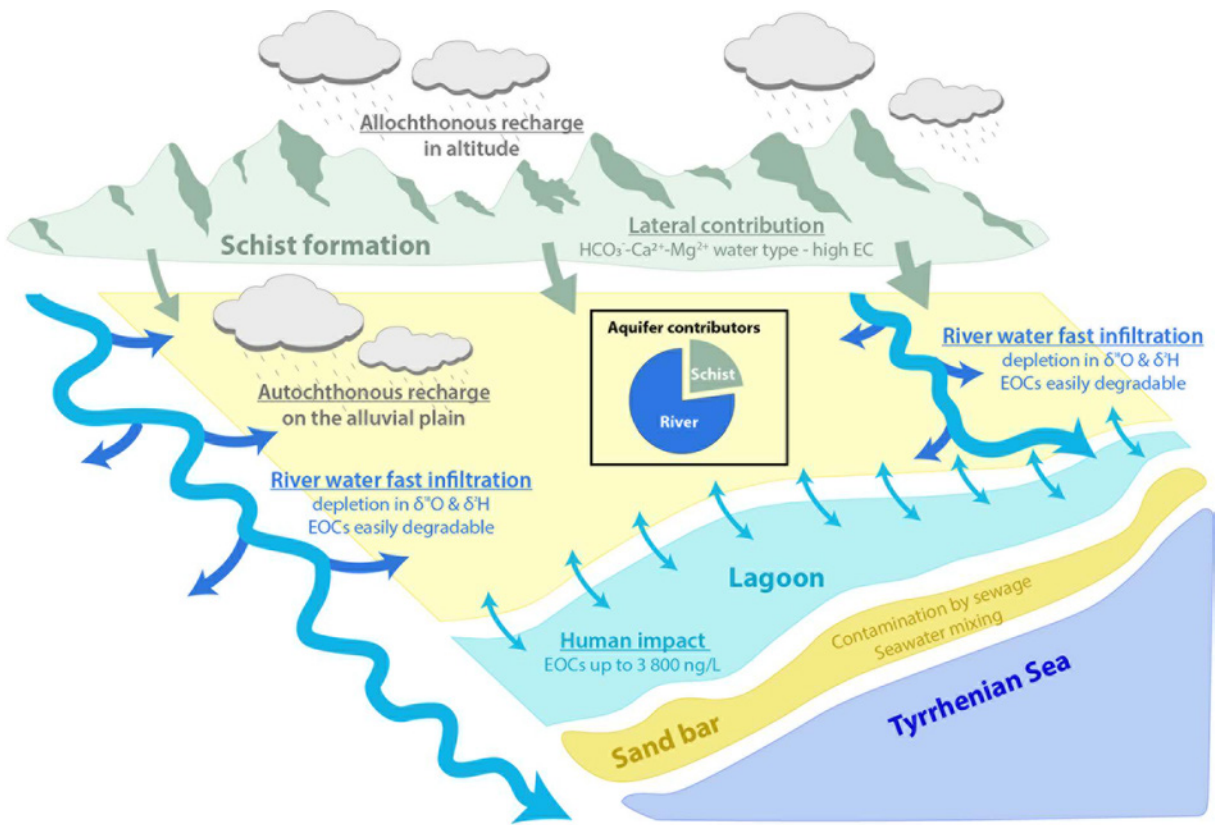

Fig. 9. Schematic representation of the alluvial aquifer's behavior.

properties of EOCs can be useful to provide relevant information about the mechanisms affecting the hydrosystems (Fig. 8). First of all, the occurrence of EOCs clearly highlights the vulnerability of the hydrosystem to anthropogenic activities. Thus, the spatial extend of the pollution makes it possible to identify pollutant's infiltration areas but also recharge areas of the aquifer (Sassine et al., 2015; Zirlewagen et al., 2016). The concentrations inform about the level of pollution affecting the resource, the archiving capacity of the system or again the potential presence/absence of attenuation processes. The type of EOCs allows the identification of the source of pollution but also provides temporal information (Robertson et al., 2013, 2016). The information provided by EOCs is particularly important in the case of non-persistent EOCs, with a short half-life time. The compounds, which decompose rapidly, will allow to precise the understanding of short-times processes such as fast vertical and/or lateral infiltration. EOCs thus provide new complementary information which can support and refine the conclusions resulting from classic chemical and isotopic approach. The absence of measurable EOCs contamination could also provide information about pristine conditions and remediation processes.

4.7. A conceptual model of the hydrosystem behavior

A conceptual model of the Marana-Casinca hydrosystem can now be proposed (Fig. 9). The piezometric map (Fig. 2a) shows very clearly the groundwater dependence of the lagoon with most of the groundwater's flow oriented from West to East, that is to say from the mountain to the lagoon. The artificial drainage network clearly affects the hydrodynamic functioning of the alluvial aquifer. The drainage channels represent a specified head boundary condition which exacerbates the natural hydraulic gradient of the alluvial aquifer. Four end-members that contribute to the alluvial aquifer were recognized: rainfall, schist groundwater, river water and seawater (Fig. 9). Isotope of the water molecule shows that the alluvial aquifer is recharged by rainwater. The local recharge by rainfall seems to contribute only slightly to the geochemistry of the alluvial aquifer. Thus, the chemical content of the investigated water bodies mainly depends on the contribution of the 3 other end-members. The lagoon's water results from groundwater, but also from surface water and seawater mixing, in very different proportions according to the location on the watershed (Fig. 2b).

The socio-economic activities developing on the Marana-Casinca alluvial plain also impact the groundwater quality. The NO_3^- (Fig. 5) and EOCs concentrations (Fig. 7) clearly highlight the anthropogenic contamination. The detection of highly degradable EOCs in groundwater emphasizes the high transmissivity of the alluvial aquifer. The alluvial deposits seem to locally favor rapid pollution infiltration towards the aquifer, which makes it more vulnerable to anthropogenic pollution (Jaunat et al., 2018). This is particularly true in the southern part of the watershed and on the sand bar. In these areas, the urbanization is largely developed (Robert et al., 2015, 2018) as well as the sanitation network. The qualitative degradation of groundwater induced by mixing processes between fresh and salt water seems limited to the freshwater lens in the sand bar (Fig. 2). Yet, some studies have already shown the vulnerability of the alluvial aquifer to saltwater intrusion in two sectors, namely the well field used for Bastia's drinking water production (A11–A18) and the quarry for aggregates extraction (A28 and A29) (BRGM, 2008, 1993).

The recharge of the alluvial aquifer involves different contributions. The northern part of the watershed receives an allochthonous recharge while the southern part benefits mostly from local rainfall (Fig. 3). The recharge brought by rainfall has a limited impact on the groundwater's geochemistry. The infiltration of river water and the lateral flow of schist groundwater represent the two major contributors (Fig. 2b). Their contribution rate varies from the northern part to the southern part, but also vertically, depending on the depth of the resource. The subsurface groundwater from the northern part benefits from a significant lateral recharge from schist groundwater (Fig. 9). The schist contribution decreases toward the southern part and rise as it gets deeper. Thus, the infiltration of river water clearly controls the recharge process (Fig. 2b). EOCs content allows to confirm the surface water inflow and to precise the rapid lateral and vertical infiltration towards the alluvial aquifer.

5. Conclusion

Anthropogenic activities, such as groundwater exploitation or artificial drainage networks developed for irrigation or sanitary issues, strongly modify the natural behavior of coastal hydrosystems and impact the groundwater quality. The appearance and detection of emerging pollutants, such as EOCs, can be very useful for understanding how these hydrosystems behave. Thanks to the study of their fate in the environment, EOCs provide new additional information which can support the observations from more common isotopic and chemical tracers. In this study, the innovative approach of combining geochemical and isotopic data with EOCs clearly allows to establish a relevant conceptual model, taking into account water–rock interactions, mixing processes, recharge and contamination modalities. This new coupling of isotope hydrology, geochemical tracers and EOCs provide precious information about the short transit time of water and identifies waters recently infiltrated as well as recharge and vulnerability areas. This approach can be helpful to understand anthropogenic influences and fast hydrological processes on any strongly anthropized hydrosystem. However, EOCs are still newly studied tracers. The progressive improvement in the knowledge of EOCs' behavior in the environment should help to further precise the temporal quantification of the fast infiltration flows. Meanwhile, in some appropriate cases, supplementing this approach by other complementary tracers such as radioactive tracers or atmospheric gases could be helpful to increase the temporal precision of the hydrological processes.

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4.3. Conclusion

L'approche multi-traceurs géochimiques et isotopiques développée sur l'hydrosystème de Biguglia a permis l'élaboration d'un premier modèle conceptuel de fonctionnement (Figure 16). Les eaux souterraines de l'aquifère de la Marana résultent des contributions de **trois pôles naturels majeurs : les précipitations, les eaux souterraines en provenance des contreforts schisteux de la Corse alpine et les eaux fluviales**. L'origine des précipitations contribuant à la recharge varie entre la partie Nord et la partie Sud du bassin versant. Les eaux souterraines du Nord du bassin versant et des alluvions du Golu bénéficient d'une **recharge allochtone assurée par les précipitations sur les contreforts schisteux de la Corse alpine**. Dans le secteur Sud et le lido, une **recharge autochtone est assurée grâce à l'infiltration directe des précipitations sur la plaine**. Le taux de recharge annuel moyen de l'aquifère par l'infiltration des eaux de pluie est estimé à 127 mm, soit environ 16% des précipitations annuelles. Les précipitations ne contribuent cependant que très faiblement à la chimie des eaux souterraines. La composition géochimique et isotopique des eaux souterraines est donc principalement influencée par l'infiltration des eaux fluviales et l'écoulement latéral des eaux souterraines en provenance des contreforts schisteux.

De manière générale**, l'infiltration des eaux fluviales contrôle clairement le processus de recharge de l'aquifère**. La contribution des eaux souterraines en provenance des contreforts schisteux est globalement moins importante, mais constitue tout de même un apport significatif. **Les contreforts schisteux constituent donc une limite à flux imposé pour l'aquifère.** Leur contribution varie du Nord au Sud de la plaine mais également verticalement. Les eaux souterraines de subsurface (jusqu'à 11 m) bénéficient d'une recharge importante par les flux latéraux en provenance des contreforts schisteux. Cette contribution diminue du Nord au Sud ainsi qu'avec la profondeur (Figure 13).

La dépendance de la lagune aux apports en eau souterraine est également quantifiée. Les eaux souterraines en provenance des contreforts schisteux constituent 3 à 11% des apports en eau douce vers la lagune et les eaux fluviales représentent quant à elles de 4 à

57%. Cependant, le modèle de mélange ne permet pas de différencier la part liée aux écoulements directs des eaux fluviales vers la lagune de celle liée aux eaux fluviales infiltrées dans l'aquifère avant de rejoindre la lagune. **La contribution des eaux souterraines est donc d'au moins 3 à 11%, à laquelle s'ajoute une proportion non quantifiée d'eau souterraine rechargée par l'infiltration des eaux de rivière**. Enfin, la contribution de l'eau de mer, majoritaire à proximité du grau, diminue progressivement vers le Sud jusqu'à devenir minoritaire à proximité du canal du Fossone.

Les activités socio-économiques développées sur la plaine ont une incidence significative sur la qualité de l'eau. **Les concentrations en NO_3^-, au-dessus du bruit de fond géochimique naturel (5 à 7 mg/L) et la présence de micropolluants organiques** sur l'ensemble de la plaine illustrent clairement une contamination d'origine anthropique. **La présence de micropolluants rapidement dégradables souligne la grande transmissivité de cet aquifère**. Les alluvions favorisent localement l'infiltration rapide de polluants vers l'aquifère, augmentant sa vulnérabilité vis à vis des pollutions anthropiques. Cette vulnérabilité est exacerbée dans les secteurs largement urbanisés et parcourus par un important linéaire d'infrastructures d'assainissement, c'est-à-dire dans la partie Sud du bassin versant et sur le lido.

L'étude du cortège de micropolluants présents dans les eaux ainsi que de leur comportement dans l'environnement prodigue de précieuses informations sur les mécanismes hydrologiques. **Les micropolluants organiques présents dans les eaux souterraines constituent des traceurs complémentaires aux traceurs isotopiques et géochimiques plus couramment utilisés**. La détection de micropolluants organiques permet notamment de préciser les zones de recharge et prodigue une résolution plus fine pour une meilleure compréhension des flux d'infiltration rapide (<1 an).

Enfin, la dégradation qualitative des eaux souterraines induite par les processus de mélange entre l'eau douce et l'eau salée semble relativement limitée, principalement localisée dans la lentille d'eau douce du lido. Les secteurs précédemment pointés pour leur vulnérabilité à l'intrusion d'eau salée, à savoir le champ captant utilisé pour l'alimentation en eau potable de Bastia et la carrière d'extraction de granulats, ne présentent pas d'anomalie détectable dans le cadre de nos travaux.

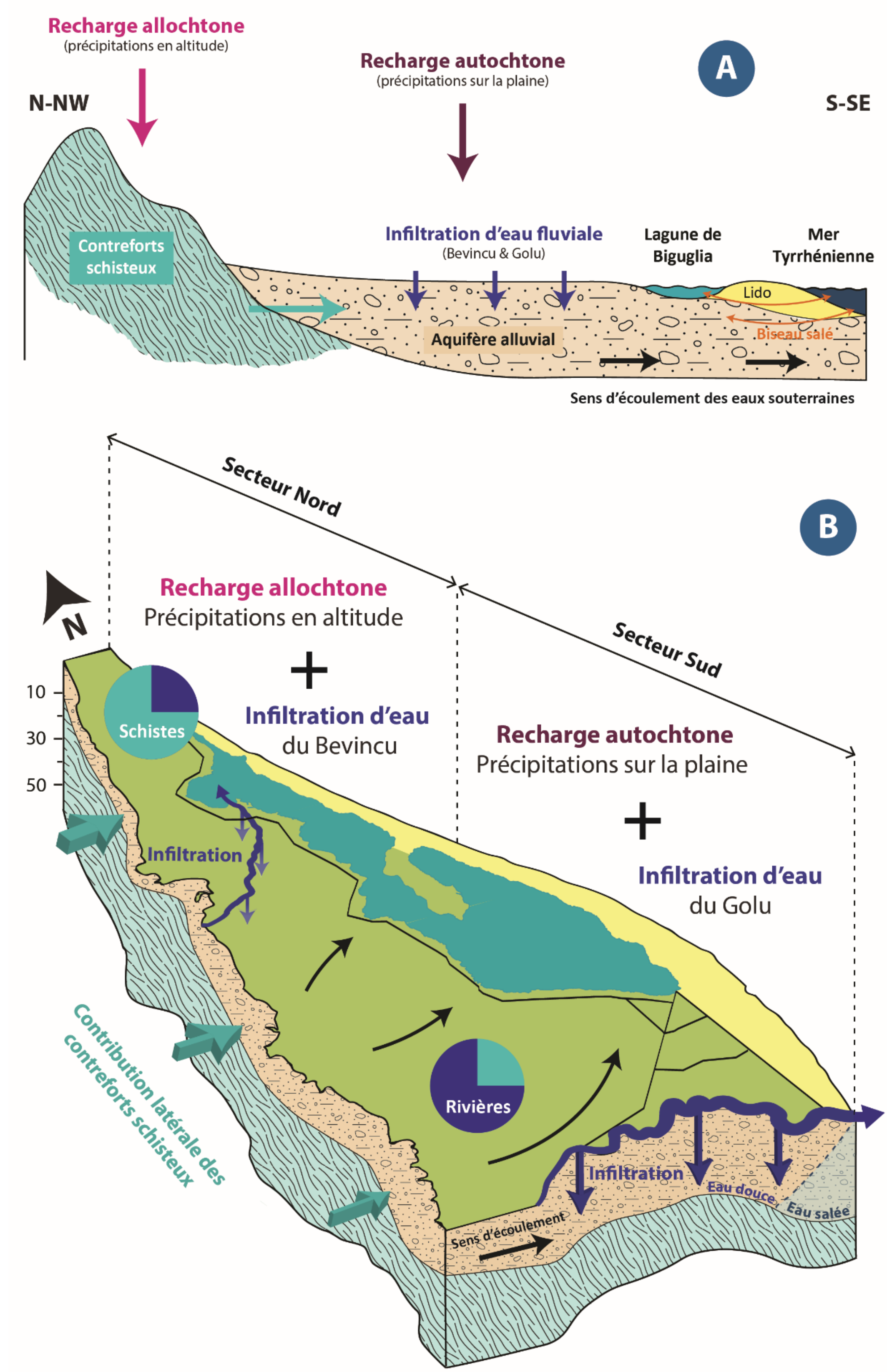

Figure 16: Représentation schématique en coupe (A) et en 3 Dimension (B) des processus hydrologiques de l'hydrosystème de la lagune de Biguglia.

Chapitre 5 :

Evolution temporelle des sources de contaminations et héritage des polluants azotés

Ce chapitre s'intéresse à l'identification des sources de pollutions d'origine azotée et à la compréhension de leurs dynamiques spatiales et temporelles grâce à l'usage des isotopes du nitrate, du bore et du tritium. Cette approche a permis de mettre en évidence la capacité d'archivage des eaux souterraines et la présence d'un legs nitraté lié aux activités historiques d'aménagement de la plaine.

5. Evolution temporelle des sources de contaminations et héritage des polluants azotés

5.1. Introduction

Les nitrates (NO_3^-) constituent la principale source de pollution des ressources en eau à travers le monde. Problématique pour l'usage de l'eau à des fins de consommation humaine (>50 mg/L), les fortes concentrations en NO_3^- sont également une source potentielle de dégradation importante des écosystèmes côtiers tributaires des eaux souterraines, en contribuant notamment au processus d'eutrophication. Compte tenu des conditions d'écoulement des eaux souterraines en milieux poreux, la vectorisation des polluants vers les écosystèmes côtiers peut être un processus long, qu'il convient d'appréhender en amont pour assurer à moyen et long terme la protection et la préservation de ces écosystèmes fragiles.

Les fortes concentrations en NO_3^- détectées précédemment dans les eaux souterraines de l'aquifère de la Marana (jusqu'à 80 mg/L – voir chapitre 3) laissent peser une forte incertitude quant à l'origine et au devenir de ces contaminants azotés. Pour la lagune de Biguglia, déjà caractérisée par des teneurs élevées en nutriments azotés (principalement sous la forme NH_4^+), le déversement des NO_3^- contenus dans les eaux souterraines pourrait encore amplifier cette dégradation qualitative du milieu. Afin de mettre en place des politiques de gestion plus adaptées, permettant de limiter au maximum les apports en NO_3^- vers les milieux aquatiques, il est donc nécessaire de mieux contraindre les sources et les dynamiques spatio-temporelles des flux nitratés.

Dans cet objectif, les signatures isotopiques du NO_3^- ($\delta^{15}N\text{-}NO_3^-$, $\delta^{18}O\text{-}NO_3^-$) peuvent prodiguer de précieuses informations quant aux sources et au devenir des NO_3^- dans les eaux souterraines. En effet, chaque source de NO_3^- se caractérise par une signature isotopique qui lui est propre, permettant alors son identification. L'inconvénient majeur de cette approche reste cependant le caractère non conservatif de l'azote, pouvant induire une modification de la signature isotopique initiale, notamment en cas de processus de dénitrification par exemple. Pour pallier à cet inconvénient, l'usage des isotopes du NO_3^- est ici complété par l'usage du ^{11}B. Cet élément, ubiquiste et co-migrant du NO_3^-, présente le double avantage de i) ne pas être affecté par les processus de transformation biogéochimiques au sein des aquifères et ii) de permettre la distinction entre le fumier d'origine animal et les eaux usées d'origine humaine. Il est par conséquent particulièrement utile pour identifier les sources de NO_3^- en cas d'altération des signatures isotopiques du NO_3^-, de dénitrification ou de contamination conjointe d'origine animale et humaine. Afin de retracer l'évolution des sources de NO_3^- et les dynamiques temporelles de ces flux, les données isotopiques ont été corrélées avec des outils de datation des eaux souterraines (^3H et CFCs) ainsi qu'avec une étude de l'évolution de l'occupation des sols sur le bassin versant.

5.2. Résultats et interprétations

Les résultats et interprétations de ces travaux ont fait l'objet de la rédaction d'un article publié dans *« Science of the Total Environment »* en décembre 2018, intitulé « Delayed nitrate dispersion within a coastal aquifer provides constraints on land-use evolution and nitrate contamination in the past», présenté ci-après. Les principales avancées concernant la capacité d'archivage des eaux souterraines de la Marana et l'impact du legs nitraté historique sont également résumées en français, en conclusion de ce chapitre.

Delayed nitrate dispersion within a coastal aquifer provides constraints on land-use evolution and nitrate contamination in the past

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Delayed nitrate dispersion within a coastal aquifer provides constraints on land-use evolution and nitrate contamination in the past

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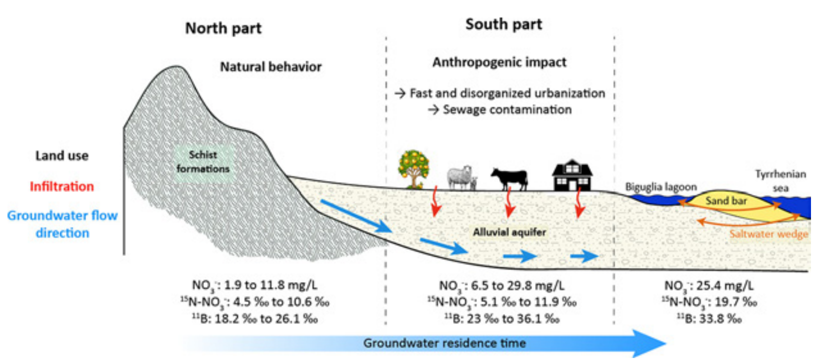
HIGHLIGHTS

- High nitrate contents are measured in an alluvial aquifer supplying a coastal lagoon.
- δ^{15}N-NO_3^- and δ^{11}B signatures identify sewage as the main nitrate source today.
- Increase in NO_3^- with groundwater age displays the archiving capacity of groundwater.
- Highest NO_3^- concentrations can be linked to agricultural activities in the past.
- Legacy pollution that accumulated in the aquifer is a threat for the lagoon.

GRAPHICAL ABSTRACT



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ABSTRACT

Identifying sources of anthropogenic pollution, and assessing the fate and residence time of pollutants in aquifers is important for the management of groundwater resources, and the ecological health of groundwater dependent ecosystems. This study investigates anthropogenic contamination in the shallow alluvial aquifer of the Marana-Casinca, hydraulically connected to the Biguglia lagoon (Corsica, France). A multi-tracer approach, combining geochemical and environmental isotopic data (δ^{18}O-H_2O, δ^{2}H-H_2O, ^{3}H, δ^{15}N-NO_3^-, δ^{18}O-NO_3^-, δ^{11}B), and groundwater residence-time tracers (^{3}H and CFCs) was carried out in 2016, and integrated with a study of land use evolution in the catchment during the last century. Groundwater NO_3^- concentrations, ranged between 2 mg/L and up to 30 mg/L, displaying the degradation of groundwater quality induced by anthropogenic activities (agricultural activities). Comparatively high δ^{15}N-NO_3^- values (up to 19.7‰) in combination with δ^{11}B values that were significantly lower (between 23‰ and 26‰) than the seawater background are indicative of sewage contamination. The ongoing deterioration of groundwater quality can be attributed to the uncontrolled urbanization development all over the alluvial plain, with numerous sewage leakages from the sanitation network and private sewage systems. Integration of contaminant and water-residence time data revealed a progressive accumulation of pollutants with time in the groundwater, particularly in areas with major anthropogenic pressure and slow dynamic groundwater flow. Our approach provides time-dependent insight into nitrogen pollution in the studied

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aquifer over the past decades, revealing a systematic change in the dominant NO_3^- source, from agricultural to sewage contamination. Yet, today's low groundwater quality is to large parts due to legacy pollution from land-use practices several decades ago, underlining the poor self-remediating capacity of this hydrosystem. Our results can be taken as warning that groundwater pollution that happened in the recent past, or today, may have dire impacts on the quality of groundwater-dependent ecosystems in the future.

1. Introduction

Coastal lagoons provide a large spectrum of socio-economic benefits for humans, such as food provision, recreation and ecotourism, water regulation, coastal protection or pollution bioremediation (Farber and Costanza, 1987; Spurgeon, 1999; Perez-Ruzafa and Marcos, 2012). However, due to their location, these unique ecosystems rely on the fragile equilibrium between freshwater and marine water inflows, and are particularly susceptible to natural hazards (climate change, sea-level rise, coastal erosion) (Ketabchi et al., 2016; Klassen and Allen, 2017). This vulnerability is accentuated by the increasing anthropogenic pressure exerted on them (Mahlknecht et al., 2017). Indeed, a large part of the world's population is concentrated in coastal territories, causing an ever-growing urbanization (Barragán and de Andrés, 2015) and exposure to human activities, like agriculture, industry or mass tourism. Such activities are often associated with pollution (sewage leakages, fertilizers) that directly impact the quality of surface and ground water (Wang et al., 2016; Senthilkumar et al., 2017; Shuler et al., 2017). Groundwater plays an important role in the water supply of coastal wetlands (Cartwright and Gilfedder, 2015; Krogulec, 2016; Paz et al., 2017) and could be an important vector of anthropogenic pollution. Hence, groundwater degradation by nitrate contamination, for example, can contribute to the eutrophication of groundwater dependent ecosystems. A better understanding of pollution sources in aquifers, and the hydraulic connectivity between groundwater and groundwater dependant ecosystems is essential to ensure the protection and conservation of these ecosystems.

The isotopic signatures of dissolved nitrogen species ($\delta^{15}N$-NO_3^- and $\delta^{18}O$-NO_3^-) can provide valuable constraints on the sources and fate of fixed (i.e., bioavailable) nitrogen (N) in groundwater. Past studies have shown that common pollutant sources of nitrate in groundwater include soil-N, manure, synthetic fertilizer and sewage effluents (Widory et al., 2013; Gooddy et al., 2014; Zhang et al., 2014; Grimmeisen et al., 2017; Vystavna et al., 2017a). To some extent, these nitrate sources have distinct isotopic signatures, which can be used diagnostically to semi-quantify the relative importance of single N sources (Aravena et al., 1993; Yakovlev et al., 2015; Grimmeisen et al., 2017; Vystavna et al., 2017b). However, N does not necessarily behave conservatively, and processes such as denitrification lead to isotope fractionation altering the primary isotopic signatures (Fukada et al., 2004; McMahon and Böhlke, 2006). At the same time, such processes can act to alleviate fixed nitrate contamination by transforming it into harmless forms of N (i.e., N_2 in the case of denitrification and anaerobic ammonium oxidation) (Jetten et al., 2001; Yenigün and Demirel, 2013).

Indirect tracers of NO_3^- sources that co-migrate with dissolved nitrate, such as boron (B), may be employed to gain additional information on groundwater pollution. In contrast to N isotopes, the B isotopic composition ($\delta^{11}B$) is not affected by biogeochemical transformation processes in aquifers. It can therefore be helpful to discriminate the NO_3^- sources where denitrification may alter primary source signatures. On the other hand, the B isotopic composition can be affected by adsorption-desorption processes on clays, organic matter or aluminium and iron oxide surfaces (Palmer et al., 1987; Bassett, 1990; Yingkai and Lan, 2001; Lemarchand et al., 2002). The great value of B is that its isotopic signature is characteristic for human and animal waste (Vengosh et al., 1994; Widory et al., 2005). Hence B isotope ratios have successfully been used to trace wastewater leakage/injections to the groundwater in various studies (Xue et al., 2009; Cary et al., 2013; Guinoiseau et al., 2018), or, in combination with NO_3^- isotopic signatures, to identify sewage contamination in groundwater, and to discriminate animal manure and fertilizer sources (Widory et al., 2004, 2005; Seiler, 2005; Xue et al., 2009).

Although correlated isotopic signatures of N and B permit the semi-quantitative identification of anthropogenic sources of pollution, they do not provide information on the timing and temporal evolution of the contamination. Yet, NO_3^- infiltration can be a very lasting process, which depends on the watershed and aquifer properties. The time-lag between the emission of a pollutant and the contamination actually determined in groundwater can vary greatly (Han et al., 2015; Vero et al., 2017). Thus, detrimental impacts of past land use often leave footprints that may only be discovered several years or decades later. Identifying current NO_3^- sources is not sufficient for understanding actual groundwater NO_3^- concentrations. It is also necessary to evaluate the contribution of legacy NO_3^- related to past activities in the catchment. Integrated information on past land-use, the groundwater age (or time since infiltration), and the concentration of nitrate allows insight into the evolution of pollutant sources in the past, which is crucial for assessing a groundwater system's resilience towards contamination (Provitolo and Reghezza-Zitt, 2015). Groundwater residence time is an essential parameter for predicting future pollutant levels, necessary to establish a sustainable water resource management (Re et al., 2014; Han et al., 2015). To this end, tracers like ^3H, CFCs and SF_6 are commonly used to gain constraints on the groundwater age (Carreira et al., 2013; Kralik et al., 2014; Prada et al., 2015; Caschetto et al., 2016). Because of their long residence time, porous aquifers are particularly prone to the detrimental impacts of anthropogenic contamination. Indeed, continuous anthropogenic pollution that infiltrated an aquifer for several decades is likely to have accumulated to high levels in the groundwater, and the measured pollution today is the result of current and past anthropogenic activities.

Our goal was to combine geochemical (major ions), environmental isotope ($\delta^{18}O$-NO_3^-, δ^2H-H_2O, ^3H, $\delta^{15}N$-NO_3^-, $\delta^{18}O$-NO_3^-, $\delta^{11}B$), groundwater residence time tracer (^3H and CFCs) data, as well as information on land use evolution for the last century, in order to 1.) identify pollution sources in an alluvial plain aquifer in Northern Corsica (France), which is hydraulically connected to a coastal Mediterranean lagoon, and 2.) assess the temporal relationships (i.e., time lag) between the actual pollution and the groundwater contamination as observed today. Understanding the dynamics and pathways of pollution within an aquifer is essential for predicting the impact on coastal groundwater dependent ecosystems, as well as their vulnerability to land-use change. The studied site in particular, and groundwater dependent ecosystems in general, are essential components of the watershed, which are often susceptible to anthropogenic pollution. They represent the terminal sink for all sorts of dissolved and particulate matter, including harmful substances that may have been released to the environment long ago in the past.

2. Study area

2.1. Geography, geomorphology and land use

The Biguglia lagoon is a shallow (maximum depth of 1.8 m), brackish coastal lagoon located in the northern part of the island of Corsica (western Mediterranean, France) near Bastia (Fig. 1). The lagoon stands within the largest wetland of Corsica. Its area is about 14,5 km^2, with a length along the coastline of 11 km and a width of up to 2.5 km in the southern part. The Biguglia lagoon is separated from the Tyrrhenian Sea by a long sand bar, and seawater exchange is controlled by a narrow, natural channel at its Northern end. Ventilation with sea water is limited (Mouillot et al., 2000), particularly because of sand accumulations that regularly fill, and thus close, the channel, impeding water exchange with the Tyrrhenian Sea. The watershed covers 182 km^2 and comprises different geological formations. The northern and the western parts are formed by lustrous schist formations, with a high relief and a maximum altitude of 1450 masl. The remainder of the catchment in the South comprises the large Marana-Casinca sedimentary plain with mostly Quaternary alluvial deposits. This alluvial plain includes a set of nested paleo-terraces from east to west, more or less eroded by the two main rivers (the Golu and the Bevincu rivers).

Agriculture and, increasingly since the 1950's, urbanization are the two main types of land use on the alluvial plain. Moreover, tourism has become an important component of the local economy, putting a lot of pressure on the environment, particularly during the summer periods. These combined anthropogenic influences have led to the eutrophication of the Biguglia lagoon (Département de la Haute-Corse,

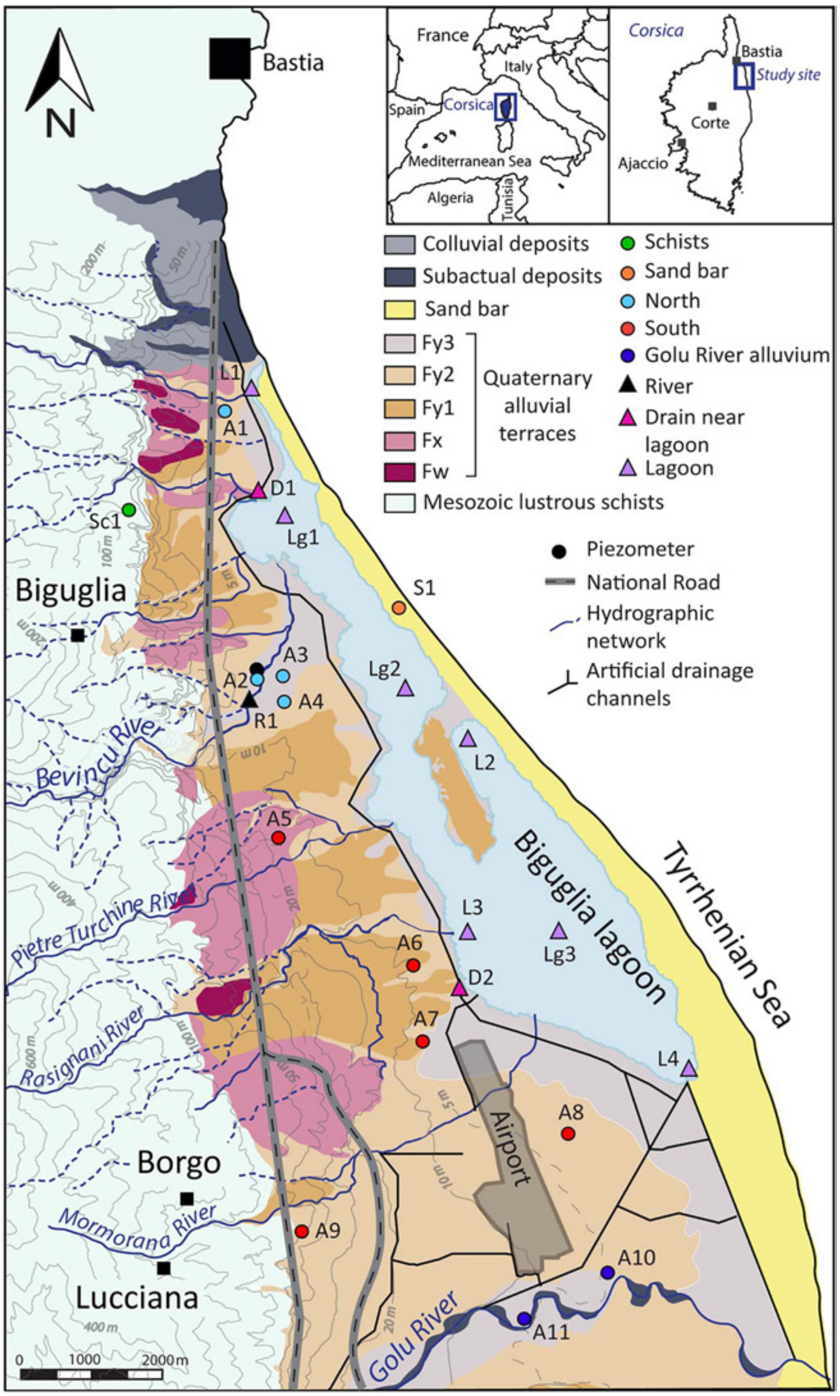

Fig. 1. Study area with simplified presentation of the regional geology and sampling sites. The study area is divided in 5 sectors to facilitate understanding of the hydrogeological setting: Schist formation (Sc 1), Sand bar (S1), North (A1–A4), South (A5–A9) and Golu River alluvium (A10–A11). Surface waters were also sampled in the Bevincu River (R1), in and around the lagoon (Lg1–Lg3 and L1–L4 respectively), and in the drainage channels near the lagoon (D1–D2).

2012; Garrido et al., 2016; Pasqualini et al., 2017). In the attempt to preserve this coastal ecosystem, the lagoon has been recognized as a RAMSAR site (Convention on Wetlands of International Importance especially as Waterfowl Habitat, 1971) in 1991, and is classified as a Nature Reserve since 1994.

2.2. Climatology and hydrology

The island of Corsica is characterized by a typical Mediterranean climate. The mean annual temperature and precipitation measured at the Bastia Météo France Station are 15.8 °C and 768 mm, respectively (average calculated between 1950 and 2016). The catchment of the study area is drained by one major river, the Bevincu River in the North, and several smaller tributaries during the rainfall periods. The regional hydrology is strongly impacted by man-made hydraulic infrastructures. For example, in the early 20th century, an artificial drainage network (Fig. 1) was constructed to collect surface groundwater in the western part of the alluvial plain in order to use the land for agricultural activities and for mosquito control. Five pumping stations, located all around the lagoon, lower the water table and reinject freshwater into artificial drainage channels directed towards the lagoon. The south of the alluvial plain is delimited by the Golu River, one of the main rivers in Corsica. It originates 2525 masl in the Hercynian Massifs of inner Corsica, and its catchment extends over 926 km^2. Naturally, it would not be part of the lagoon's watershed, however, an active artificial drain links the river to the lagoon in the South (Fig. 1).

2.3. Geology and hydrogeology

The Marana-Casinca alluvial plain consists of Quaternary sediment deposits from lustrous schist erosion products, transported by the Golu River, the Bevincu River and smaller, transient tributaries. The alluvial terraces were formed during the last glacial phases and were altered during interglacial periods (Conchon, 1972, 1978). Independent of their age, all terraces are composed of gravels, pebbles or blocks, in a sandy-loamy matrix. The aquifer, as it is today, has developed mostly in Middle and Upper Wurmian alluvial deposits (Fy2 and Fy3, Fig. 1), and is perched on an impermeable clay-deposit layer that formed during the Lower Wurmian (Fy1). The alluvial plain represents the main aquifer system of the region and includes the Bevincu and Golu aquifers. The total area of the shallow aquifers is estimated at 80 km^2, with a thickness of 3 to 40 m, and an average hydraulic conductivity around 10^{-3}–10^{-4} m/s (Orofino et al., 2010). The Bevincu aquifer is used for the drinking water supply of the Bastia urban area, while the Golu aquifer is less exploited, mostly used for irrigation. Piezometric records (piezometer 11071X0062/CASATO; http://www.ades.eaufrance.fr/) indicate that groundwater level fluctuations are closely related to variations in precipitation (Fig. 2), indicating a rather short response time and good connectivity to the surface. The seasonal groundwater level fluctuates by up to 2 m. The piezometric map, based on piezometric measurements at the boreholes/groundwater wells investigated in this study (Sc1, A1–A11) (Fig. 3), shows the groundwater levels and flow towards the lagoon during the high-water period (April 2015), with flow gradients between 3 and 5‰ (Orofino et al., 2010; Garel et al., 2016). Exact estimates are difficult, yet there is no doubt that the aquifer contributes markedly to the freshwater input of the lagoon, making the latter an important coastal groundwater-dependant ecosystem (GDE).

3. Sampling and analytical methods

3.1. Sampling and geochemical analyses

A comprehensive sampling and measurement campaign was conducted during the high-water period between May 19 and May 23, 2016. Geochemical analyses included the determination of major ion

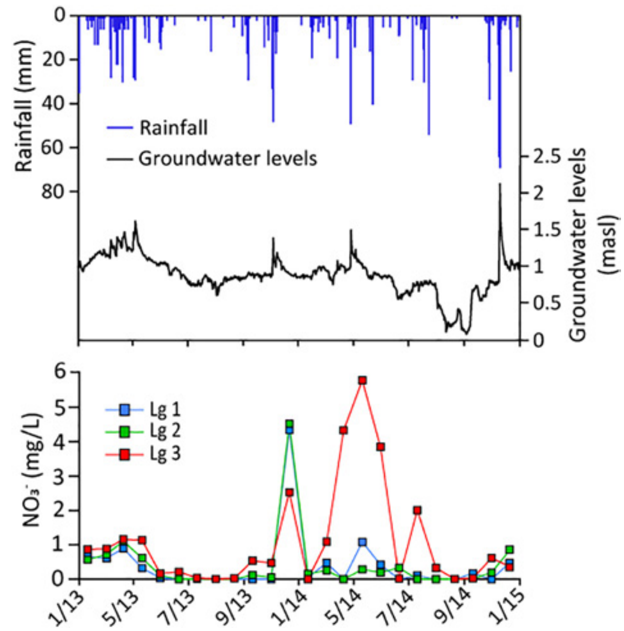

Fig. 2. Groundwater levels, rainfall (in mm), and NO_3^- concentrations in the lagoon, from January 2013 to December 2015.

and trace element concentrations, as well as the measurement of stable isotope ratios of water ($\delta^{18}O$-H_2O and δ^2H-H_2O), nitrate ($\delta^{15}N$-NO_3^-, $\delta^{18}O$-NO_3^-) and boron ($\delta^{11}B$). A total of 20 sites distributed more or less evenly over the entire alluvial plain were sampled: 3 piezometers,

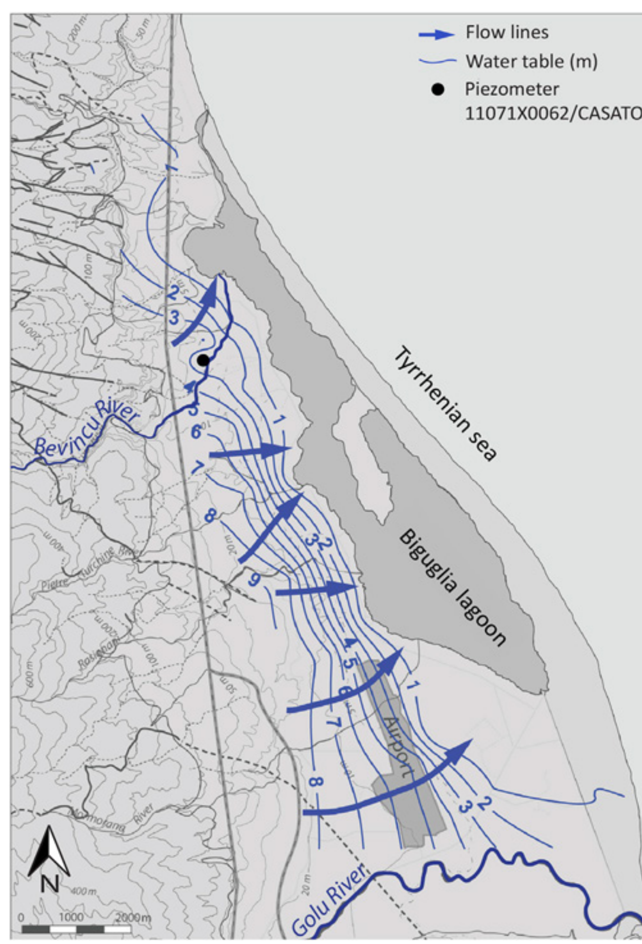

Fig. 3. Spatial distribution of aquifer water levels in the study area.

4 boreholes, 6 wells, 4 sampling sites in the lagoon, 2 sampling sites in the artificial drain near the lagoon, and 1 sampling site in the Bevincu River. The groundwater samples were collected using Grundfoss MP1 (A4, A10) or Comet submersible pumps (A5, A7, A8 and A9) and a nylon tube, or directly at the tap avoiding any air contamination (Sc1, A1, A2, A3, A6, A11 and S1). Prior to sampling, groundwater was pumped until physico-chemical parameters (i.e., T, pH) had stabilized. In addition, a monthly monitoring campaign dedicated to the analysis of NO_3^- concentrations was conducted in the lagoon (Lg1, Lg2 and Lg3) between January 2013 and December 2015. All samples for major cation and anion analysis (Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-}) were filtered through 0.45 μm nitrocellulose membrane filters, collected in two 50 mL polyethylene bottles and stored at 4 °C.

In situ physico-chemical parameters (Electrical conductivity (EC), Temperature (T), pH and dissolved O_2) were measured using a WTW 3310 Conductivity meter, a WTW 3310 pH meter and a WTW 3310 IDS Oxymeter. Alkalinity measurements were performed in the field using a HACH digital titrator. The concentration of dissolved major ions was determined at the Hydrogeology Department (CNRS UMR 6134 SPE), University of Corsica, France, using a Dionex ICS 1100 chromatograph. Water samples for trace element analysis were filtered through 0.20 μm nitrocellulose membranes, acidified using ultrapure HNO_3, and collected in 50 mL polyethylene bottles before stored at 4 °C. Trace element analyses were carried out at the AETE technical platform, University of Montpellier, France, using a Q-ICPMS X series II Thermo Fisher, with an analytical precision better than 8%.

3.2. Isotope analysis

Samples for the analysis of stable isotopes of the water molecule were collected in 20 mL amber glass bottles. δ^2H-H_2O and $\delta^{18}O$-H_2O measurements were performed using a laser-based liquid-vapour stable isotope analyser DLT-100 (Los Gatos Research) at the Hydrogeology Department (CNRS UMR 6134 SPE), University of Corsica, France. Analyses were conducted based on the analytical scheme recommended by the IAEA (Penna et al., 2010). The analytical precision was better than 1‰ for δ^2H-H_2O and 0.1‰ for $\delta^{18}O$-H_2O.

Samples for $\delta^{15}N$-NO_3^- and $\delta^{18}O$-NO_3 were filtered through 0.20 μm nitrocellulose membrane filters, collected in 50 mL polyethylene bottles, and frozen. Analyses of nitrate (N) and oxygen (O) isotope ratios were carried out at the Department of Environmental Science, University of Basel, Switzerland, using the denitrifier method (Sigman et al., 2001; Casciotti et al., 2002; Casciotti and McIlvin, 2007). Briefly, 20 nmol of NO_3^- were converted to N_2O by cultured denitrifying bacteria (Pseudomonas chlororaphis ATCC 43928 and P. chlororaphis ATCC 13985), which lack the nitrous oxide reductase enzyme. The produced N_2O was the automatically purged, purified and analyzed using an isotope ratio mass spectrometer (Thermo Finnigan DELTA^{plus} XP). Blank contribution was generally <0.3 nmol (1.5% of the sample). Oxygen isotope exchange with H_2O during the reduction of NO_3^- to N_2O was corrected for according to Casciotti et al. (2002), and was never higher than 5%. $\delta^{15}N$-NO_3^- and $\delta^{18}O$-NO_3 values are referenced relative to atmospheric air (AIR) and standard mean ocean water (VSMOW), respectively. Isotopic values were normalized using internal, as well as international KNO_3^- reference materials with reported $\delta^{15}N$-NO_3^- and $\delta^{18}O$-NO_3 values of 4.7‰ and 25.6‰ (IAEA-N3) and −1.8 ‰ and −27.9‰ (USGS 34), respectively (Gonfiantini et al., 1995; Wenk et al., 2014). All samples were analyzed in duplicates or triplicates. The reproducibility based on repeat standard and sample measurements was generally better than 0.3‰ for $\delta^{15}N$-NO_3^- and better than 0.5‰ for $\delta^{18}O$-NO_3.

Water sample for the analysis of B isotope ratios were collected in 250 mL polyethylene bottles and stored at 4 °C. Chemical separation of B was performed using IRA columns, and B isotope measurements were carryout out using a ThermoScientific Neptune Plus inductively coupled mass spectrometer (MC-ICP-MS) at the ALS Scandinavia AB Laboratory, Sweden. NIST SRM 951 was used as B isotopic standard to correct for instrumental mass bias (Lemarchand et al., 2002; Guerrot et al., 2011). See ALS website (https://www.alsglobal.se/en) for details.

3.3. Tritium and CFCs analysis

Water samples for tritium analyses were collected in 500 mL polyethylene bottles and analyzed by liquid scintillation counting (Thatcher et al., 1977) after electrolytic enrichment (Kaufman and Libby, 1954) at the Hydrogeology Department of the University of Avignon, France. Tritium concentration is expressed in TU (tritium units).

Water samples for chlorofluorocarbon (CFC) analyses were collected in stainless-steel ampoules after rinsing them with at least three times the sample volume, without any contact with atmospheric air during sampling. CFCs concentrations used for the calculation of groundwater residence times were measured using a gas chromatograph with an electron capture detector GC-ECD (Labasque et al., 2006; Ayraud et al., 2008), at the Plateforme CONDATE eau (OSUR, University of Rennes 1, France). The concentrations of three CFCs (CFC-11, CFC-12 and CFC-13) were measured with an analytical uncertainty of 1% (Labasque et al., 2006; Ayraud et al., 2008). CFC concentrations were used to calculate atmospheric mixing ratios (pptv), and compared to the atmospheric evolution curve (NOAA HATS program) to determine the apparent groundwater piston age. For each sampling point, three hypothetical lump parametric models, a piston flow model (PFM), an exponential model (EM), and binary mixing model (BMM), were systematically tested to determine the apparent groundwater residence time (Maloszewski and Zuber, 1996). The overall uncertainty of derived groundwater age estimates, including sampling biases and analytical errors, as well as errors that result from uncertainty of the recharge temperature, under the consideration of dispersion-adsorption effects, is estimated to be ±3 a (Labasque et al., 2006).

Noble gases were extracted using a head-space technique with a He gas phase, and dissolved Ne and Ar concentrations were determined by micro-gas chromatography (CG 3000, SRA instrument). The analytical error for Ne and Ar measurements was 3% and <2%, respectively. The analysis of Ne/Ar ratios allows calculating excess air and recharge temperatures (Heaton and Vogel, 1981). Excess N_2 is reported with respect to the equilibrium N_2 concentration value at 12 °C and an atmospheric pressure at 10 masl.

3.4. Land-use data

Land-cover/-use changes since the beginning of the 20th century were assessed based on available data for the watershed of the Biguglia Lagoon. The CORINE land-cover program established a land-cover map for the study site for the years 1990, 2006 and 2012. For these numerical vector maps, data were georeferenced with Lambert 93 coordinates, and we considered 4 generic land-cover types, as defined by the CORINE land-cover program: urbanized areas (continuous and discontinuous urban fabric, industrial areas, roads and airport), agricultural areas (orchards, vineyards, crops, grasslands under agricultural use), natural vegetation (forest, shrubland, grassland) and wetlands (coastal lagoon, marshes, bogs, beaches and sand). Natural areas occur mainly in the North and the Western portions of the catchment, covered by forests and shrubland. Urbanized areas are widespread throughout the alluvial plain, and particularly to the South of the plain, where urban development is still ongoing. An earlier survey in the Biguglia watershed published by the French fiscal services (Source: *Archives départementales de Haute-Corse*; Corvol (1999)), provided additional information on land-use for the year 1913, but in a non-spatially explicit format. Here, three generic land-cover types were considered: urbanized areas, agriculture areas and natural vegetation.

4. Results and discussion

4.1. Field parameters and hydrochemistry

The groundwater temperature ranged from 13.8 to 19.0 °C (mean 16.9 °C) and closely followed the annual air temperature trends in Bastia (mean 15.8 °C, Météo France). Water temperatures in the lagoon during the sampling campaign ranged from 20.9 to 25.5 °C, depending on the depth of sampling. Oxygen saturation ranged between 0.9% and 82.0%, with an average of 40.6%. Some groundwater samples (A4, Sc1, S1 and A5) were characterized by very low oxygen saturation levels (1–14%), yet throughout most of the aquifer, the oxygen saturation ranged from 45% to 82%. The correlation of groundwater and atmospheric temperatures in combination with mostly high oxygen saturation levels (indicating rapid heat and gas exchange) is consistent with a shallow, unconfined type of aquifer. Groundwater and fresh surface water displayed EC values between 225 μS/cm and 5710 μS/cm, with a median value of 428 μS/cm. Brackish water (from the lagoon and the drain near the lagoon) showed EC values from 1687 to 28,400 μS/cm with an average of 23,300 μS/cm. The pH in the groundwater varied from 6.2 to 8.1, with more elevated pH values derived from the North of the watershed, where mostly calcareous schist crops out. The pH in the lagoon is slightly more alkaline (from 7.8 to 8.9).

Highest groundwater temperatures (19 °C) at Sc1 were associated with a high EC (819.0 μS/cm) and low oxygen saturation (1.1%) indicating that at this borehole (90 m deep) groundwater is disconnected from the (near-) surface hydrological dynamics. The lowest temperatures (A11, 13.8 °C) and EC values (A11, 1224 μS/cm and A10, 311 μS/cm) were measured in groundwater located near the Golu River. The Golu River has its source at high altitude and the low mineral content is consistent with the geology of the upper watershed. Particularly in the South, Golu River water contributes significantly to aquifer recharge, and infiltration of its cold and relatively low ion-strength water tends to lower the EC and temperature of the alluvial aquifer. Groundwater from the sand bar (S1) shows the most elevated EC values (5710 μS/cm), due to high concentrations in Cl^-, Na^+, Mg^{2+} and Ca^{2+} from the admixture of seawater. In this area, the aquifer represents a fresh water lens that is constrained by seawater on the one side and lagoonal water on the other, making it particularly susceptible to sporadic saline water intrusions.

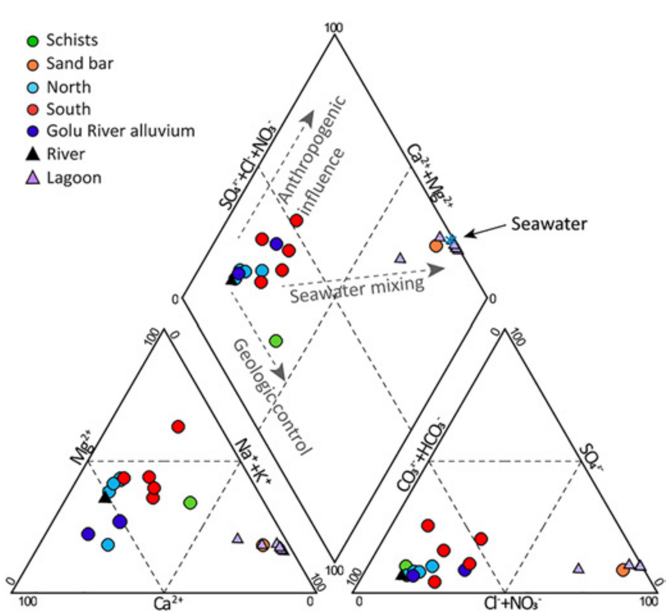

Fig. 4. Piper diagram for all water samples.

Most groundwater samples from the alluvial plain displayed a typical HCO_3^--Ca^{2+}-Mg^{2+} type hydrochemistry (Fig. 4). Groundwater from the South showed, in addition, elevated, Cl^--NO_3^- concentrations. Generally, NO_3^- levels were above the natural regional baseline (between 5 and 7 mg/L) (Appelo and Postma, 2005; Santoni et al., 2016a, 2016b) at almost all groundwater sites, indicating the anthropogenic influence throughout the alluvial plain. The HCO_3^--Na^+-K^+-type groundwater hydrochemistry within the calcareous-schist formations indicates its geologic control. Sand bar groundwater and water from the lagoon display high Cl^--Na^+-K^+ concentrations, consistent with the increased influence from seawater mixing (Fig. 4).

4.2. Groundwater origin

The δ^2H-H_2O and $\delta^{18}O$-H_2O values for river, ground, lagoon and sea water samples are listed in Table 1. The isotopic signature of groundwater ranges between −9.11‰ and − 1.21‰ for $\delta^{18}O$-H_2O and between −59.9‰ and −11.5‰ for δ^2H-H_2O (Fig. 5), a typical range along the fresh- to seawater continuum. The O and H isotopic signatures of the freshwater samples are close to the Global Meteoric Water Line (GMWL) proposed by Craig (1961), and to the Western Mediterranean Meteoric Water Line (WMMWL) from Celle-Jeanton et al. (2001). The water stable isotopic signatures allow discriminating between two major water recharge processes/sources within the floodplain. Firstly, groundwater from the northern part of the alluvial plain (A1-A4) is characterized by $\delta^{18}O$-H_2O values between −8.11‰ and −7.06‰ and δ^2H-H_2O values from −48.9‰ to −41.3‰, consistent with a high-altitude water source. For comparison the $\delta^{18}O$-H_2O and δ^2H-H_2O from rainwater at the Corte station (450 masl) is −7.27‰ and −47.7 ‰ respectively (Huneau et al., 2015). Groundwater from the southern part of the catchment (A5-A9) is comparatively enriched in ^{18}O-H_2O and ^2H-H_2O ($\delta^{18}O$-H_2O between −7.00‰ et −6.61‰, δ^2H-H_2O between −42.6‰ and −39.0 ‰), close to the weighted mean isotopic rainfall isotope signature at the Bastia station (1 masl, −6.06‰ and −36.1‰, respectively) (Huneau et al., 2015). Finally, groundwater from the most southern sites (A10 and A11) located in the Golu river alluvium, again displays very low water isotope values (−8.00‰ and −9.11‰ for $\delta^{18}O$-H_2O and −49.9 ‰ and −59.9‰ for δ^2H-H_2O, respectively). This relative depletion in the heavier water isotopes is indicative of groundwater recharge with contribution from an allochthonous, higher-altitude water source, i.e., from the headwater catchment. Hence, while in the south of the alluvial plain, groundwater is recharged mainly from local sources, groundwater in the North of the alluvial aquifer and close to the Golu River is primarily recharged with water from higher up in the catchment.

The $\delta^{18}O$-H_2O and δ^2H-H_2O signatures of the water in the lagoon range from −6.83‰ to −1.21‰ and from −40.7‰ to −11.6‰, respectively. The water isotope data fall slightly below the seawater-freshwater mixing line, indicating evaporative isotope enrichment within the lagoon (Gemitzi et al., 2014).

4.3. Groundwater residence time and mixing processes

Tritium concentrations of groundwater ranged between 2.2 ± 0.2 to 4.3 ± 0.3 TU, with an average of 3.2 ± 0.3 TU (Table 2). The ^3H concentrations provide qualitative information of groundwater residence time. Low ^3H concentrations (<2 UT) indicate a long residence time, >60 years, high concentrations (>4 UT) suggest a more recent infiltration (<60 years), and intermediate concentrations result from mixing processes. In order to independently validate and precise the relative age, ^3H concentrations were compared to CFC-based ages. Indeed, a binary mixing model with an "old" groundwater end-member without any anthropogenic CFCs (i.e., recharged before the 1950's), and a "young" groundwater end-member recently infiltrated (i.e., recharged in 2016) allows us to estimate the apparent age of groundwater. Thus, for A3, A4, A6 and A10, ^3H concentrations can be correlated to CFC-

Table 1
Type of sampling site, localization, sampling depth and results from geochemical and isotopic analyses for the different wells/sampling sites (April 2016 campaign).

Sample ID	Type	Localization (Lambert 93)		Altitude (masl)	Depth (m)	T (°C)	O_2 (%)	pH	EC (μS. cm^{-1})	HCO_3^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	$\delta^{15}N$-NO_3 (‰)	$\delta^{18}O$-NO_3 (‰)	^{11}B (‰)	δ^2H (‰)	$\delta^{18}O$ (‰)
Sc1	Borehole	1,227,113.3	6,192,715.3	80	90	19.0	1.1	7.0	819	462	42.0	2.7	84.8	3.0	42.7	48.8	5.9	1.0	−3.6	−41.5 ± 0.1	−7.15 ± 0.01
A1	Borehole	1,228,441.0	6,194,233.9	37	–	16.5	70.1	7.0	474	196	31.0	7.3	28.3	1.7	13.7	73.9	6.4	3.6	26.1	−41.3 ± 0.3	−7.06 ± 0.09
A2	Well	1,228,907.9	6,190,418.8	–	–	17.0	29.6	6.9	439	214	23.0	11.8	16.4	0.7	25.0	37.3	4.5	3.4	18.2	−44.8 ± 0.3	−7.39 ± 0.02
A3	Well	1,229,725.9	6,189,979.6	2	14	16.3	82.0	7.4	367	166	19.0	2.8	12.4	0.6	21.0	35.8	5.3	1.2	23.6	−48.5 ± 0.1	−7.95 ± 0.02
A4	Piezometer	1,229,094.0	6,189,984.0	11	50	17.8	0.9	7.9	345	179	18.0	1.9	10.3	0.7	16.3	31.2	10.6	6.3	21.2	−48.9 ± 0.2	−8.11 ± 0.03
A5	Well	1,229,156.1	6,188,083.4	175	7	14.1	13.8	6.4	523	215	20.0	6.5	17.6	2.5	29.4	44.9	11.9	9.4	23.0	−42.6 ± 0.1	−6.71 ± 0.04
A6	Borehole	1,231,088.1	6,186,248.4	8	23	17.9	64.0	6.9	413	188	28.0	17.9	21.3	1.6	22.9	27.4	5.1	2.6	26.2	−40.8 ± 0.2	−6.94 ± 0.09
A7	Well	1,231,209.0	6,185,141.0	9	9	16.5	56.7	7.3	428	134	29.0	–	22.8	4.3	21.1	28.0	7.3	4.3	26.4	−40.0 ± 0.1	−6.61 ± 0.06
A8	Piezometer	1,233,242.0	6,183,809.4	2	12	17.8	50.7	6.5	384	101	24.0	26.8	24.1	2.3	36.3	15.5	5.4	3.5	36.1	−39.0 ± 0.2	−6.63 ± 0.05
A9	Well	1,229,490.7	6,182,415.9	45	12	16.8	65.6	8.1	689	270	38.0	29.8	49.4	3.7	37.6	57.3	11.5	7.0	23.5	−42.0 ± 0.1	−7.00 ± 0.03
A10	Piezometer	1,234,045.0	6,181,835.0	4	19	18.5	44.7	6.2	311	111	22.0	23.8	18.4	1.8	12.9	43.9	8.7	4.5	34.0	−49.9 ± 0.2	−8.00 ± 0.02
A11	Well	1,232,652.8	6,181,163.4	8	–	13.8	45.7	6.7	224	105	9.1	7.5	7.5	0.9	6.8	32.7	8.8	5.9	24.8	−59.9 ± 0.2	−9.11 ± 0.04
S1	Borehole	1,230,919.2	6,191,414.1	1	–	17.6	3.5	7.5	5710	244	1700	25.4	950	73	130	100	19.7	9.6	33.8	−34.7 ± 0.2	−5.83 ± 0.03
R1	River	1,228,766.8	6,190,080.5	4	–	16.8	99.8	8.1	321	161	15.0	0.0	9.8	0.6	15.1	35.4	–	–	26.4	−47.6 ± 0.1	−7.86 ± 0.05
L1	Lagoon	1,228,788.2	6,194,529.8	0	–	22.7	78.2	7.8	27,900	174	10,000	0.0	6500	180	710	230	–	–	–	−13.6 ± 0.7	−1.38 ± 0.08
L2	Lagoon			0	–	25.5	–	8.9	28,400	133	10,000	0.0	6600	170	680	220	–	–	–	−16.6 ± 0.3	−1.92 ± 0.02
L3	Lagoon	1,231,845.6	6,186,731.0	0	–	23.3	95.1	8.1	26,500	129	9600	0.0	6200	170	700	230	–	–	–	−33.6 ± 0.3	−5.69 ± 0.04
L4	Lagoon	1,235,023.8	6,184,716.8	0	–	22.8	74.7	7.8	24,500	116	9300	0.3	5400	170	690	240	7.3	5.3	–	−40.7 ± 0.1	−6.83 ± 0.03
D1	Drain near lagoon			0	–	22.8	114.8	8.2	26,200	150	9400	1.1	6000	170	700	230	12.2	5.7	37.0	−13.2 ± 0.7	−1.64 ± 0.24
D2	Drain near lagoon			0	–	18.3	64.4	7.1	1687	210	390	18.4	280	4	47	60	6.6	6.3	–	−11.6 ± 0.2	−1.21 ± 0.11

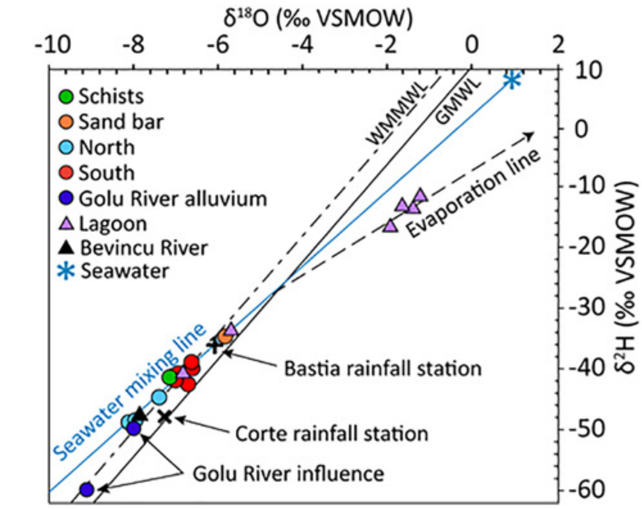

Fig. 5. δ^2H-H_2O vs $\delta^{18}O$-H_2O plot with data for seawater, groundwater and weighted mean values for rainwater collected at Corte and Bastia. Also shown is the Global Meteoric Water 943 Line (GMWL) from Craig (1961) and the Western Mediterranean 944 Meteoric Water Line (WMMWL) from Celle-Jeanton et al. (2001).

based ages, and linear regression then yields groundwater residence time for each groundwater sample (Table 2).

The binary mixing model based on CFC concentrations also allows estimating the relative contribution of water recently infiltrated (e.g. from the river), versus water with longer residence time (e.g., from the schists formation) (Table 3). Given the obvious links between origin and age of the different water masses, additional, independent information on the partitioning between different water sources (and hence on the mixed age) can be obtained using a geochemical binary mixing model considering Ca + Mg/Na + Cl ratios (Table 3). Indeed, the hydrochemical fingerprints with regards to Mg^{2+}, Ca^{2+}, Na^+ and Cl^- concentrations are quite distinct for surface water (low ionic strength) on the one hand, and groundwater from the schist formations (higher ionic strength) on the other hand, permitting the discrimination between the two end-members (and estimates on respective relative contributions).

Comparatively low ^3H concentrations (from 2.2 TU to 3.0 TU), corresponding to the longest mean residence times (from 20 years to 45 years), are measured in groundwater from the South of the floodplain (A6-A8), the sand bar (S1), and the schist formation (Sc1). The sample from the schist formation (Sc1) displays one of the lowest ^3H concentrations (2.3 ± 0.2 TU) indicating the long residence times for water resources from the relatively impervious sediments, with minimal connectivity to surface water. Binary mixing modelling for A6 indicated a comparatively low contribution from modern water, between 23% and 34%. Similarly, A7 and A8 groundwater is essentially disconnected from the inflow of surface water, as indicated by the combined ^3H, CFC, and hydrochemical data, suggesting an important contribution of old groundwater of at least 60 years (Table 2). In this context, A5 appears as an exception with comparatively high ^3H concentrations (4.2 TU). A greater influence of short transit-time surface water is very likely here, considering the vicinity to the Pietre Turchine River.

Table 2
Concentrations of ^3H (in TU) and calculated groundwater residence time based on a linear regression between CFC piston flow model ages (recharge temperature of 12 °C and elevation of 10 masl) and ^3H concentrations.

Sample ID	^3H (TU)	Calculated residence time (years)
Sc1	2.3 ± 0.2	41
A1	3.8 ± 0.3	9
A2	3.1 ± 0.3	16
A3	3.1 ± 0.3	16
A4	3.7 ± 0.3	10
A5	4.2 ± 0.3	5
A6	2.2 ± 0.2	44
A7	3.0 ± 0.3	23
A8	2.6 ± 0.2	34
A9	3.1 ± 0.3	16
A10	3.0 ± 0.3	23
A11	4.3 ± 0.3	4
S1	2.9 ± 0.3	26

Groundwater from the North (A1–A4) displays markedly higher ^3H concentrations (from 3.1 TU to 3.7 TU), in the same range as the mean weighted ^3H content of water at the Corte rainfall station (3.8 TU from September 2016 to March 2017). Intermediate ^3H concentrations between 3 and 4 TU indicate mixing between groundwater with longer residence time and groundwater that infiltrated more recently (i.e., with a low water residence time between 9 and 16 years). Based on binary mixing model calculations for A3 and A4, between 75% and 83%, and between 73% and 84%, respectively, of the groundwater recharge can be attributed to the infiltration of river water (Table 3).

Finally, the two samples located in the Golu River alluvium (A10 and A11) also display relatively high ^3H concentrations (3 TU and 4.3 TU respectively). In accordance with the "young" ^3H signature, the water $\delta^{18}O$-H_2O and δ^2H-H_2O values, as well as the physico-chemical parameters confirm that the Golu River contributes significantly to the groundwater recharge in this location.

Independent of their residence time, the majority of groundwater sites (except Sc1, A3, A4 and A5) display NO_3^- concentrations above the natural baseline (between 5 and 7 mg/L) (Appelo and Postma, 2005; Santoni et al., 2016a, 2016b). Groundwater with the longest residence time shows the highest NO_3^- concentrations (Fig. 8), suggesting that the high groundwater NO_3^- contents are the result of a progressive accumulation over several decades, related to both current and past anthropogenic land use.

4.4. Land use evolution and anthropogenic influence on groundwater in the past

Over the last century, the urbanized areas of the catchment have grown five-fold in size, from 4 km^2 in 1913 to 21 km^2 in 2012 (Table 4). The rate of urbanization has particularly increased since the beginning of the 21st century. Urbanization has developed mainly on the alluvial plain (Fox et al., 2012). Strong urban pressure (Robert et al., 2015), lack of urban planning (Prévost and Robert, 2016), and the construction of isolated residential areas has amplified the risk of localized pollution and leakage from the extensive sanitation network (i.e. leakage from water pipelines and septic tanks). Agricultural land-use has decreased by almost 40%, from 71,1 km^2 in 1913 to 43,9 km^2 in 2012. Shrinking of the agricultural area was accompanied by modification of agricultural practices. Over the last decades, orchard and vineyard farming was progressively replaced by cattle breeding and vegetables production. Natural areas comprising mostly forests and shrublands have increased in size by 50% between 1913 and 1990 (from 31,6 km^2 to 46,5 km^2), as result of socio-economic changes such as the abandonment of the countryside and mountain areas (Baessler and Klotz, 2006; Geri et al., 2011). Since 1990, forest and shrubland areas have remained more or less constant.

NO_3^- concentrations vary from 2 to 30 mg/L with an average of 14 mg/L (Table 1), always below the drinking standard (50 mg/L). Most strikingly, a marked difference between nitrate concentrations in groundwater from the North (A1-A4) and from the South (A5-A9) of the watershed was observed. Towards the South, average NO_3^- concentrations (22.0 mg/L) are four times higher than in the northern parts (5.5 mg/L). NO_3^- concentrations in the lagoon and in river water were very low (<1 mg/L) during the sampling campaign in May 2016. However, the NO_3^- survey conducted in 2013 and 2014 revealed a NO_3^-

Table 3
Concentrations of CFCs in groundwater samples, apparent groundwater age, and proportions of recent versus old water based on binary mixing model calculations (recharge temperature of 12 °C and elevation of 10 masl).

Sample ID	CFC-11		CFC-12		CFC-113		Apparent ages	Binary mixing model		N_2	N_2 excess
	pmol/kg	pptv	pmol/kg	pptv	pmol/kg	pptv	years	CFCs	(Ca + Mg)/(Na + Cl)	mol/L	%
A3	8.48	454.21	3.28	669.79	0.31	53.90	16	Old +75% of recent water	Schist GW + 83% of river water	6.2E−04	−1.3
A4	2.54	135.83	2.13	435.18	0.53	92.03	10	Old +72% of recent water	Schist GW + 73% of river water	6.5E−04	+3.7
A6	1.05	56.08	0.91	184.83	0.17	29.06	44	Old +34% of recent water	Schist GW + 23% of river water	7.1E−04	+11.7
A10	13.46	721.50	15.35	3132.37	0.27	46.90	23	Old +66% of recent water	Schist GW + 52% of river water	6.8E−04	+7.9

peak in May 2014 in the southern basin of the lagoon, during the high-water period (Fig. 2). It appears that the sporadic increase in lagoonal NO_3^- concentrations is most often associated with strong episodic rainfall events, bringing in large amounts of nitrate leached from the watershed, via the aquifer. Indeed, comparison with groundwater piezometric levels support that the main NO_3^- inputs to the lagoon are from the groundwater, hydraulically connected to the lagoon (Garel et al., 2015). Groundwater, therefore, has a predominant impact on NO_3^- supplies to the lagoon.

Elevated NO_3^- concentrations by themselves represent an unequivocal indicator of the ongoing aquifer water quality deterioration due to anthropogenic influence. In addition, nitrate isotope ratios can be used to further identify the origin and fate of contaminant sources (Widory et al., 2005; Pastén-Zapata et al., 2014; Zhang et al., 2015; Wang et al., 2017). The dual isotope approach to trace nitrate contamination is based on the fact that nitrate from different origins have distinct isotopic signatures. A bi-plot of $\delta^{18}O\text{-}NO_3^-$ vs $\delta^{15}N\text{-}NO_3^-$ was constructed, adopting typical end-member source isotopic "fingerprints" from the literature (Kendall, 1998; Kendall et al., 2008; Xue et al., 2009) (Fig. 6). Values for $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ ranged from 4.5 to 19.7‰ and from 1.0 to 9.6‰, respectively (Table 1). Highest $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ were observed in groundwater from the sand bar. When using nitrate N and O isotope signatures to constrain nitrate contamination sources, it is important to understand that both N and O isotopic source signatures can be modified by biological processes. For example, partial nitrate loss by microbial denitrification causes the $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ values to increase from its primary source values. While the nitrate N:O enrichment ratio in association with denitrification ($\delta^{15}N\text{-}NO_3^-/\delta^{18}O\text{-}NO_3^-$) can vary (Böttcher et al., 1990; Cey et al., 1999; Lehmann et al., 2003; Granger et al., 2004; Panno et al., 2006), probably as function of the relative importance of co-occurring nitrate regeneration (Lehmann et al., 2003; Wenk et al., 2014; Granger and Wankel, 2016), a linear, concomitant increase in nitrate $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ has been considered indicative of microbial denitrification in groundwater. Indeed, the correlation between $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ ($r^2 = 0.7$) suggests that denitrification occurs to some extent in the studied aquifer where O_2 is low. This conclusion is also supported by excess N_2 concentrations of up to 10% at some of the wells (Table 3). While a clear link between low O_2 levels (i.e., reducing conditions) and excess N_2 concentrations did not exist, it is plausible to assume that at already low O_2 saturation levels, O_2 can further be reduced to suboxic levels in microenvironments.

Table 4
Land use evolution of the Biguglia lagoon catchment from 1913 to 2012 (CORINE land-cover program and *Archives départementales de Haute-Corse* (Corvol, 1999)).

Land use	1913	1990	2006	2012	Surface evolution (%)
					1913–2012
Urbanization (km^2)	4.1	15.8	18.5	21.0	+519.5
Agriculture (km^2)	71.7	43.9	42.7	43.9	−61.2
Forest and shrublands (km^2)	31.6	46.5	45.0	46.5	+147.0

Denitrification in sandy marine sediments is very common (Kaspar, 1982; Devol, 2015; Kirstin and Bo, 2016), and we have no doubt that the highest nitrate $\delta^{15}N\text{-}NO_3^-$ values at S1 (in the sand bar) can be attributed to N and O isotope fractionating effects during partial denitrification. While to some extent denitrification (in situ, or by exchange with actively denitrifying environments) has likely impacted also the groundwater nitrate pool at the other sites, we argue that the overall nitrate elimination potential of the aquifer is rather low, and that the isotopic imprint of denitrification was, if at all, rather weak (Fig. 6). Assuming that denitrification within the aquifer of our study region is of minor importance, we consider the nitrate isotope data reliable indicators of the original nitrate source.

The measured nitrate isotope values are relatively well constrained within the $\delta^{15}N\text{-}NO_3^-$-vs- $\delta^{18}O\text{-}NO_3^-$ space. More precisely, the isotope signatures fall within the range reported for soil organic N and manure/sewage (Fig. 6). Hence, within the watershed, sewage from leaky septic waste system appears as the principal anthropogenic N pollution source (besides soil N), while aquifer contamination by NO_3^- originating from artificial fertilizer only plays a marginal role. This is consistent with the fact that urban land-use is playing a predominant role today and in the more recent past.

Within the lagoon, the relatively high nitrate $\delta^{15}N\text{-}NO_3^-$ values at low NO_3^- concentrations may partly be due to phytoplankton uptake in the lagoon, but are also consistent with a significant fixed N contribution from groundwater-derived sewage contamination. Indeed, anthropogenic impacts and the on-going deterioration of the water quality of the lagoon has also been observed by others (Mouillot et al., 2000; Garrido et al., 2016; Pasqualini et al., 2017).

To further verify the proposed anthropogenic influence as highlighted by the N isotope measurements, we also analyzed $\delta^{11}B$ (Seiler, 2005; Widory et al., 2005). The use of B isotopes as contamination indicator is based on the fact that different sources have a distinctive B isotopic signature. For example, the $\delta^{11}B$ of seawater is 39‰ (Foster et al., 2010), while that of average continental crust is 0 ± 5‰ (Vengosh et al., 1994). Sewage-derived B sources were found to have $\delta^{11}B$ values ranging from 5 to 13‰, significantly different from uncontaminated groundwater (~30‰) and seawater. Hence, groundwater contaminated by recharge of treated sewage will yield distinctive $\delta^{11}B$ signatures in between the end-member values. The $\delta^{11}B$ values observed in this study ranged from −3.6‰ to 37.0‰ (Table 1), yet, the majority of the groundwater $\delta^{11}B$ isotopic signatures fell between 18 and 27‰, which suggests a certain degree of groundwater contamination or mixing with non-marine natural B from the crust (Vengosh, 1999).

The combined isotopic signatures of $\delta^{11}B$ and $\delta^{15}N\text{-}NO_3^-$ allow us to even better delineate the different groundwater contamination sources, and to semi-quantify the degree of contamination. Firstly, groundwater from the southern portions of the floodplain, near the coast, from the sand bar and in water from the lagoon (A8, A10, S1 and D1), was more enriched in ^{11}B ($\delta^{11}B$ from 33.8‰ to 37‰), indicating significant marine influence (Fig. 7). Secondly, groundwater samples (mostly from the Southern flood plain) displaying elevated $\delta^{15}N\text{-}NO_3^-$ values (up to 11.9‰) compared to the northern part, in combination with lowered $\delta^{11}B$ (from 26.2‰ to 23‰) and high NO_3^- concentrations (from 6.5 mg/L to 29.8 mg/L) suggest systematic contamination with

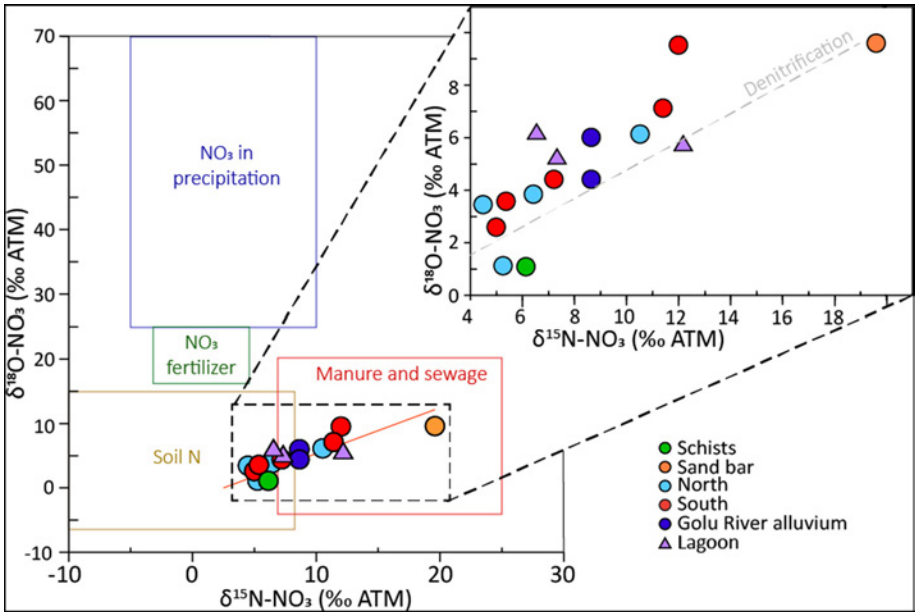

Fig. 6. δ^{15}N-NO_3^- vs δ^{18}O-NO_3^- values for all analyzed samples. Depicted are also the dominant compositional ranges of δ^{15}N and δ^{18}O data for nitrate from different sources (Kendall, 1998; Xue et al., 2009).

sewage (Vengosh et al., 1994; Widory et al., 2013) in these portions of the aquifer. Finally, mixing with pristine, old water from the schist formation seems to generate a third vector within the δ^{15}N-NO_3^--δ^{11}B biplot. Significantly lowered δ^{11}B values in combination with comparatively low NO_3^- concentrations (from 2.7 mg/L to 11.8 mg/L) and low δ^{15}N-NO_3^- values highlight the importance of groundwater recharge from the schist formation mostly at sites in the Northern part of the aquifer.

4.5. Groundwater contamination as function of time

Our data suggest that the nitrate elimination potential of the aquifer system is rather weak underlining its poor self-remediating capacity. As a consequence, given water residence times >30 years in some parts of the aquifer, today's low groundwater quality is at least in parts due to the contamination by legacy pollutants. Groundwater with a large enough hydraulic residence time can be used as valuable recorder of climatic and hydrological changes that happened in the past (Fontes et al., 1993; IAEA, 1993; Jiráková et al., 2011; Varsányi et al., 2011). Similarly, the time-lag between pollution emission and groundwater contamination, a well-known aspect in contaminant hydrology (Worrall and Burt, 1999; Wang et al., 2013; Hrachowitz et al., 2016; Vero et al., 2017), provides insight into past anthropogenic activities. More precisely, with the groundwater age constraints in hand, the isotopic and geochemical data allow us to reconstruct nitrate contamination and land use evolution though time. The extensive historical information available on land use for the study area, in turn, permits validation of such a space-for-time approach.

In order to use the isotope/hydrochemical signatures for "tagging" legacy N contamination sources in a chronological way, a graph of δ^{15}N-NO_3^- vs ^{3}H was constructed (Fig. 8). Again, a general pattern arises, in which the data points can be divided into three groups: Groundwater

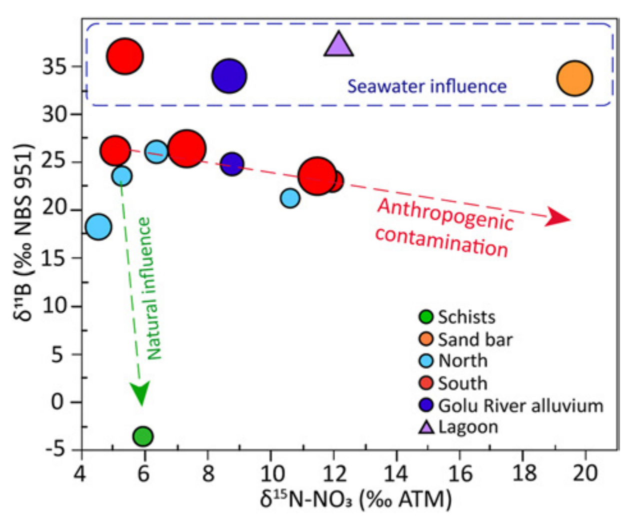

Fig. 7. δ^{11}B vs δ^{15}N-NO_3^- values for groundwater and surface water samples. Isotope compositional ranges and mixing/process vectors as presented in (Vengosh, 1999; Vengosh et al., 1994; Widory et al., 2013).

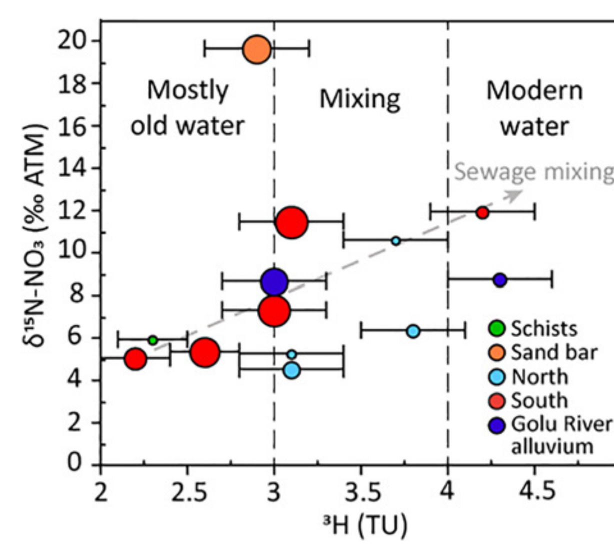

Fig. 8. δ^{15}N-NO_3^- vs ^{3}H concentrations in groundwater samples. The symbol size is proportional to NO_3^- concentrations measured in groundwater samples.

with the longest average residence time (from 2 TU to 3 TU), mostly at the Southern sites (A6, A7, A8), display the highest NO_3^- concentrations and lowest δ^{15}N-NO_3^-. The relatively low δ^{15}N-NO_3^- indicates a soil N origin. Culture type changes in the past, and massive soil remodeling for the construction of new urban areas has probably led to significant soil N mobilization in the 70/80's resulting in high fixed-N inputs to the aquifer (as indicated by the relatively high nitrate concentrations in the oldest groundwater samples) (Daum, 1997). Groundwater with an intermediate average water residence time (i.e., age) (Tables 2 and 3) represents a mix of relatively old, already anthropogenically impacted, groundwater (with high N from soil sources) and modern surface water, contaminated with sewage N (e.g., A1, A2, A3 and A4). Finally, groundwater with the shortest residence time (above 4 TU) displays the lowest nitrate concentrations and, on average, the highest δ^{15}N-NO_3^- values. This "modern" groundwater, with a mean residence time <30 years, carries an N and B isotopic tag that is indicative of recent near-surface N contamination primarily from sewage sources (i.e., leaky wastewater/sanitation network).

Our data clearly indicate that over the last decades, contamination by sewage N became increasingly important. Overall, the NO_3^- concentrations increased significantly with residence time and with the degree of mixing of pristine groundwater with contaminated water sources over the last 50 years. The progressive contamination within the aquifer, due to pollution from sources that changed over time, confirms the poor self-remediating capacity of the Biguglia groundwater system. Given the observed time lag of several decades between pollution and groundwater contamination, even a complete halt of anthropogenic fixed N inputs to the groundwater would not result in an immediate improvement of the groundwater quality. In the same vein, modern aquifer contamination will have detrimental effects on the quality of the groundwater that will be felt for several decades to come. There is no doubt that such legacy effects are problematic for the future conservation and quality management of groundwater resources and associated ecosystems.

5. Conclusion

Our study demonstrates the value of multi-isotope approaches to provide a solid basis for the identification of contaminant sources, as well as an integrated understanding of time-dependent contaminant dispersal in groundwater systems. We highlighted the close links between transport, residence time and progressive accumulation of nitrate in a coastal aquifer in Corsica (France). As of today, sewage contamination appears to be the main source of nitrate pollution to the aquifer. However, the highest nitrate concentrations in the aquifer can in large part be attributed to anthropogenic/agricultural activities in the recent past. While the relatively low nitrate contamination in modern groundwater underlines that ongoing management practices to reduce surface nitrate pollution take effect, progressive nitrate contamination in "older" portions of the aquifer indicate the poor self-remediating capacity of the system. The legacy effects of past anthropogenic pollution in groundwater described here lead to questions regarding the resilience and long-term protection of ecosystems that directly rely on the groundwater supply. Anthropogenic groundwater pollution that happened over the last decades will represent a continuous threat for such groundwater dependent ecosystems in the future. The timing of such "blasts from the past" will depend on the intrinsic hydraulic capacity of a given aquifer.

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5.3. Conclusion

Le temps de résidence des eaux souterraines permet de mieux appréhender la dynamique globale de l'hydrosystème. Mises en regard des teneurs en ^3H mesurées dans les pluies, les teneurs en ^3H des eaux souterraines apportent une information qualitative sur leur temps de séjour dans l'aquifère. Pour les eaux souterraines de la plaine de la Marana, les faibles teneurs en ^3H (<2 UT) indiquent des temps de résidence longs, supérieurs à 60 ans (eaux « anciennes »). Les fortes teneurs en ^3H (>4 UT) suggèrent elles, une infiltration plus récente, inférieure à 60 ans (eaux « récentes »). Les teneurs intermédiaires résultent des processus de mélange entre des eaux à long (> 60 ans) et à court (<60 ans) temps de séjour. Les concentrations en ^3H ne permettant pas de datation précise, celles-ci ont été corrélées avec les mesures des CFCs afin de quantifier l'âge apparent des eaux souterraines. Cette approche par régression linéaire a ainsi permis de proposer un temps de séjour calculé pour chacune des eaux souterraines échantillonnées. Les temps de séjour les plus longs, mesurés dans les eaux souterraines du centre de la plaine alluviale, du lido et des contreforts schisteux, traduisent la dynamique inertielle de ces secteurs. À l'inverse, les eaux souterraines de la partie Nord de la plaine alluviale et les alluvions du Golu se caractérisent par des temps de séjour plus faibles, liés à l'infiltration des eaux de surface en provenance du Bevincu et du Golu. Ces observations sont également confirmées par les deux modèles de mélange binaires utilisés, l'un basé sur les concentrations en CFCs et l'autre sur le rapport Ca+Mg/Na+Cl, ce dernier étant possible grâce aux concentrations ioniques très contrastées entre les eaux de surface (faible concentration ionique) et les eaux souterraines en provenance des formations schisteuses (concentration ionique élevée).

La majorité des eaux souterraines échantillonnées présente des concentrations en NO_3^- supérieures au bruit de fond géochimique naturel (5 à 7 mg/L), démontrant la présence d'une contamination anthropique diffuse sur l'ensemble de la plaine. Une différence marquée est cependant observable entre les eaux souterraines du Nord et du Sud du bassin versant. Vers le Sud, les concentrations moyennes en NO_3^- (22 mg/L) sont quatre fois plus élevées que celles mesurées dans la partie Nord (5,5 mg/L). Les eaux souterraines « récentes » montrent les concentrations en NO_3^- les plus faibles. Ces concentrations augmentent progressivement avec le temps de séjour des eaux souterraines. Ce processus d'accumulation met en évidence la **faible capacité de**

remédiation de l'aquifère vis-à-vis des contaminations en NO_3^-. En effet, bien que la dénitrification puisse probablement se produire dans certains secteurs - principalement sur le lido du fait de la nature marine des sédiments, sujets à ce type de processus – son empreinte isotopique reste faible, voire nulle, sur la majorité du bassin versant. **Les processus d'élimination des NO_3^-, marginaux voire inexistants, permettent alors leur accumulation et leur stockage dans l'aquifère au cours du temps**.

Grâce à l'usage combiné du $\delta^{11}B$, du $\delta^{15}N$-NO_3^- et du $\delta^{18}O$-NO_3^-, **deux sources de NO_3^- ont pu être identifiées : le sol et les fumiers/eaux usées.** Ainsi, pour les eaux souterraines du Sud de la plaine, ayant les temps de séjour moyens les plus longs (de 2 à 3 UT), les NO_3^- proviennent principalement du sol. Les changements des pratiques agricoles ainsi que le remodelage massif du sol pour la construction de nouvelles zones urbaines ont induit une mobilisation importante de l'azote du sol dans les années 70 et 80, provoquant par conséquent des apports élevés d'azote vers l'aquifère. Les eaux souterraines « récentes » (>4 UT) présentent des signatures isotopiques plus enrichies en ^{15}N-NO_3^- et ^{11}B. Ces observations indiquent une contamination par les flux azotés de surface, liée principalement à l'infiltration récente d'eaux usées. La forte urbanisation, le manque de plan d'urbanisme et la construction de zones résidentielles isolées au cours des dernières années ont amplifié les risques de pollutions localisées. Les eaux usées, en provenance des fuites sur le réseau d'assainissement collectif ou sur les ouvrages d'assainissement individuel, constituent actuellement la principale source anthropique de pollution azotée. Enfin, les eaux souterraines avec un temps de séjour intermédiaire (entre 3 et 4 UT) résultent d'un mélange entre des eaux souterraines « anciennes » - contaminées indirectement par les activités humaines ayant entrainées une mobilisation importante de l'azote du sol - et des eaux « récentes » contaminées par l'infiltration d'eaux usées.

Les sources de contamination azotée ont donc évolué au cours du temps (Figure 17). Les fortes concentrations en NO_3^- actuellement observées résultent de la perturbation massive des sols débutée dans les années 1960. De nos jours en revanche, la contamination par les eaux usées représente la principale source de dégradation. **L'état contemporain dégradé de la ressource découle donc en grande partie de l'héritage des pollutions liées aux politiques d'aménagement passées. Ce legs historique montre la capacité « d'archivage » des eaux souterraines vis-à-vis des activités**

humaines. L'accumulation progressive des polluants dans les eaux souterraines constitue ainsi une problématique autant sur le plan humain qu'environnemental.

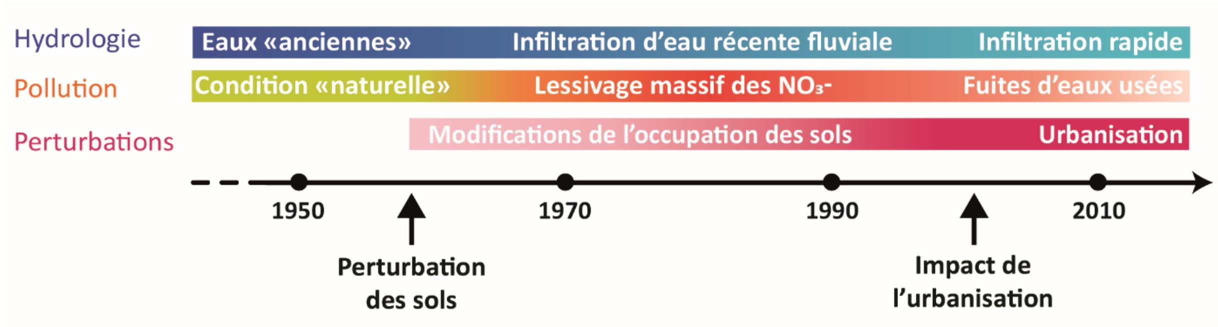

Figure 17: Représentation schématique de l'évolution des processus de contamination en lien avec les pressions exercées sur l'hydrosystème de la lagune de Biguglia.

En effet, si le stockage des NO_3^- représente un risque pour la consommation humaine, leur migration inéluctable vers la lagune menace également cet écosystème fragile. Bien que les concentrations en NO_3^- mesurées dans les eaux du Bevincu soient très faibles (<2 mg/L), la lagune présente tout de même des hausses de NO_3^- importantes et occasionnelles, dont l'amplitude est particulièrement forte dans le bassin Sud, au niveau des stations de pompage qui sollicitent le plus les eaux souterraines. **L'excellente corrélation des dynamiques entre les niveaux piézométriques et les teneurs en NO_3^- dans la lagune laisse présumer que les eaux souterraines ont un impact prédominant sur l'approvisionnement en NO_3^- de la lagune.** L'augmentation soudaine et ponctuelle des NO_3^- dans la lagune semble reliée aux épisodes de fortes précipitations, entraînant un lessivage massif des NO_3^- vers la zone saturée avant de rejoindre la lagune, via l'écoulement ou le pompage des eaux souterraines.

Les concentrations modérées en NO_3^- dans les eaux souterraines « récentes » soulignent une amélioration en cours des pratiques de gestion permettant une diminution des flux azotés vers l'aquifère. Cependant, étant donné le décalage de plusieurs décennies observé entre les activités à l'origine de la pollution et la dégradation effective de la ressource, même l'arrêt complet des apports anthropiques en azote ne saurait induire une amélioration immédiate de la qualité des eaux souterraines. La contamination contemporaine de l'aquifère pourrait donc avoir des effets néfastes sur la qualité des eaux souterraines et des écosystèmes associés pendant encore plusieurs décennies.

Chapitre 6 :

Vulnérabilité des aquifères côtiers face aux pressions anthropiques actuelles et historiques

Ce chapitre s'intéresse à la vulnérabilité actuelle et passée des eaux souterraines en mettant en relation des cartographies de vulnérabilité intrinsèque et spécifique avec l'étude des contaminations azotées héritées d'activités anthropiques passées.

6. Vulnérabilité des aquifères côtiers face aux pressions anthropiques actuelles et historiques

6.1. Introduction

La cartographie de la vulnérabilité des eaux souterraines est le plus souvent effectuée dans l'objectif de connaître la situation actuelle de la ressource vis-à-vis d'un, ou plusieurs processus de contamination en cours. La question reste cependant en suspens dans les cas des aquifères soumis à une dégradation héritée de pratiques ou de contaminations passées et/ou pour lesquels l'influence anthropique a évolué de manière significative au cours du temps. Pourtant, à l'image de l'aquifère de la Marana, les activités anthropiques historiques peuvent influencer l'état qualitatif des eaux souterraines sur plusieurs décennies et représenter ainsi de potentielles menaces pour l'exploitation des eaux souterraines et les écosystèmes qui en sont tributaires. Les gestionnaires ont alors besoin de stratégies de gestion à long terme, qui intègrent la vulnérabilité des eaux souterraines liées aux contaminations passées et actuelles pour assurer leur pérennité future.

Dans ce chapitre, les vulnérabilités intrinsèques et spécifiques des eaux souterraines de la plaine de la Marana sont testées et comparées à l'aide de quatre méthodes de cartographie. La vulnérabilité intrinsèque - se basant sur les caractéristiques naturelles du milieu pour déterminer la sensibilité des eaux souterraines à la pollution par les activités humaines - a été cartographiée avec les méthodes DRASTIC et SINTACS. La vulnérabilité spécifique - permettant d'évaluer la vulnérabilité des eaux souterraines vis-à-vis d'un polluant ou groupe de polluants en tenant compte des caractéristiques propres au milieu et aux polluants - a été calculée à l'aide des méthodes SI et DRASTIC-Modifié, intégrant toutes deux l'occupation des sols comme paramètre de calcul. Les résultats sont exprimés sous forme cartographique comprenant 5 degrés de vulnérabilité : très faible, faible, modéré, fort et très fort.

6.2. Résultats et interprétations

Les résultats et interprétations de ces travaux ont fait l'objet de la rédaction d'un article publié dans *« Science of the Total Environment »* en mars 2019, intitulé « Combinations of geoenvironmental data underline coastal aquifer anthropogenic nitrate legacy through groundwater vulnerability mapping methods», présenté ci-après. Les principales avancées concernant la vulnérabilité des eaux souterraines de la Marana sont également résumées en français, en conclusion de ce chapitre.

Combinations of geoenvironmental data underline coastal aquifer anthropogenic nitrate legacy through groundwater vulnerability mapping methods

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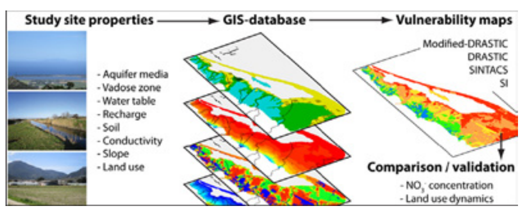
HIGHLIGHTS

- The vulnerability of a Mediterranean coastal aquifer with high nitrate legacy is mapped.
- 4 index-based groundwater vulnerability assessment methods are compared.
- Specific and intrinsic groundwater vulnerability are computed.
- Vulnerability classes are validated by nitrate concentration resulting from legacy-type contamination.

GRAPHICAL ABSTRACT



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ABSTRACT

Groundwater quality is strongly dependent on land use. Past and current anthropogenic activities can lead to the diffusion of contaminants in aquifers. This diffusion can threaten the resource exploitation for decades, thereby endangering the ecological health of groundwater dependent ecosystems. Thus, groundwater stakeholders need methods for long-term management which integrate groundwater vulnerability. This study was conducted on the shallow alluvial aquifer of the groundwater-dependent Biguglia lagoon on Corsica Island, France. The aquifer is exposed to anthropogenic contamination for many decades with nitrate contamination legacy linked to agricultural activities, uncontrolled urbanization and sewage leakages.
In most cases, vulnerability mapping is done in the objective of comparing groundwater situation regarding an on-going contamination process. But the question is still pending for aquifers where contamination is inherited from past practices or contaminations and where anthropogenic influences have changed through time.
To propose an effective and innovative method for territorial management in Mediterranean alluvial aquifers, four index-based groundwater vulnerability mapping methods were tested and compared: two intrinsic vulnerability mapping methods (DRASTIC and SINTACS) and two specific vulnerability mapping methods (Modified-DRASTIC and SI), the latter integrating land use in the accuracy of groundwater vulnerability. Novelty is coming from the comparison between vulnerability maps and their application and validation in a hydrosystem affected by nitrate legacy-type contamination. The specific vulnerability mapping methods are more likely to represent the current pressures to which groundwater are subject. Thus, specific vulnerability methods such as the SI one revealed here very relevant to assess groundwater quality and to react retrospectively. The comparison between groundwater nitrate legacy and intrinsic groundwater vulnerability methods appeared also useful to

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define priority protection areas in long-term territorial management planning (EU Water Framework Direction). In this sense, the SINTACS method seems to be the more appropriate in the Mediterranean and alluvial context of this study.

1. Introduction

Groundwater degradation is generally due to anthropogenic activities and land use evolution over time. During infiltration and circulation in the vadose zone, water can record markers of anthropogenic activities. Because of the low groundwater velocity, depending on circulation media and geochemical conditions, human activities legacy can be persistent after several decades and up to several centuries in aquifers (e.g. Cao et al., 2018; Erostate et al., 2018). This is true for nitrates that are clear evidence of agricultural activities, sewage infiltration and/or nitrate storage in the vadose zone, especially when the natural groundwater baseline is exceeded (Djebebe-Ndjiguim et al., 2013; Yakovlev et al., 2015; van Meter et al., 2016; Ascott et al., 2017; Vystavna et al., 2017a, 2017b). Groundwater nitrate legacy can then act as witnesses of past land uses. In order to insure the long-term groundwater quality, it is important to plan land use with regards to the spatial variability of aquifer vulnerability.

The term of vulnerability was first applied to groundwater by Margat (1968) and was later adopted worldwide (e.g. Vrba and Zaporozec, 1994; Tesoriero, 1998; Sener et al., 2009; Marin et al., 2015). It is based on the assumption that the physical environment may provide some degree of protection to groundwater against the natural and human influences, especially with regard to contaminants entering the subsurface environment (Vrba and Zaporozec, 1994). Among the numerous other definitions proposed in the literature (e.g. Albinet and Margat, 1970; Villumsen et al., 1982; Vrba and Zaporozec, 1994; Gogu and Dassargues, 2000), authors distinguish between intrinsic and specific vulnerabilities. The intrinsic vulnerability can be defined as a quantitative, relative, non-measurable and dimensionless property, considering the hydrogeological characteristics of an aquifer, that is independent of the type of contamination scenario (Vrba and Zaporozec, 1994; Kavouri et al., 2011; Huneau et al., 2013). Specific vulnerability is the assessment of vulnerability associated with contaminant sources that take into consideration the contaminant's characteristics and its connection to the various components of intrinsic vulnerability (Gogu and Dassargues, 2000; Brindha and Elango, 2015; Vaezihir and Tabarmayeh, 2015). Thus, intrinsic vulnerability is defined solely as a function of hydrogeological factors and specific vulnerability is defined by the potential impacts of specific land uses and contaminants (Stigter et al., 2006).

Different approaches are proposed for groundwater vulnerability mapping in intergranular porosity contexts: statistical ones that express vulnerability in terms of probability of contamination (Teso et al., 1996; Troiano et al., 1997; Burkart et al., 1999); processed-based simulations based on numerical modelling (Rao et al., 1985; Jury and Ghodrati, 1989; Wu and Babcock, 1999; Pineros Garcet et al., 2006; Tiktak et al., 2006) and overlay and index-based techniques that are based on field parameters notation (Aller et al., 1987; Foster, 1987;Civita, 1994; Vrba and Zaporozec, 1994; Plagnes et al., 2010). Overlay and index-based methods are comparatively simpler and provide a good balance between the difficulties of data acquisition, computational complexity and reliability of the results. Despite the use of unavoidable simplifying assumptions, index-based techniques are the most employed for groundwater vulnerability assessment (Kumar et al., 2015).

Many index-based methods have been developed since the 80's. Some examples of overlay and index methods are DRASTIC (Aller et al., 1987), GOD (Foster, 1987), SINTACS (Civita, 1994), modified-DRASTIC (Secunda et al., 1998), or SI (Susceptibility Index, Ribeiro, 2000). To limit uncertainties due to each method, the combination of selected overlay and index-based methods may represent an advantage for gaining confidence in vulnerability mapping by comparing the results and analysing their consistency in practical case studies where contamination has already occurred (Stigter et al., 2006). Thus, numerous researchers have compared the results of different vulnerability mapping methods on specific sites (for example Murat et al., 2004; Anane et al., 2013; Brindha and Elango, 2015). They all inferred that vulnerability results vary significantly with the type of method used and that each method is clearly more appropriate to specific hydrological contexts.

In most cases, vulnerability mapping is done in the objective of comparing the situation of groundwater regarding an on-going contamination process. But the question is still pending for aquifers where contamination is inherited from past practices or contaminations and where anthropogenic influences have changed through time. In this study, we propose an innovative approach where intrinsic and specific groundwater vulnerabilities are computed and compared with the past and current land use effects on the hydrogeological vulnerability regarding the nitrate legacy. A Mediterranean alluvial coastal aquifer in Corsica (France) was selected for its intergranular porosity and for its groundwater nitrate contamination attributed to a progressive accumulation of pollutants over time. The current low groundwater quality is principally due to legacy pollution from land-use practices several decades-old, agricultural fertilization, and uncontrolled urbanization with severe sewage leakage (Pasqualini et al., 2017; Erostate et al., 2018). Thus, both the socio-economical context and the hydrogeological situation of this aquifer appeared well adequate to test different vulnerability assessment methods. The intrinsic vulnerability was mapped with DRASTIC and SINTACS methods. SI and modified-DRASTIC were used to compute specific vulnerability. The good level of knowledge on land use evolution history, aquifer functioning and groundwater quality of this aquifer make it an ideal study site to test the relevance of such an approach in the context of rapid land use change.

2. Methods

2.1. Study site

2.1.1. Geological and hydrological settings

Corsica Island is characterized by a typical Mediterranean climate associated with marked inter-annual variations of precipitation and temperature. The mean annual temperature and precipitation measured at the Bastia Météo France Station are 15.8 °C and 768 mm respectively (average calculated from 1950 to 2016).

The Biguglia lagoon is a shallow and brackish coastal lagoon located in the northern part of Corsica Island (France), just south of Bastia city (Fig. 1; Garrido et al., 2016). The lagoon is part of the largest wetland of Corsica (14.5 km^2) formed by the marine reworking of the Golu River alluvial deposits. It is isolated from the Tyrrhenian Sea by a sandy bar and seawater exchanges are controlled by a narrow and natural channel to the North. The alluvial part of the Biguglia catchment is composed by the large Marana sedimentary plain. It is constituted by a set of nested paleo-terraces from east to west, more or less eroded by the two main rivers (Golu and Bevincu rivers). This alluvial plain is comprised of Quaternary deposits made of lustrous schist erosion products transported by the Golu River, the Bevincu River and temporary minor streams from the western high relief of the lagoon watershed. The terraces were formed during glacial phases and were altered during interglacial periods (Conchon, 1972, 1978). They are composed of gravels,

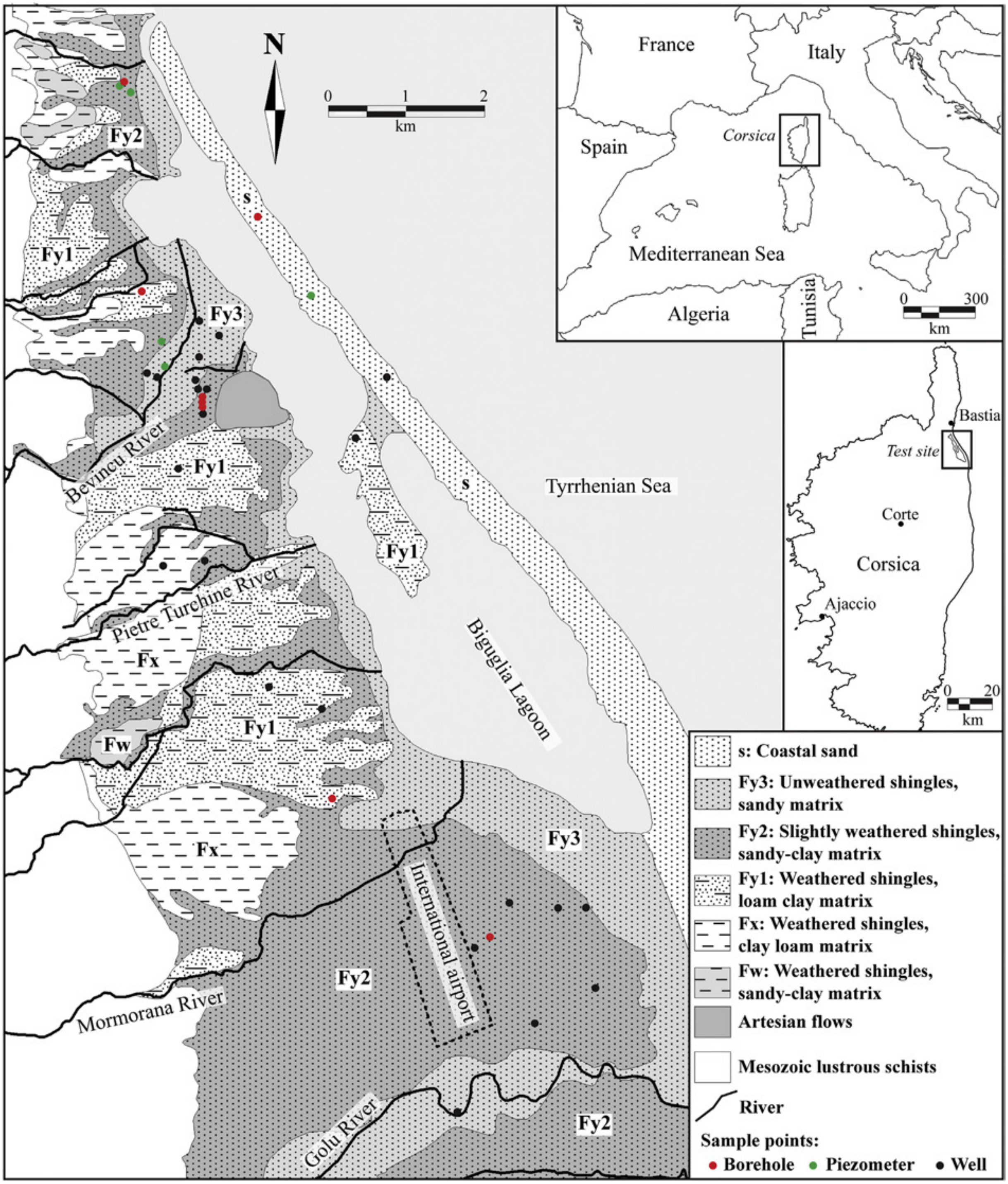

Fig. 1. Localisation and geology of the Biguglia watershed (adapted from Conchon et al., 1994 and Lahondère, 1983) and distribution of the sampling points.

pebbles or blocks, and are surrounded by a sandy-loamy matrix more or less developed. The aquifer is located mostly in the Middle and Upper Wurmian alluvial deposits (Fy2 and Fy3, Fig. 1) and overlays an impermeable substratum formed by the Lower Wurmian clay deposits (Fy1).

This groundwater resource corresponds to the main aquifer of the region. The total surface area of this shallow aquifer is estimated at 80 km^2 with a thickness of 3 to 40 m and a hydraulic conductivity (K) of 10^{-7} $m \cdot s^{-1}$ to 10^{-3} $m \cdot s^{-1}$. Groundwater is used mostly for small local irrigation purposes but also for drinking water supply through two main pumping areas located at the Bevincu mouth for the water supply of Bastia city and in a meander of the Golu River south of the study area that supplies all the southern plain in drinking water (needs about 5 M m^3 annually). The generally short response time (from 4 to 15 years) with pressure transfers reflecting its good connection with the surface during recharge episodes (Erostate et al., 2018). Seasonal variations of the groundwater level reach a maximum of 2 m. However, in the central part, the aquifer displays longer residence times, above 25 years (Erostate et al., 2018). Besides, the nitrate concentrations clearly increase with the residence time, showing the progressive accumulation of pollutants in the aquifer. Thus, the low groundwater quality is in large parts due to the contamination by legacy pollutants dating back to the 70/80's (Erostate et al., 2018). All the groundwater flow line directions converge to the lagoon and the gradient calculated from the water level data ranges between 3 and 5‰, showing hydrodynamic behaviour favourable to groundwater flow (Orofino et al., 2010; Garel et al., 2016). The aquifer represents a significant contribution to freshwater input for the lagoon, making it a coastal Groundwater Dependant Ecosystem (Kløve et al., 2011; Balestrini et al., 2016; Krogulec, 2016).

2.1.2. Socio-economical context

The catchment area of the Biguglia lagoon, and especially the plain of the Marana, is subject to an important urban development as shown by a clear increase in the population living around the pond since the 1950's (Fig. 2); this is due to its flat topography and privileged geographical location just south of the second largest city of Corsica. Moreover, the area is crossed by an important road network associated with a dynamic economic context, including an airport, several business parks, and a major tourism zone on the lido (highly populated in summer months).

Agricultural activities (fruits and vegetable production, cattle breeding) and increasing urbanization are the two main kinds of anthropogenic pressure in the area. Since 1950, the intensification of urbanization is to the detriment of agricultural areas (Table 1). Indeed, because of a delay in the development and application of local urban zoning laws, the study area has experienced a significant urban sprawl (Prévost and Robert, 2016). For instance, the four towns around the lagoon (Furiani, Biguglia, Borgo, Lucciana) increased their artificial areas by about 3,04% per year between 2002 and 2011 (Robert et al., 2015; Table 1). Nevertheless, forest and Mediterranean shrub lands have remained stable in the area since the mid-20th century after having strongly been destroyed in the 19th century.

Considering the various potential pressures on the watershed and the important development of urbanization along the Corsican shore, and because of its remarkable biodiversity, the Biguglia lagoon was recognized as a RAMSAR site (Convention on Wetlands of International Importance especially as Waterfowl Habitat, 1971) since 1991 and classified as a nature reserve since 1994 (Département de la Haute-Corse, 2012).

2.2. Vulnerability assessment using index methods

2.2.1. Mapping methods

Overlay and index methods rely mainly on the quantitative or semi-quantitative compilation and interpretation of mapped parameters (Gogu and Dassargues, 2000). These parameters, their number and

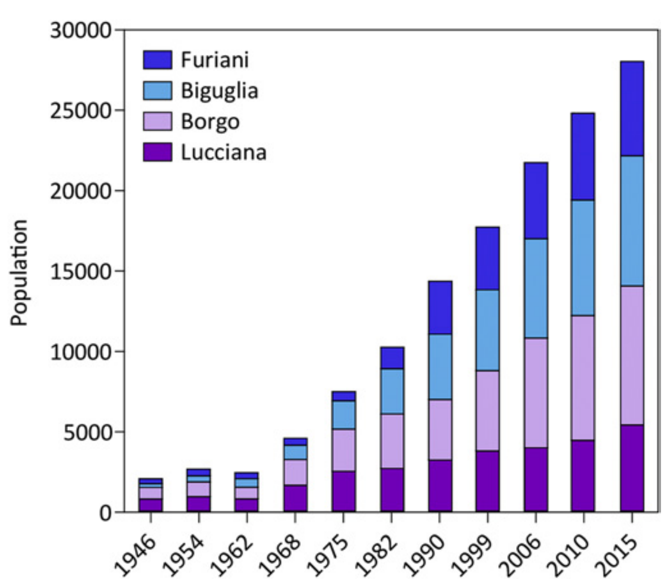

Fig. 2. Demographic evolution of the 4 main municipalities standing in the watershed from 1946 to 2015 (INSEE).

Table 1
Land use evolution of the Biguglia lagoon catchment from 1913 to 2012 (CORINE land-cover program and Archives départementales de Haute-Corse; Corvol, 1999).

Land use	1913	1990	2006	2012	1913–2012 surface evolution (%)
Urbanization (km^2)	4.1	15.8	18.5	21.0	+519.5
Agriculture (km^2)	71.7	43.9	42.7	43.9	−61.2
Forest and shrublands (km^2)	31.6	46.5	45.0	46.5	+147.0

their definition, vary from one method to another, but the estimation of groundwater vulnerability is linked to three major factors: soil conditions, the unsaturated zone of subsoil and bedrock, and transport in the saturated zone. All the parameters are described in details later. Their combination leads to a final vulnerability index value that quantifies sensitivity to contamination (Gogu and Dassargues, 2000; Kumar et al., 2015). Land use is a supplementary factor implemented in specific vulnerability methods that distinguish them from intrinsic ones.

The DRASTIC model was developed by the U.S. Environmental Protection Agency (EPA) to evaluate the intrinsic vulnerability of groundwater pollution for the entire United States (Aller et al., 1987). The acronym DRASTIC stands for the seven parameters used in the model which are: Depth to water, net Recharge, Aquifer media, Soil media, Topography, Impact of vadose zone and K (Table 2). Each parameter is rated from 1 (low sensitivity to pollution) to 10 (high sensitivity) based on their relative impact on aquifer vulnerability. The seven parameters are then assigned constant weights ranging from 1 to 5 reflecting their relative importance (Babiker et al., 2005).

SINTACS is the most widely used method for intrinsic groundwater vulnerability assessment in Italy, and it is the standard method in Italian legislation on the protection of groundwater against pollution (Italian Law by Decree No. 152, 11/5/1999, 1999; Guastaldi et al., 2014; Busico et al., 2017). It represents an adaptation of the DRASTIC method to the hydrogeological, climatic, and impact settings that are typical of the Italian territory and of the Mediterranean basin (Hamza et al., 2007). The SINTACS method takes into account the same parameters as the DRASTIC method but parameter weighting is different (Table 2); higher weights are attributed to the soil properties and to the topography in the SINTACS model.

SI is also adapted from DRASTIC. It was developed with the intention of evaluating aquifer vulnerability on large to medium scales (Ribeiro, 2000), with respect to diffuse agricultural pollution in hydrogeological settings typically found in Portugal. The main difference is the addition of a parameter defining land cover (abandoning the concept of a purely intrinsic vulnerability assessment method) and the abandonment of three DRASTIC parameters (soil media, unsaturated zone and K; Table 2). In Ribeiro (2000), parameter rates are from 0 (less vulnerable) to 100 (the most vulnerable) and parameter weights are <1. So as to facilitate the comparison of final vulnerability results from each method in this study, SI parameter rates were divided by 10 and parameter weights were multiplied by 10. This multiplication procedure was also adapted for cross-comparison of vulnerability assessment methods in earlier studies (e.g. Stigter et al., 2006; Brindha and Elango, 2015).

Numerous modifications of the DRASTIC method have been proposed since its first application (e.g. Khosravi et al., 2018). One of them is modified-DRASTIC from Secunda et al. (1998) which added a land use parameter within the original method as a supplementary parameter to the DRASTIC model. The same notations and the same weights are applied to the basic parameters (Table 2), but the addition of land cover allows the mapping of specific groundwater vulnerability.

Table 2
Parameters and associated weights used in the DRASTIC, SINTACS, SI and modified-DRASTIC methods (x signify that this parameter is not used in the method; SI parameter weights have been multiplied by 10 in this study).

	Parameter weights							
Mapping method	Aquifer media	Vadose zone	Depth to water	Net recharge	Soil	Hydraulic conductivity	Slope	Land use
DRASTIC (Aller et al., 1987)	3	5	5	4	2	3	1	x
SINTACS (Civita, 1994)	3	5	5	4	3	3	3	x
SI (Ribeiro, 2000)	0.259	x	0.186	0.212	x	x	0.121	0.222
Modified- DRASTIC (Secunda et al., 1998)	3	5	5	4	2	3	1	5

Final vulnerability index (VI) is then computed in the whole studied area thanks to the Eq. (1).

$$VI = \sum_{i=1}^{n} P_{ri} \times P_{wi} \tag{1}$$

where
"Pr_i" is the rating value of the parameter i,
"Pw_i" is the theoretical weight of the parameter i,
"n" is the number of parameters to be implemented.

VI will give a relative measure of the vulnerability of groundwater to pollution. Regions classified with higher index are more prone to pollution compared to low index areas.

2.2.2. Parameter description, data acquisition and compilation

The needed parameters were compiled from different sources in a GIS database. For all parameters, the dataset was mapped in vector format with MapInfo Professional 7.8 (Mapinfo Corporation, 2003). Because the data came from different sources, data layers were resampled to be suitable to the chosen resolution of 20 m. This resolution is a reasonable compromise between the different resolutions of the datasets, the regional extent of the test area, and the accuracy necessary to compare the results of the four methods.

The vector files were then discretized into grid files of 20 m × 20 m cells with a vector-to-raster conversion using Vertical Mapper 3.1. The process simply converts the existing region vector file to a grid file where every grid node is given the same value as the region it falls in. These grid files were finally added together according to Eq. (1) that was automatically applied in each cell of the grid to reach the final vulnerability maps.

2.2.2.1. Aquifer media parameter. The "Aquifer media" parameter refers to the consolidated or unconsolidated rock constituting the saturated zone. Its properties can allow a contaminant attenuation that depends on the amount and sorting of fine grains. It also defines the route and path length that is followed by a contaminant, which is one of the important controls in determining the time available for attenuation processes such as sorption, reactivity and dispersion to occur (Aller et al., 1987). The classification and rating of this parameter in this study are based on the geological map (Lahondère, 1983; Conchon et al., 1994; Orofino et al., 2010) and on the guidelines from Aller et al. (1987) for DRASTIC, modified-DRASTIC and SI, and from Civita (1994) and Civita and De Maio (2004) for SINTACS. Classifications and corresponding ratings are given in Supplementary Table 1.

2.2.2.2. Vadose zone parameter. The "vadose zone media" parameter refers to the material below the typical soil horizon and above the water table, which is unsaturated or discontinuously saturated. Biodegradation, neutralization, mechanical filtration, chemical reaction, volatilization and dispersion are processes which may occur within the vadose zone (Aller et al., 1987). Furthermore, as in the saturated zone, the vadose zone media controls the path length and routing, thus affecting the time available for attenuation. The classification and rating of this parameter in this study are based on the geological map (Lahondère, 1983; Conchon et al., 1994) and on the guidelines from Aller et al. (1987) for DRASTIC and modified-DRASTIC, and from Civita (1994) and Civita and De Maio (2004) for SINTACS. This parameter is not considered for the SI method. Classification and corresponding ratings are given in Supplementary Table 2.

2.2.2.3. Depth to water table parameter. The "depth to water table" parameter corresponds to the thickness of the unsaturated zone. It represents the distance that a potential contaminant has to travel before reaching the aquifer water table (Ouedraogo et al., 2016). The vulnerability to pollution decreases with the thickness of the unsaturated zone because there is generally a greater chance for attenuation to occur as this thickness increases (Aller et al., 1987). This parameter was computed by subtracting the ground surface elevation (from BD ALTI® V 2.0, IGN, 25 m resolution) by the piezometric level interpolated from field campaign conducted in April 2015 in 38 groundwater points. High water period was chosen in order to obtain vulnerability assessment results in the most critical situation. Ranges and corresponding ratings were obtained from Aller et al. (1987) for DRASTIC, modified-DRASTIC and SI, and form Civita (1994) for SINTACS (Supplementary Table 3).

2.2.2.4. Net recharge parameter. The "Net recharge" is the amount of water that reaches the water table. Its role is very significant in aquifer vulnerability assessment because of its capacity to transmit pollutants from the soil surface to the water table. The susceptibility of groundwater to pollution generally increases with the amount of net recharge, but the positive effect of dilution beyond a certain value is also integrated into the SINTACS method (Civita and De Maio, 2004). Net recharge was computed by the combination of annual effective precipitation produced by CGDD et al. (2016) and the Index of Development and Persistence of the River networks (IDPR) database (InfoTerre). The IDPR, developed by Mardhel et al. (2004) and modified by Gay et al. (2016), characterizes the landscape in terms of soil infiltration/runoff.

It assumes that the organization of the drainage network is dependent on the underlying geological formations and describes the theoretical river network established due to morphological parameters only and the real river network that has developed under heterogeneous geological conditions (Gay et al., 2016). This index provides a quantitative estimation of the capacity of subsurface formations to infiltration or runoff. Therefore, a partition coefficient between runoff and infiltration can be estimated from this index. Impermeable surfaces (road, airport runway, densely urbanized area, commercial zones and parking) are considered as null recharge areas and are rated 1 for this parameter for the four applied methods. The net recharge values obtained from these computations were then rated in accordance with the guidelines provided by the authors for each method (Supplementary Table 4).

2.2.2.5. Soil media parameter. The "Soil media" parameter is used only in DRASTIC, SINTACS and Modified-DRASTIC, with the same classification and rating for the three methods (Supplementary Table 5). It refers to the uppermost portion of the vadose zone characterized by significant biological activity. The soil has a significant impact on the amount of recharge which can infiltrate and hence on the ability of a contaminant to move vertically into the vadose zone. The presence of fine-textured materials such as silts and clays can decrease relative to soil permeability and restrict contaminant migration (Aller et al., 1987). For this study, a 1:25,000 soil map of Corsica from Demartini and Favreau (2011) was used.

2.2.2.6. Hydraulic conductivity parameter. The "hydraulic conductivity" parameter (K) is not used in the SI method. Ranges and ratings are different for DRASTIC, modified-DRASTIC and SINTACS. Applicable values for this study are available in Supplementary Table 6. This parameter describes the ability of the aquifer to transmit water and the potentially associated contaminants when submitted to a hydraulic gradient. High conductivity values will be associated with high contamination risks (Rahman, 2008). In this study, this parameter has been mapped by assuming that K was homogeneous within each different geological formation of the aquifer. For each formation, existing data were been used (Orofino et al., 2010; Erostate et al., 2018; French geological database: BSS; InfoTerre).

2.2.2.7. Topography parameter. The "topography" parameter determines the infiltration capacity of surface water into the soil and hence the capacity to introduce pollution into the soil (Ouedraogo et al., 2016). A low slope permits greater infiltration and a greater potential for contaminant migration while a high slope fosters lower infiltration and greater runoff (Shrestha et al., 2016). The slope of the study area was computed from the BD ALTI® V 2.0, IGN (25 m resolution) database and rated according to authors' recommendations (Supplementary Table 7).

2.2.2.8. Land use parameter. The "land use parameter" is used only in specific vulnerability assessment methods, i.e. modified-DRASTIC and SI in this study. It defines the activity on the ground surface that can act as potentially pollution sources. In this study, a detailed land use map based on aerial photographs and compliant with CORINE Land Cover classification (CGDD, 2009) was used (Robert et al., 2015) and completed with the French governmental database RPG (Graphical Parcel Register; ASP, 2012) for agricultural areas. The rating of land uses was based on the same classification as Ribeiro (2000) for both SI and modified-DRASTIC methods (Supplementary Table 8).

2.3. Comparison and validation of the results

2.3.1. Reclassification of vulnerability indexes

In every 400 m^2 (20×20 m) cell of the mapped area, the application of Eq. (1) results in a different vulnerability value for each method. The final index of all these methods gives a relative measure of the vulnerability of groundwater to pollution. Regions classified with higher indices are more prone to pollution compared to low index areas. In order to compare these results, a reclassification is necessary. Different ranges of vulnerability are proposed in the literature. Table 3 summarizes the different ranges of vulnerability classification used in the literature. No universal method of classification is available. For comparison of vulnerability index computed with different methods, the quantile classification is appropriate (Rahman, 2008; Güler et al., 2013; Brindha and Elango, 2015). Thus, this uniform method was adopted in this study, i.e. quantile classification that range data into five categories (very low, low, moderate, high and very high vulnerability) with an equal number of units in each category.

2.3.2. Validation of the results

2.3.2.1. Field measurements and laboratory investigations. To validate the vulnerability maps obtained, geochemical investigations were carried out in the field in order to sample groundwater and quantify the local nitrate fingerprint on the aquifer. Groundwater sampling sites ($n = 38$) including wells, boreholes and piezometers were selected over the study area in order to cover as much of the territory as possible with a homogeneous density (Fig. 1 and Supplementary Table 9).

Groundwater table was measured at each site with an OTT KL010 contact gauge. Groundwater Electrical Conductivity (EC) and Temperature (T) were measured in the field with a WTW Cond 3310 m (WTW GmbH, Weilheim, Germany). Bicarbonates were determined in the field using a HACH digital titrator (HACH Company, Loveland, USA). Samples for nitrates as well as for other major ions (Cl^-, SO_4^{2-}, Na^+, Ca^{2+}, Mg^{2+} and K^+) were filtrated through 0.45 μm nitrocellulose membranes before collection in two 50 mL polyethylene bottles and stored at 4 °C before analysis. One of them was acidified using ultrapure HNO_3 for cation analysis. The ionic concentrations were determined with a Dionex ICS 1100 chromatograph (Thermo Fischer Scientific, Waltham, USA) at the Hydrogeology Department of the University of Corsica, France. The chemical analysis quality was checked by computation of the ionic balance error and rejected if >5%.

These chemical results were used to validate the vulnerability assessment maps and to observe if, in the context of this study, one method is more reliable than the others. Since there is no geologic source of nitrate in the study area and because only human activities can lead to a noticeable NO_3^- concentration in groundwater of this aquifer, this ion was chosen. In addition, Erostate et al., 2018, highlighted the nitrate legacy in the groundwater of the Biguglia alluvial aquifer with an increasing of NO_3^- concentrations with the groundwater ageing and in relation with the ancient agricultural practices.

3. Results and discussion

3.1. Parameter maps

3.1.1. Aquifer media, vadose zone and hydraulic conductivity

Mapping of these three parameters was based on the geological features of the study area (Fig. 1 and Supplementary Tables 1, 2 and 6). For aquifer media, the same notation was applied for DRASTIC, Modified-DRASTIC and SI. According to the author's

Table 3
Examples of ranges of vulnerability used in the literature for vulnerability indexes classification.

Method	References	Vulnerability classes based on vulnerability indexes From very low vulnerability (left) to very high vulnerability (right)						
DRASTIC and DRASTIC based methods	Engel et al. (1996); Anane et al. (2013)	1–120	121–160	161–200	>200			
	Ouedraogo et al. (2016)	<84	84–114	115–145	146–175	>176		
	Krogulec (2016); Shrestha et al. (2016)	<100	100–125	126–150	151–175	176–200		
	Stigter et al. (2006)	<79	80–99	100–119	120–139	140–159	160–179	180–199
	Babiker et al. (2005)	Five classes based on the fixed interval of area percentage						
SI	Ribeiro (2000)	<45	45–64	65–84	>85			
SINTACS	Civita (1994); Al Kuisi et al. (2006); Pisciotta et al. (2015)	<25	l 25–39	40–49	50–75	>75		
All methods	Rahman (2008); Güler et al. (2013); Brindha and Elango (2015), This study	Five classes based on quantile classification						

guidelines, the computation for these three vulnerability assessment methods led to an aquifer media rating of about 4 for 15% of the mapped area (Fw and Fx formations and the artesian zone; Fig. 1) and about 5 for another 15% (Fy1; Fig. 1). 39% of the area is rated 8 (Fy2 formation; Fig. 1) for this parameter and 31% is rated 9 (Fy3 and the sand bar; Fig. 1). For SINTACS, 15% of the surface area is rated 6 (Fw, Fx and the artesian area), 15% is rated 7 (Fy1), 39% is rated 8 (Fy2) and 31% is rated 9 (Fy3 and the sand bar).

The vadose zone parameter is not used in SI. The vadose zone of the sand bar offers little protection to groundwater pollution and is rated 9 for the three other methods. The rating for Fw, Fx, Fy1 and the artesian zone is more favourable to protection (rate about 4 to 5 for DRASTIC and Modified-DRASTIC and 3 to 4 for SINTACS). The rest of the study area (Fy2 and Fy3 formations) is rated 7 to 8 for DRASTIC and Modified-DRASTIC and 5 to 6 for SINTACS.

The K values are highly variable with values ranging from $1 \times 10^{-4}\ m \cdot s^{-1}$ to $1 \times 10^{-7}\ m \cdot s^{-1}$. These K values are based on the local literature and the sparsely available results of pumping tests conducted in the different formations (Orofino et al., 2010; Erostate et al., 2018; French geological database: BSS; InfoTerre). This led to the rating of this parameter from 1 (DRASTIC and Modified-DRASTIC) and 2 (SINTACS) in the artesian zone (K about $1 \times 10^{-7}\ m \cdot s^{-1}$) to 9 (DRASTIC and Modified-DRASTIC) and 9.75 (SINTACS) for Fy2 and Fy3 formations (K about $5 \times 10^{-3}\ m \cdot s^{-1}$). The rest of the aquifer (K about 1 $10^{-4}\ m \cdot s^{-1}$) for Fw, Fx and Fy1 is rated 3 (DRASTIC and Modified-DRASTIC) and 7.5 (SINTACS).

3.1.2. Depth to groundwater

The depth to groundwater was mapped with the combination of the DTM (Fig. 3A) and the interpolated piezometric levels in high water level. Results of this computation are given in Fig. 3B. The thickness of the non-saturated media decreases from west to east (i.e. from the inland relief to the coast), conversely to the depth to groundwater rating. The maximum thickness of the non-saturated media is about 80 m in the western extremity of the alluvial formations. Nevertheless, only 2% of the surface area is concerned by a depth to groundwater >40 m (rating about 1 for the four tested methods; Supplementary Table 3). 18% of the surface area is rated under the moderate value for all the methods (2

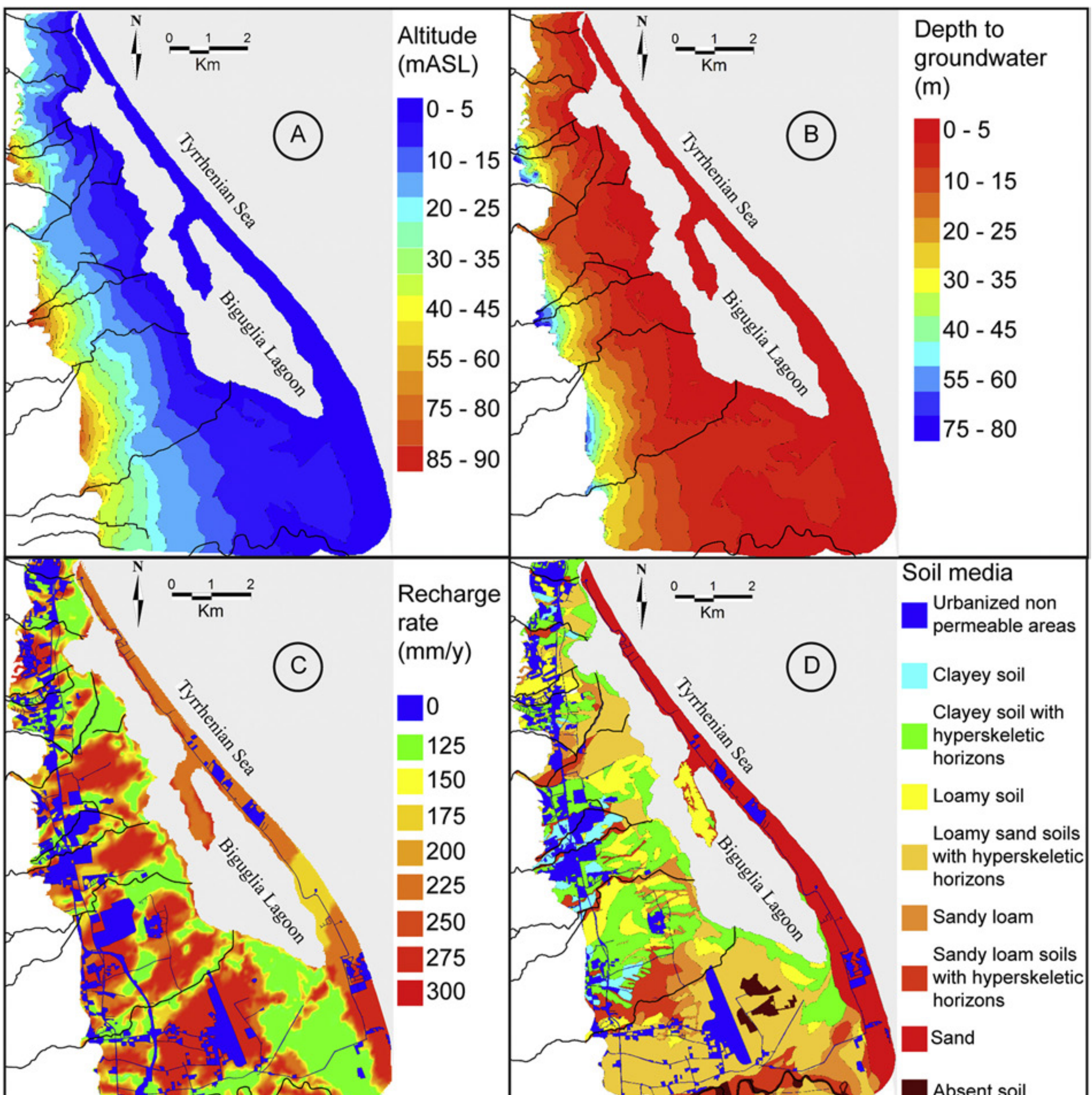

Fig. 3. Maps of some parameters used for groundwater vulnerability mapping (A: topography, B: Depth to groundwater table in April 2014, C: Net recharge, D: Soil media).

to 3 for DRASTIC, Modified-DRASTIC and SI, and 2 to 4 for SINTACS), which correspond to a non-saturated media from 15 m to 40 m thick. Finally, because of the low depth to groundwater of the Biguglia aquifer in the alluvial plain, 79% of the area has a rating >5, especially in the eastern half of the study area where the slope is gentler. Depth to groundwater value is from 5 m to 15 m for 37% of the surface area (rating about 5 to 7 for DRASTIC, Modified-DRASTIC and SI and 5 to 7 for SINTACS) and from 0 m to 5 m for 43% of the area (rating about 9 to 10 for DRASTIC, Modified-DRASTIC and SI and 8 to 9 for SINTACS).

3.1.3. Net recharge

The net recharge calculated by application of the IDPR varies spatially between 100 $mm \cdot y^{-1}$ and 276 $mm \cdot y^{-1}$, corresponding to 13% to 36% of the average annual rainfall (768 mm, calculated from 1950 to 2016 at the Bastia Météo France Station), respectively. This is in good agreement with the recharge rate of about 23–26% of annual rainfall computed for the neighbouring porous coastal aquifer of Bonifacio (Southern Corsica, France) by Chesnaux et al., 2018 and Santoni et al., 2018. The spatial variation of net recharge is presented in Fig. 3C. The impermeable surfaces (road, airport runway, parking…) rated 1 amount to 13% of the studied area. The net recharge of the remaining study area is >100 $mm \cdot y^{-1}$. For DRASTIC, Modified-DRASTIC and SI, the rated values for this parameter are 6 for 37% of the area, 8 for 24% and 9 for 26%. For SINTACS, 25% of the area is rated 6 for the net recharge parameter; 5% are rated 7; 8% are rated 8; 42% are rated 9 and 7% are rated 10. The ranges of rating are given in Supplementary Table 4.

3.1.4. Soil media

In the coastal zone of the study area, the soil is sandy (rating about 9 for all methods). To the west, the alluvial deposits of the Biguglia watershed have led to the development of soil, which is more or less clayey with frequent hyperskeletic horizons. Sandy, loamy and clayey soils can be observed in various proportions (Fig. 3D). The classification of the soil parameter for vulnerability rating was based on the proportions of sand, clay or loam and on the presence or not of stone- or gravel-rich horizons (Supplementary Table 5) with the same ranges and ratings for the four methods, according to the author's guidelines.

The soil is absent in some areas (about 10% of the area), especially around the Golu river bed and where quarries are exploited. Artificial impermeable zones were rated 1 (13% of the surface) because infiltration can be considered as null in these areas. The rating on the rest of the study area is distributed in the following way: 3% rated 3; 17% rated 4; 10% rated 5; 27% rated 6; 7% rated 7; 9% rated 8 and 12% rated 9. Thus, 71% of the study area has a rating less than or equal to 6 for the soil parameter. Therefore, this parameter should not lead to a major increase in final vulnerability index values.

3.1.5. Slope

The topography of the Biguglia watershed is characterized by three different compartments as can be observed in Fig. 3A. A small part has a high slope, superior to 10% with a maximum of about 32%. It is the area just under the schist formations located at the west of the study area. These surfaces, rated 1 to 3 for DRASTIC, Modified-DRASTIC and SI and rated 4.5 to 1 in the SINTACS method (Supplementary Table 7), represent only 0.5% of the study area. The second compartment is between this high slope area and the alluvial plain. The slope here is between 5% and 10% representing only 2.5% of the study area. These surfaces are rated 5 to 9 in the DRASTIC, Modified-DRASTIC and SI methods and the ratings are from 5.5 to 7.5 for the SINTACS method. Finally, the major part of the study watershed is a large plain with a very low slope. Indeed 97% of the area has a slope under 5% and 45% is inferior or equal to 1%. The ratings for these areas are 10 (DRASTIC, Modified-DRASTIC, SI) and 9.5 (SINTACS) when the slope is <2%. They are about 9 (DRASTIC, Modified-DRASTIC, SI) and from 7.5 to 8.5 (SINTACS) when the slope is between 2% and 5%. The ratings for this parameter will clearly have a major impact on the final vulnerability indexes.

3.1.6. Land use

Land use is an important factor in the Biguglia watershed because of its rapid evolution over the last decades and its high diversity, from protected natural areas to intensely urbanized zones as well as agricultural and industrial activities (Fig. 4). Thus, specific vulnerability assessment by the SI and Modified-DRASTIC could show interesting results besides the intrinsic vulnerability assessment.

The computation based on the classification proposed by Ribeiro (2000; Supplementary Table 8) led to a rate of 0 for this parameter for 41% of the studied area, which corresponds to the sand bar and to the natural protected area around the lagoon. Conversely, 42% of the watershed is rated from 7 to 10 (agricultural, commercial and industrial activities, urban spaces and transport pathways). 17% of the watershed is rated 5, corresponding to the grassland and pasture areas.

3.2. Groundwater vulnerability maps and results comparison

3.2.1. Spatial and visual comparison

The computed vulnerability indexes obtained are very different, but the quantile classification allowed us to compare the results by ranging them into five classes of vulnerability (Table 4). Resulting maps are presented in Fig. 5 with the percentage of area in each class of vulnerability.

For every model used, the very low vulnerability class is the least represented (from 1% of the surface for SI to 6% for DRASTIC). For intrinsic vulnerability assessment method, the low vulnerability class is the second one in terms of area (13% for DRASTIC and 10% for SINTACS) whereas it is the highly vulnerable class for the specific vulnerability methods (8% for SI and 7% for Modified-DRASTIC). The two classes the most represented for DRASTIC are the very high and high classes (31% and 32% of the surface respectively). For SINTACS, it is the moderate and high classes (respectively 22% and 48%). The high and moderate vulnerability classes are the more represented for SI (36% and 42%) and it is the moderate and high classes for Modified-DRASTIC (36% and 37%). These results emphasize the differences in the computation of the four methods.

High differences appear between results from intrinsic and specific vulnerability assessment methods. Indeed, the high and very high vulnerability classes are the most represented for DRASTIC and SINTACS (addition of the two classes: 64% and 63% respectively) while they are <50% for SI and Modified-DRASTIC (44% for the both). The same distribution is highlighted for the moderate class (around 20% of the surface for intrinsic vulnerability method and 36% to 42% for the specific ones). The surface areas represented by the very low and low classes are not very different from one method to another (14% for SI, 21% for Modified-DRASTIC, 13% for SINTACS and 18% for DRASTIC). Thus, it seems that in the context of this work, the specific vulnerability assessment methods are less severe than intrinsic vulnerability methods, with a translation of the high and very high vulnerability classes towards the moderate one.

There are also large differences in results between intrinsic vulnerability assessment methods themselves. DRASTIC results are more worrying than those of SINTACS with a proportion of highly vulnerable areas almost double those of SINTACS. For SI and Modified-DRASTIC, results are similar.

3.2.2. Comparison by discrepancy mapping

In order to identify the source of the differences observed in vulnerability maps and to localize the discrepancies, a numerical classification was used with a score of 1 for the very low vulnerability class, of 2 for the low vulnerability class and so on until a score of 5 for the very high vulnerability class. This method was first proposed by Brindha and Elango (2015). The resulting maps (Fig. 5) were then subtracted one by one to obtain Fig. 6. A difference about $|2|$, which is the maximum discrepancy observed, signifies that the computed vulnerability is different by 2 classes (e.g. moderate and very low), a difference about $|1|$ indicates one class of difference between the two methods (e.g. very high and high) and the results are the same if the difference is 0.

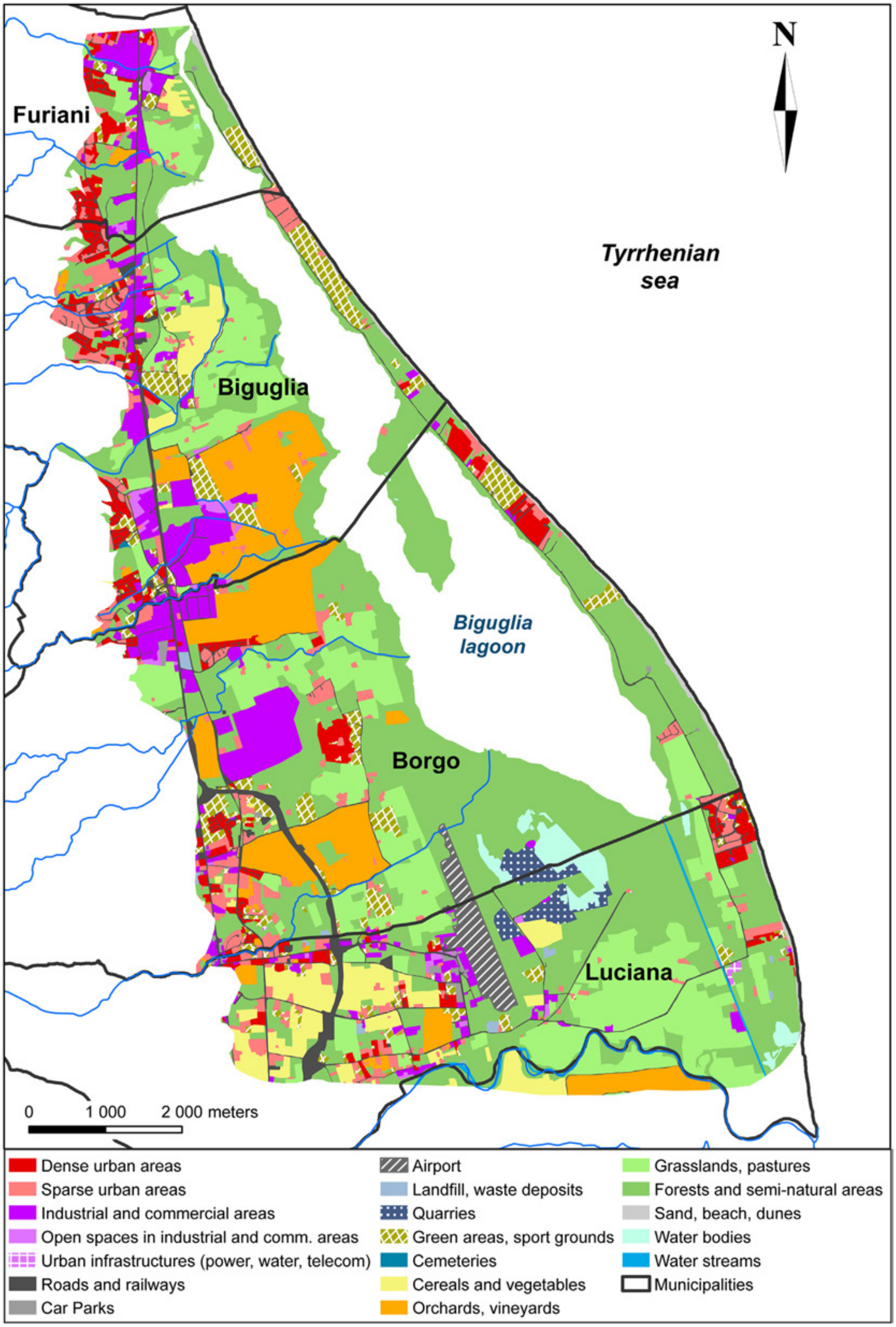

Fig. 4. Map of land use in the Biguglia watershed, adapted from Robert et al. (2015) and Graphical Parcel Register (ASP, 2012).

The maximum "no difference" area is observed for comparison of the two specific vulnerability assessment methods (SI vs Modified-DRASTIC: 74% of the area with same results) and for the two intrinsic vulnerability assessment methods (SINTACS vs DRASTIC: 69%). Therefore, adding the "land use" parameter has a major influence on the vulnerability maps.

For the intrinsic vulnerability assessment methods, the very high vulnerability class appears mainly between the coast and the Biguglia lagoon because of the combination of high ratings of almost all parameters (Supplementary Tables 1 to 8). This class is also strongly present in the south-eastern part of the watershed and along the entire bank of the lagoon,

Table 4
Descriptive statistics of vulnerability indexes computed and ranges of vulnerability classes (St. Dev. = Standard Deviation).

	Min.	Max.	Mean	St. Dev.	Vulnerability ranges				
					Very low	Low	Moderate	High	Very high
DRASTIC	53	213	157	38	53–85	86–117	118–149	150–181	182–213
SINTACS	71	238	177	33	71–104	105–138	139–171	172–205	206–238
Modified-DRASTIC	70	263	177	35	70–109	110–147	148–186	187–224	225–263
SI	26	95	66	11	26–40	41–54	55–68	69–81	82–95

especially for DRASTIC results. Here, the principal differences between DRASTIC and SINTACS methods are the "vadose zone parameter" which is more severe in DRASTIC. Fig. 6 also highlights that in some places groundwater is more vulnerable according to SINTACS than to DRASTIC, mainly in the north-western half of the watershed. This is due to the "hydraulic conductivity" parameter rates and to the "aquifer media" parameter that is more severe in the SINTACS method than in the DRASTIC one.

Between Modified-DRASTIC and SI results, differences are very low. For 7% of the study area, the groundwater vulnerability is higher according to Modified-DRASTIC than to SI. This can be explained by the absence of three parameters in SI (vadoze zone, K, soil media) and by the parameter weights applied in these two methods (Table 2). The same reasoning is applied for the places where SI notation is greater than in Modified-DRASTIC because the three parameters more in DRASTIC all have low

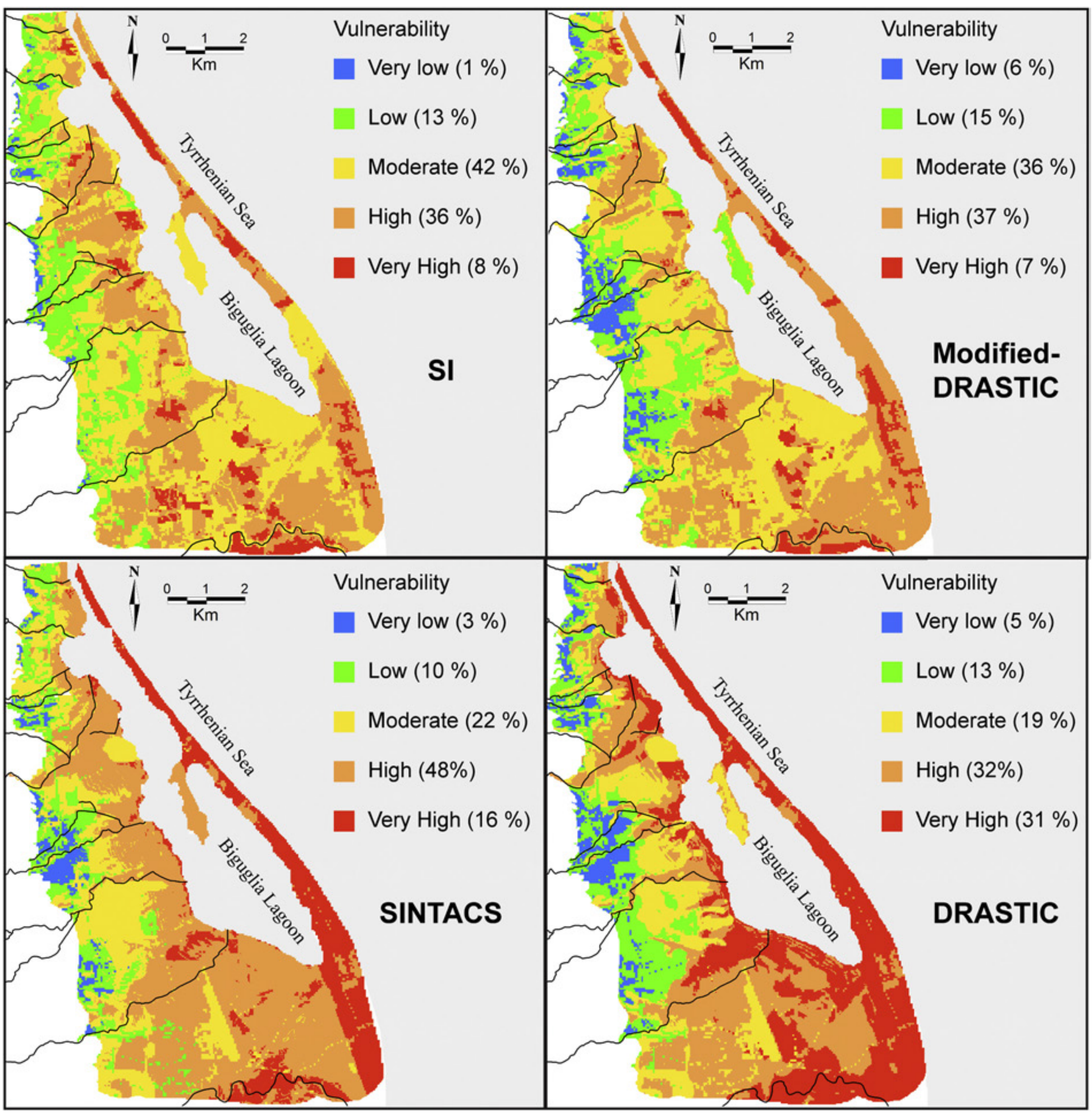

Fig. 5. Groundwater vulnerability maps of the Biguglia watershed resulting from the application of SI, Modified-DRASTIC, SINTACS and DRASTIC methods (percentage of the vulnerability classes area are given in parenthesis).

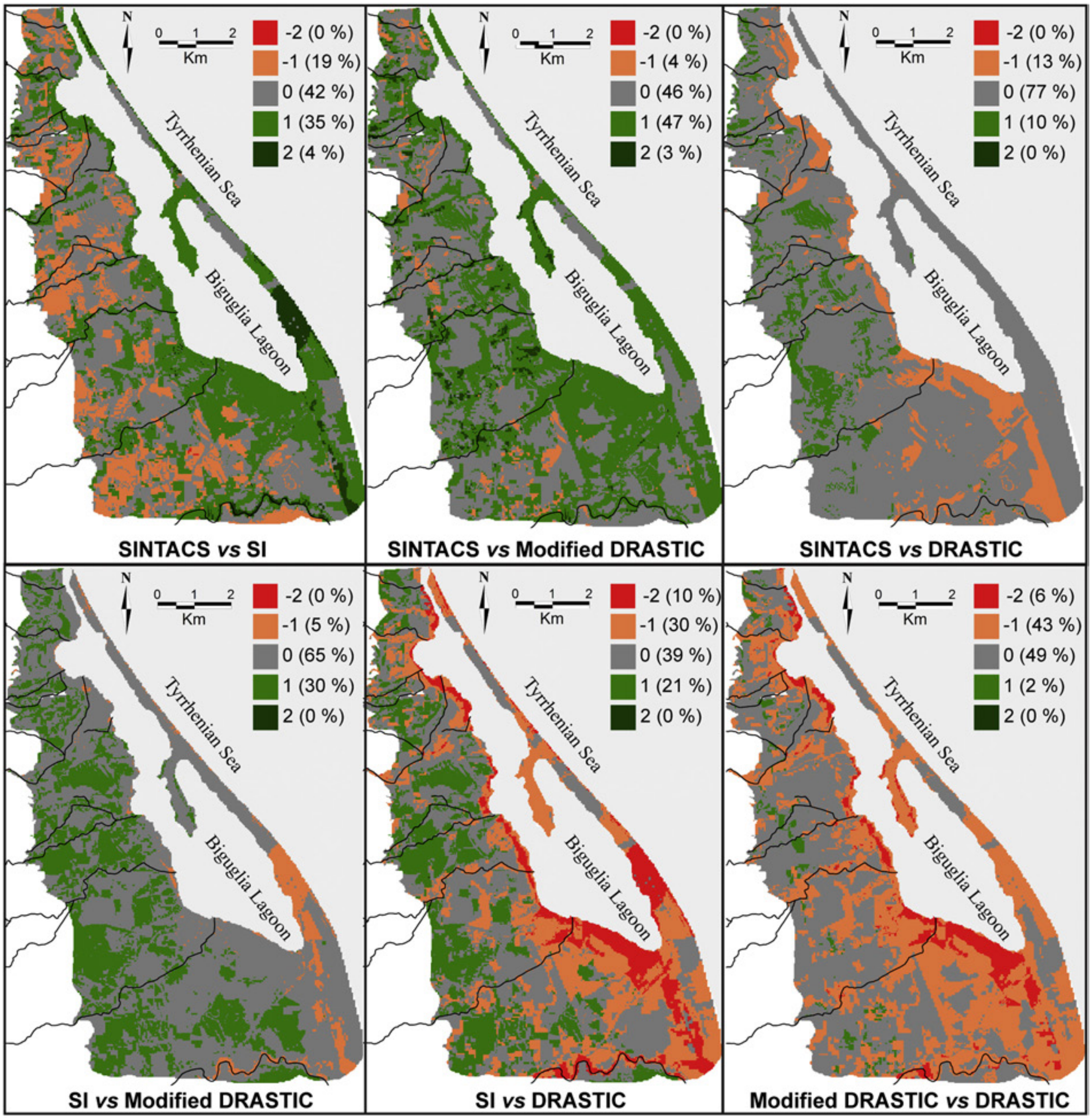

Fig. 6. Pairwise comparison of groundwater vulnerability classes from the 4 methods (area percentage of discrepancies is given in parenthesis).

ratings (4 for the "vadose zone media" parameter, 3 for the "hydraulic conductivity" parameter and 1 for the "soil" parameter).

When comparing results from intrinsic vulnerability assessment methods with those of specific vulnerability assessment methods, differences between parameter notations and weights exist, but the addition of "land use" also adds gaps; these are highlighted by the Fig. 6 (SINTACS vs SI, SINTACS vs Modified-DRASTIC, SI vs DRASTIC and Modified-DRASTIC vs DRASTIC). The major differences observed here are south of the Biguglia Lagoon (groundwater more vulnerable according to intrinsic vulnerability mapping method than the specific one) and along the western half of the watershed (groundwater more vulnerable according to specific vulnerability mapping method than the intrinsic one). South of the lagoon, the surface is occupied mainly by natural and semi-natural areas which compensate the unfavourable classification obtained through DRASTIC and SINTACS, minimizing the final groundwater vulnerability for Modified-DRASTIC and SI. In contrast, agricultural, industrial activities, and urbanized areas tend to increase the vulnerability in the west of the watershed for specific vulnerability methods.

3.3. Chemical validation

3.3.1. Groundwater quality

Nitrate concentrations measured in groundwater samples (Fig. 7) display a clear anthropogenic fingerprint on groundwater with some concentrations above the drinking water standards limit of 50 $mg.L^{-1}$ and most of the values are above the natural baseline of 5 mg L^{-1} (Appelo and Postma, 2005; Santoni et al., 2016). Highest concentrations can be found along the shore of the Tyrrhenian Sea, at the mouth of the Bevincu River and at the south-western end of the lagoon (Fig. 7 and Supplementary Table 9). These results are in good agreement with the results of Erostate et al. (2018) who clearly evidenced a nitrate contamination within the aquifer and managed to source and date the process. Erostate et al. (2018) also showed that the highest nitrate concentrations in the aquifer can in large part be attributed to anthropogenic/agricultural activities in the recent past. While the relatively low nitrate contamination in modern groundwater underlines that ongoing management practices to reduce surface nitrate pollution are efficient,

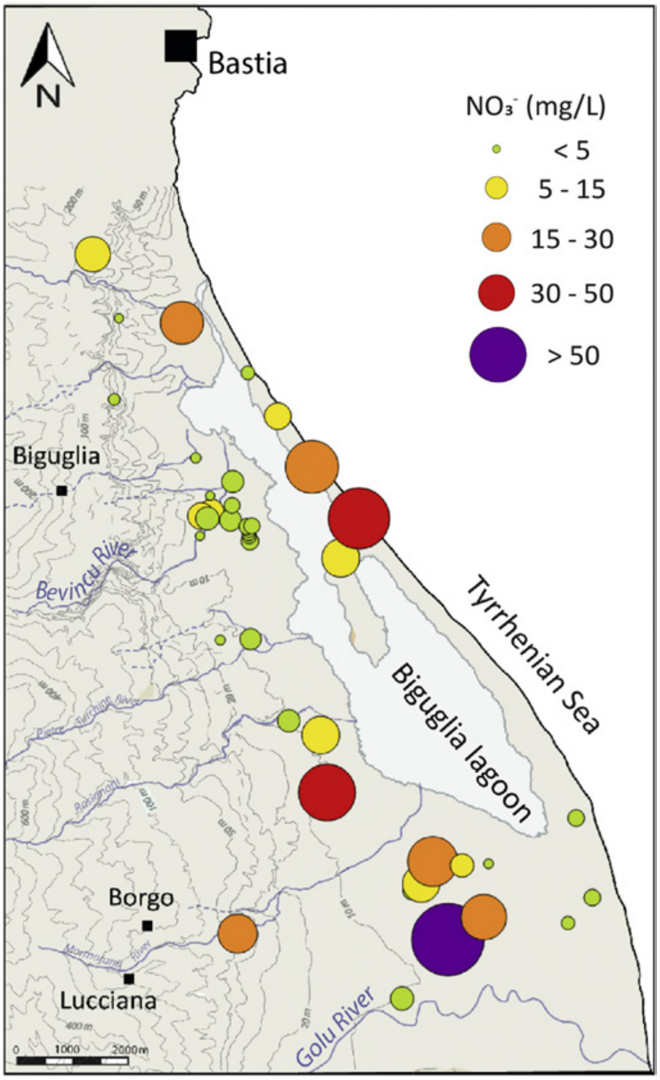

Fig. 7. groundwater nitrate concentrations for the 38 points of the monitoring network sampled in September 2016.

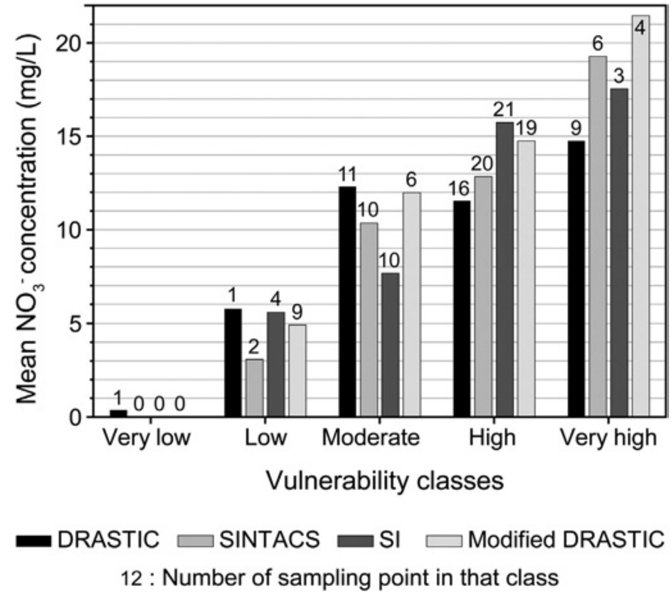

Fig. 8. Average groundwater nitrate concentrations function of vulnerability classes for the four applied mapping methods.

progressive nitrate contamination in “older” portions of the aquifer indicate the poor self-remediating capacity of the system. This geographical zonation is in agreement with the vulnerability maps of Fig. 5. Indeed, the sample sites with the highest NO_3^- concentrations are situated in the high or very high vulnerability classes.

3.3.2. Validation of vulnerability maps

In order to compare the groundwater vulnerability maps with the measured quality of the groundwater, the average groundwater NO_3^- concentrations were calculated for each vulnerability class. Thus, the concentration of groundwater from all the sample points mapped in very high vulnerability class for a method was averaged, and so on for the other ranges and methods. The results are represented in Fig. 8, in which the average NO_3^- concentrations are classified according to the vulnerability ranges of the sampled points. It is clearly noticeable that, for most methods, the NO_3^- average concentrations increase with increasing vulnerability. This confirms that index-based methods applied in this study are relevant for predicting aquifer vulnerability to pollution. However, for DRASTIC, the nitrate average concentration does not increase from moderate to high vulnerability ranges.

Furthermore, there are some discrepancies in the accuracy of these results regarding the Pearson correlation coefficient between the calculated average nitrate concentrations and the final vulnerability index. The Pearson correlation coefficient is the most commonly used correlation coefficient and is proposed to validate vulnerability mapping results by correlation with NO_3^- concentration in numerous studies (e.g.: Javadi et al., 2010; Güler et al., 2013; Wu et al., 2018). This coefficient is about 0.074 for DRASTIC, which confirms that this method is not the most appropriate in this context. It is about 0.12 for SINTACS, about 0.22 for SI and about 0.24 for Modified-DRASTIC. Then, Pearson factors are better for SI and Modified-DRASTIC results associated with NO_3^- groundwater concentrations. Nevertheless, these correlation coefficients are low, that can be explained by a lack of sampling points available for the determination of nitrate concentrations in groundwater. Furthermore, horizontal pollutant transfer is not considered in these methods and the transit time of groundwater in the aquifer can be underestimated in the diffusion of a pollution. Thus, today's low groundwater quality of the Biguglia catchment is to a large part due to legacy pollution from land-use practices several decades ago when land uses were quite different (between 1913 and 2012, urbanization has increased about 519.5% and agriculture has decreased about 61.2%; Table 1). Thus, the vulnerability maps proposed here are based on actual parameters while nitrate concentrations are a partly the legacy of former activities.

Finally, it is clear that Modified-DRASTIC and SI methods show the closest results to the geographical repartition of NO_3^- contamination. Therefore, despite low correlation coefficients between vulnerability classes and groundwater quality evolving a loss of accuracy, these methods describe well the contamination state of the groundwater in this hydrogeological and socio-economical context. The observed gap can be explained by the historical legacy of nitrates in the groundwater versus the current land use and soil parameters used to map the specific vulnerability. The specific vulnerability results are mapped with the current land cover situation, not with the land uses contemporaneous of the contamination.

4. Conclusion

In the hydrological context tested in this study, the DRASTIC method is more severe than the SINTACS one. For specific vulnerability, SI and Modified-DRASTIC results are close to each other, even if SI slightly minimizes vulnerability compared to modified-DRASTIC.

Between intrinsic and specific vulnerability mapping, the intrinsic one clearly shows more worrying results than the specific one. This is explained by the land use of the studied watershed, where natural protected areas and semi-natural environments compensate the high rates of intrinsic vulnerability. Finally, the comparison between groundwater nitrate concentrations and vulnerability classes allowed us to validate all the methods with a lower accuracy for the DRASTIC method.

Groundwater quality of the Biguglia watershed is dependent on both past activities (legacy traces of agricultural activities) and actual pressures (mainly sewage waters) with land uses that drastically evolved in the last few decades. Thus, the specific vulnerability results are representative of the actual situation in terms of pressure with the development of natural areas and the decrease in risky farming practices, but with a greater risk related to urban areas.

The specific vulnerability results are then more likely to represent the actual risks incurred by groundwater quality, with better accuracy for Modified-DRASTIC. Specific vulnerability methods are good tools to make an inventory of the groundwater pressures. In this case, the SI method is appropriate because it gives good results (almost equivalent to those of Modified-DRASTIC) with three fewer parameters to consider (i.e. significant time-saving). Nevertheless, intrinsic groundwater vulnerability methods are very useful to define priority protection areas in long-term space management planning. The SINTACS method seems to be the most appropriate in the Mediterranean and alluvial contexts comparable to this study. Even if more accurate techniques exist to specify aquifer vulnerability like artificial neural network methods or numerical hydrodynamic modelling, index mapping method are the most used in vulnerability groundwater management. We demonstrated that these methods are able to well describe the vulnerability situation of aquifers even in case of fast land use evolution implying ancient legacy contamination. This first-time-demonstration will have to be generalized in other aquifers subject to legacy contamination.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.12.249.

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6.3. Conclusion

Quelles que soient les méthodes, l'ensemble de l'aquifère se caractérise principalement par une **vulnérabilité modérée à très importante sur plus de 80% de sa superficie.** La classe de vulnérabilité très faible est très peu représentée avec seulement 1 à 6% de la surface de la plaine. Les superficies présentant une faible vulnérabilité sont également limitées et font consensus entre les méthodes (13% pour SINTACS, 18% pour DRASTIC, 14% pour SI et 21% pour DRASTIC-Modifié).

Des différences importantes résident cependant entre les méthodes d'évaluation de la vulnérabilité intrinsèque et spécifique. **Les classes de vulnérabilité élevée et très élevée sont les plus représentées pour les méthodes d'évaluation de la vulnérabilité intrinsèque SINTACS et DRASTIC, avec 63% et 64% respectivement, alors qu'elles ne représentent que 44% pour les méthodes d'évaluation de la vulnérabilité spécifique SI et DRASTIC-Modifié** (Tableau 2). D'après les méthodes d'évaluation de la vulnérabilité intrinsèque, et particulièrement la méthode DRASTIC, **le lido mais également la partie Sud-Est du bassin versant et les rives de la lagune, sont sujets à une vulnérabilité très élevée.** La classe de vulnérabilité modérée est en revanche beaucoup plus représentée par les méthodes SI et DRASTIC-Modifié, avec 42% et 36% respectivement, contre seulement 19% pour la méthode DRASTIC et 22% pour SINTACS (Tableau 2). **Ainsi, les résultats obtenus avec les méthodes d'évaluation de la vulnérabilité intrinsèque sont les plus alarmants, avec une expression majoritaire des classes de vulnérabilité élevée et très élevée.**

Tableau 2: Tableau récapitulatif des surfaces de la plaine de la Marana concernées par une vulnérabilité modérée ou élevée/très élevée.

	Vulnérabilité intrinsèque (surface en %)		**Vulnérabilité spécifique (surface en %)**	
	SINTACS	DRASTIC	SI	DRASTIC-Modifié
Elevée/Très élevée	63	64	44	44
Modérée	22	19	42	36

Résultats plus alarmants

La prise en compte du paramètre « occupation des sols » (vulnérabilité spécifique) a une influence majeure sur les cartes de vulnérabilité. Les principales différences sont observées au Sud de la lagune de Biguglia et à l'Ouest du bassin versant. Au Sud de la lagune, les eaux souterraines sont estimées comme plus vulnérables par les méthodes de cartographie de la vulnérabilité intrinsèque (DRASTIC et SINTACS). Cependant, l'occupation du sol principalement constituée de zones naturelles et semi-naturelles, entraîne une diminution importante de ce niveau de vulnérabilité par les méthodes DRASTIC-Modifié et SI. En revanche, à l'Ouest du bassin versant, la vulnérabilité estimée à l'aide de méthodes de cartographie de la vulnérabilité spécifique (DRASTIC-Modifié et SI) est accrue par la nature agricole, industrielle et urbaine de l'occupation des sols.

Afin d'évaluer la validité des cartographies de vulnérabilité, une inter-comparaison avec la qualité géochimique des eaux souterraines a été réalisée. **Les concentrations en NO_3^- les plus élevées concordent avec les secteurs caractérisés par une vulnérabilité élevée à très élevée, c'est-à-dire le lido, l'extrémité Sud-Ouest de la lagune et, dans une moindre mesure, la partie aval du Bevincu.** Afin de confirmer ces déductions cartographiques, les concentrations moyennes en NO_3^- mesurées dans les eaux souterraines ont été calculées pour chacune des classes de vulnérabilité. **Pour la plupart des méthodes (3/4), les concentrations moyennes en NO_3^- augmentent avec le degré de vulnérabilité.** Cette concordance confirme la pertinence des méthodes de cartographie à index pour la prédiction de la vulnérabilité des aquifères aux pollutions humaines. La méthode DRASTIC se distingue cependant par des concentrations moyennes en NO_3^- n'augmentant pas avec les classes de vulnérabilité modérée à élevée. Cette méthode apparait par conséquent comme peu appropriée dans le cas de l'aquifère de la Marana.

Afin de quantifier la cohérence entre le degré de vulnérabilité et les concentrations en NO_3^-, le coefficient de corrélation de Pearson a été calculé pour chaque méthode. Dans le cas de la méthode DRASTIC, la faible valeur du coefficient de corrélation de Pearson (0,074) confirme sa faible pertinence dans ce contexte. Cependant, bien que légèrement meilleures, les valeurs du coefficient de Pearson restent faibles pour l'ensemble des méthodes avec seulement 0,12 pour la méthode SINTACS, 0,22 pour la méthode SI et 0,24 pour la méthode DRASTIC-Modifié. Ces faibles valeurs peuvent s'expliquer notamment par le nombre insuffisant de points de prélèvement disponibles pour la détermination des concentrations en NO_3^- dans les eaux souterraines. De plus, le transfert horizontal de polluants n'est pas pris en compte avec les méthodes utilisées ici et le temps de séjour des eaux souterraines dans l'aquifère peut-être largement sous-estimé en cas de pollution diffuse.

Malgré les faibles coefficients de Pearson, les cartographies de vulnérabilité obtenues par les méthodes DRASTIC-Modifié et SI donnent des résultats très proches de la répartition géographique de la contamination en NO_3^- observée. **Les méthodes de cartographie de la vulnérabilité spécifique reflètent ainsi bien l'état de dégradation actuel des eaux souterraines dans le contexte hydrogéologique et socio-économique de l'aquifère de la Marana, autrement dit dans le cas d'un aquifère alluvial côtier soumis à une forte pression anthropique, dont la nature et l'importance ont considérablement évolué au cours des dernières décennies**. L'écart observé, traduit par les faibles coefficients de Pearson, peut alors s'expliquer par l'héritage historique en NO_3^- dans les eaux souterraines. En effet, la dégradation actuelle des ressources en eaux souterraines du bassin versant est en grande part liée aux legs nitratés hérités des pratiques et de l'occupation des sols mises en place dans les années 1960 (voire chapitre 5). Or, les cartes de vulnérabilité établies dans cette étude sont basées sur les paramètres et l'occupation des sols actuels. Les résultats de la vulnérabilité spécifique représentent donc la situation actuelle de la couverture du sol, et non la situation contemporaine de la contamination.

Les méthodes de cartographie de la vulnérabilité spécifique apparaissent comme les plus pertinentes dans le cas de l'aquifère de la Marana, pour évaluer la qualité des eaux souterraines et prendre en compte l'aspect historique de la dégradation. Les méthodes d'évaluation de la vulnérabilité intrinsèque des eaux souterraines sont très utiles pour la détermination des zones de protection prioritaires dans la planification territoriale à long terme (Tableau 3). Dans cette optique, la méthode SINTACS semble être la plus appropriée pour l'aquifère de la Marana mais également dans le cas des aquifères méditerranéens et alluviaux comparables à cette étude.

Tableau 3: Méthodes de cartographie à privilégier dans le cas de l'hydrosystème de la lagune de Biguglia.

Vulnérabilité intrinsèque	Vulnérabilité spécifique
Détermination des zones de protection prioritaires dans la planification territoriale à long terme	Evaluation de la qualité des eaux souterraines tenant compte de l'aspect historique de la dégradation

Conclusion & Perspectives

Conclusion

Par leur aspect appliqué, les travaux menés dans le cadre de cette thèse ont permis l'étude détaillée de la composante hydrogéologique de l'hydrosystème littoral de la lagune de Biguglia. La pertinence, les atouts et les limites des outils hydrologiques utilisés revêtent eux d'une recherche à caractère plutôt fondamental, permettant d'apporter de nouveaux éléments de réflexion quant à la pertinence des traceurs des flux en hydrogéologie. De cette double approche est née **un premier modèle conceptuel à grande échelle, permettant de mieux appréhender le fonctionnement hydrogéologique de l'aquifère et sa continuité hydraulique avec la lagune de Biguglia.**

1. Fonctionnement global de l'hydrosystème de la lagune de Biguglia

- ### Condition et quantification de la recharge

La corrélation entre les signatures isotopiques des isotopes stables de la molécule d'eau dans les précipitations et les eaux souterraines a permis d'élaborer une droite météorique locale caractérisant la signature des eaux de pluie. L'usage de ces isotopes a également permis de mettre en évidence **la complexité de la recharge autochtone et allochtone dans le cas de l'hydrosystème de la lagune de Biguglia**. Si le Sud du bassin versant profite d'une recharge autochtone par l'infiltration directe des précipitations sur la plaine, le Nord du bassin versant bénéficie lui d'une recharge allochtone par les précipitations en provenance des contreforts schisteux.

Ces résultats confirment la pertinence des isotopes stables de la molécule d'eau pour la compréhension des processus de recharge complexe, dans le cas des bassins versants à fort contraste altitudinal.

- ### Temps de résidence des eaux souterraines

Mises en regard des teneurs en ^3H mesurées dans les pluies, les teneurs en ^3H des eaux souterraines ont permis d'obtenir une information qualitative sur leur temps de séjour. Grâce à la corrélation développée entre les concentrations en CFCs et les teneurs en ^3H, **il a été possible de proposer un temps de séjour estimé par calcul pour chacune des eaux souterraines échantillonnées.** Ainsi, les eaux souterraines du Sud de la plaine alluviale, du lido et des contreforts schisteux présentent des temps de séjour longs et une dynamique particulièrement inertielle. A l'inverse, les eaux souterraines de la partie nord de la plaine alluviale et des alluvions du Golu se caractérisent par des temps de séjour plus faibles, liés à l'infiltration des eaux de surface en provenance des rivières.

Malgré les influences pouvant affecter les concentrations en CFCs, ces traceurs peuvent prodiguer des informations complémentaires très utiles dans le cas des aquifères alluviaux fortement anthropisés. Il convient cependant de faire preuve de la plus grande précaution au moment de l'interprétation des données. La corrélation avec d'autres traceurs/indicateurs des dynamiques et des temps de séjour des eaux souterraines constitue une étape essentielle et *sine qua non*.

- **Quantification des interactions entre les masses d'eau**

Grâce aux concentrations géochimiques et aux signatures isotopiques très contrastées entre les eaux de surface (faible concentration ionique/signature isotopique très appauvrie), les eaux souterraines en provenance des formations schisteuses (concentration ionique élevée/signature plus enrichie) et l'eau de mer (concentration ionique très élevée/signature très enrichie), **un modèle de mélange a pu être élaboré afin de mieux quantifier et évaluer i) la contribution des eaux fluviales à la recharge de l'aquifère, ii) le rôle des contreforts schisteux bordant l'hydrosystème à l'Ouest, iii) les relations aquifères/lagune et iv) les processus d'intrusion saline.**

En plus d'apporter des éléments de compréhension hydrodynamique, l'élaboration de ce modèle montre la pertinence du couplage géochimique et isotopique pour l'estimation semi-qualitative des mélanges au sein des aquifères alluviaux côtiers. A l'aide de paramètres très communément mesurés en hydrogéologie, faciles d'obtention et relativement peu couteux (HCO_3^-, Cl^- et $\delta^{18}O$), il est donc possible de préciser de manière fiable les processus de mélange complexes au sein de l'aquifère (incertitude <10%).

- **Dépendance de la lagune aux eaux souterraines**

Il apparaît de manière claire que **la lagune est partiellement tributaire des eaux souterraines**. Les processus de décharge de l'aquifère directement dans la lagune semblent cependant très fortement contraints et quasi-totalement contrôlés par le système de drainage artificiel de la plaine. **L'apport des eaux souterraines à la lagune est principalement assuré par les stations de pompage. Celles-ci pompent l'eau des**

canaux drainant massivement la nappe et la rejette vers la lagune. Les eaux fluviales participent également de manière significative aux apports en eau douce vers la lagune. Actuellement, il n'est cependant pas possible de faire la distinction entre i) les écoulements directs des eaux fluviales vers la lagune et ii) les processus d'infiltration des eaux fluviales, transitant via l'aquifère, avant de rejoindre la lagune. La proportion d'eau souterraine contribuant aux apports en eau douce à la lagune est donc très probablement sous-estimée.

- ### Modèle conceptuel de fonctionnement de l'hydrosystème

Les eaux de l'aquifère résultent principalement de i) l'infiltration des eaux de rivière du Bevincu et du Golu, ii) la contribution latérale en provenance des eaux souterraines des contreforts schisteux de la Corse alpine et iii) la double recharge allochtone et autochtone par les précipitations. Le système de drainage développé sur la plaine modifie grandement le fonctionnement hydrodynamique de l'aquifère. Les canaux de drainage bordant la lagune constituent une charge imposée pour l'aquifère, accélérant l'écoulement naturel des eaux souterraines. Il est donc important de préciser ici que **les canaux de drainage ne sont pas seulement des collecteurs des eaux de ruissellement de surface mais drainent également une part importante des eaux souterraines en provenance de l'aquifère (Figure 18).** Les travaux menés permettent maintenant de proposer un premier modèle conceptuel de fonctionnement de l'hydrosystème de la lagune de Biguglia dans sa globalité, à grande échelle (Figure 19).

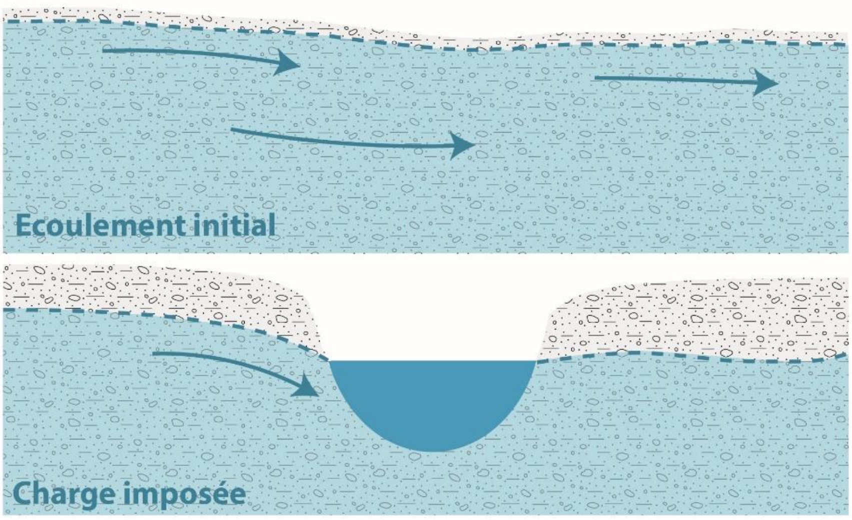

Figure 18: Illustration de l'effet de « charge imposée » des canaux sur les eaux souterraines.

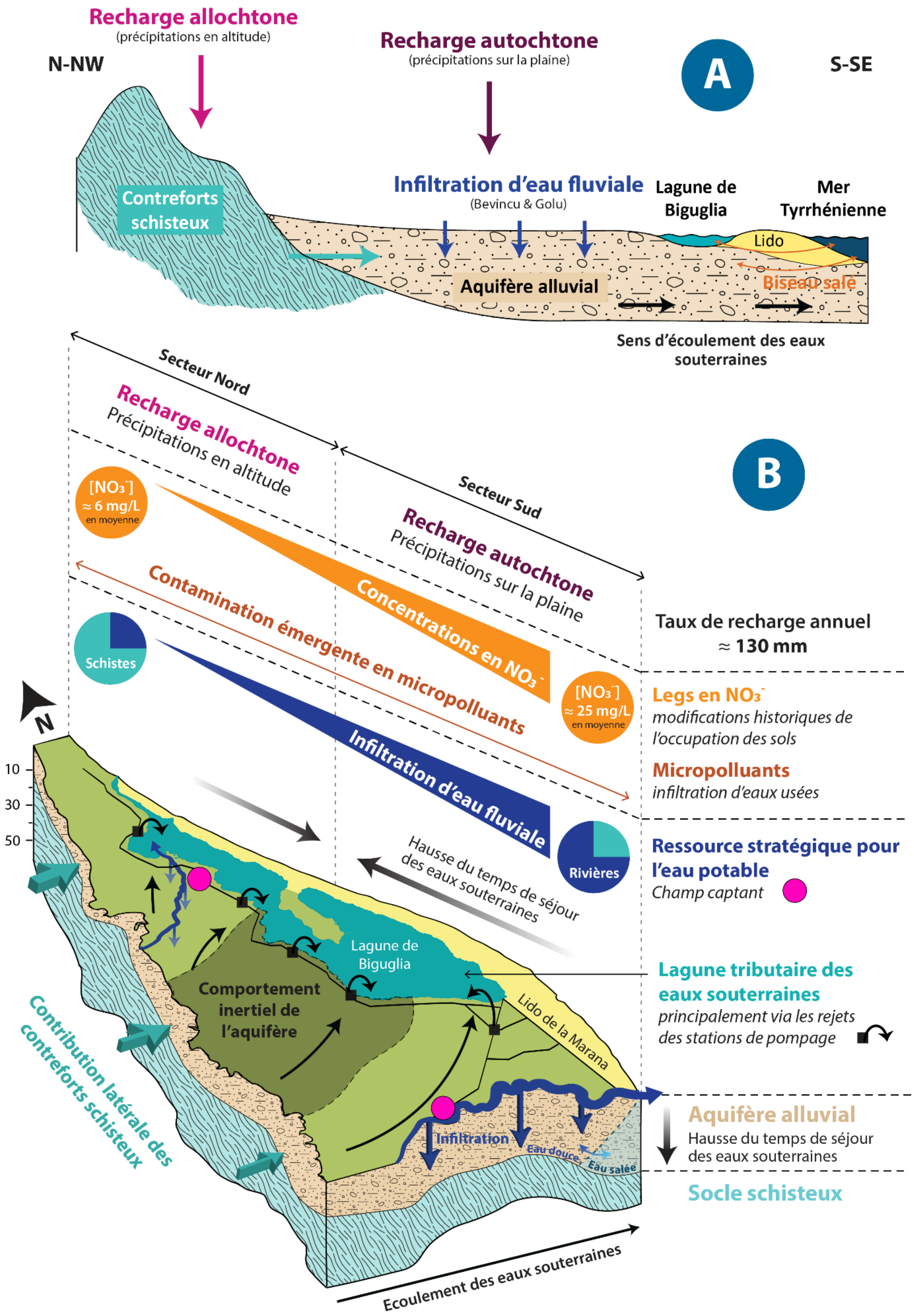

Figure 19: Vue en coupe de l'hydrosystème de la lagune de Biguglia présentant les principaux processus hydrologiques (A) et schéma conceptuel de fonctionnement global de l'hydrosystème (B).

2. Identification et traçage de l'origine des polluants

Les concentrations en NO_3^- attestent d'une contamination anthropique diffuse sur l'ensemble de la plaine. **La corrélation entre les concentrations en NO_3^- et les teneurs en ^3H a permis d'évaluer la dynamique temporelle des polluants nitratés dans les eaux souterraines.** Ainsi, on constate une augmentation des concentrations en NO_3^- avec le temps de résidence des eaux souterraines. A l'inverse, les eaux souterraines avec des temps de séjour courts, montrent les concentrations en NO_3^- les plus faibles. **Les polluants nitratés sont donc accumulés et stockés dans l'aquifère au cours du temps.** Les eaux souterraines, en s'écoulant vers la lagune, constituent alors un vecteur potentiel de pollution à prendre en compte. La diminution significative des teneurs en NO_3^- dans les eaux souterraines les plus récentes témoigne d'une amélioration des pratiques et des stratégies de gestion à l'échelle du bassin versant, permettant de réduire les flux azotés vers l'aquifère. La lagune de Biguglia continue cependant de présenter des teneurs en NO_3^- excessivement fortes (Orsoni et al., 2001; Souchu et al., 2010; Pasqualini et al., 2017).

Si les concentrations en NO_3^- constituent d'elles-mêmes un indicateur de dégradation anthropique, **les isotopes du NO_3^-, ^{15}N-NO_3^- et ^{18}O-NO_3^-, et du bore ^{11}B ont permis de retracer les sources des flux nitratés.** Deux sources majeures ont pu être mises en évidence : une origine naturelle depuis le sol et une origine anthropique liée à l'impact des eaux usées. Les processus de dénitrification sont faibles et extrêmement localisés. L'aquifère se caractérise donc par un faible potentiel d'élimination des NO_3^-, permettant ainsi leur accumulation.

L'usage couplé des isotopes du nitrate ^{15}N-NO_3^-, ^{18}O-NO_3^- et du bore ^{11}B permet une bonne appréhension des sources de contaminations dans le cas des aquifères alluviaux fortement anthropisés. Le ^{11}B est essentiel à l'interprétation des données isotopiques du nitrate car il permet de s'affranchir des biais liés aux processus de dénitrification. **La corrélation entre les isotopes du nitrate et les teneurs en ^3H permet une approche historique de la pollution, bien au-delà du simple constat qualitatif de la ressource à un instant donné.** Elle permet de mettre en évidence les legs historiques en polluants, conséquences des aménagements et des pratiques passés, qui continuent de mettre en péril la qualité actuelle de la ressource en eau. **Dans le cas des aquifères alluviaux**

côtiers fortement anthropisés et sujets à un développement toujours plus important, le potentiel d'archivage des eaux souterraines constitue une information clé pour l'instauration de stratégies de gestion pertinentes assurant la pérennité future des ressources en eau.

3. Pertinence des micropolluants organiques pour le traçage des flux hydrologiques

L'amélioration constatée au niveau des concentrations en NO_3^- est contrebalancée par la découverte de micropolluants organiques dans l'ensemble des ressources en eau de l'hydrosystème. Ces polluants, d'origine uniquement anthropique, démontrent la vulnérabilité du système. **L'importante urbanisation de la région représente actuellement la principale menace pour l'aquifère alluvial.** Même si les concentrations en micropolluants organiques sont très élevées dans les eaux usées, leurs concentrations dans les eaux naturelles sont relativement faibles, témoignant d'une contamination encore limitée.

La détection de micropolluants organiques dans les eaux souterraines de la Marana a permis de préciser les zones d'infiltration des eaux de surface et donc de recharge de l'aquifère. **La présence de micropolluants organiques très rapidement dégradables témoigne de processus d'infiltration rapide et met en exergue la forte transmissivité de l'aquifère alluvial de la Marana.** Les alluvions favorisent localement l'infiltration rapide de pollutions vers la zone saturée, ce qui augmente considérablement la vulnérabilité de la ressource. La partie Sud du bassin versant et le lido, où l'urbanisation et le réseau d'assainissement des eaux usées sont largement développés, semblent particulièrement concernés.

A l'heure actuelle, la complexité des processus et des paramètres entraînant la conservation, la transformation ou la remédiation des micropolluants organiques fait encore l'objet de nombreuses recherches. Les micropolluants ne peuvent donc pas constituer à eux seuls des traceurs d'une fiabilité suffisante. Cependant, **corrélés à des traceurs plus conventionnels et mieux maîtrisés, les micropolluants organiques peuvent apporter des informations complémentaires concernant i) le traçage des eaux récemment infiltrées, ii) l'identification des zones de recharge et iii)**

l'amélioration de l'évaluation de la vulnérabilité des aquifères fortement anthropisés.

4. Importance du travail collaboratif

- **Reconstitution des trajectoires socio-environnementales**

De par leur complexité, les hydrosystèmes littoraux requièrent une approche transdisciplinaire afin de comprendre l'évolution du système dans son ensemble. **La corrélation des données géochimiques avec les dynamiques socio-économiques d'aménagement du territoire a permis d'identifier les liens de causalité entre le développement anthropique et l'état des ressources**. Il a alors été possible de retracer la chronologie des processus de dégradation et d'identifier l'évolution progressive des sources de pollutions au cours du temps. Historiquement, la dégradation des eaux souterraines est principalement causée par la perturbation massive des sols, liée à la modification des pratiques agricoles depuis les années 1950 et à la construction de nouveaux espaces urbanisés. La déstabilisation des sols a conduit à un lessivage massif des NO_3^- qui se sont progressivement accumulés dans l'aquifère. Depuis les dernières décennies, l'infiltration d'eaux usées non traitées vers l'aquifère représente la principale source de dégradation des eaux souterraines sur le secteur d'étude. L'extension importante du réseau d'assainissement en réponse à l'urbanisation désorganisée de la plaine a entraîné la multiplication des fuites sur le réseau. **Les eaux souterraines agissent donc comme une « archive » des activités anthropiques développées sur la plaine et leur dégradation actuelle résulte des politiques d'aménagement menées sur les 70 dernières années.**

- **Vers un travail collaboratif entre scientifiques et gestionnaires**

Les résultats acquis dans le cadre de cette thèse montrent une dégradation diffuse et actuellement en cours des eaux souterraines de l'aquifère de la Marana, due à l'ensemble des activités humaines développées sur le bassin versant. Bien que largement propre à la consommation et présentant des concentrations encore relativement faibles en polluants, il apparait cependant comme essentiel d'enrayer la dégradation de cette ressource stratégique. **Mais plus important encore, ces travaux démontrent la forte interdépendance existante entre les eaux souterraines, fluviales et lagunaires et ainsi le besoin crucial d'améliorer la prise en compte des eaux souterraines dans**

les stratégies de gestion de l'hydrosystème. Il est maintenant nécessaire que les décisionnaires et les gestionnaires s'approprient pleinement la complexité des interactions entre les masses d'eau. La lagune de Biguglia ne dépend pas uniquement des apports en eau de surface. **Les apports en eau souterraine, via les stations de pompage, contribuent aux conditions physico-chimiques de la lagune ainsi qu'au drainage des polluants (notamment nitratés) vers la lagune. Les eaux souterraines contribuent ainsi activement à l'ensemble des services écosystémiques produit par la lagune. La prise en compte du continuum hydrologique dans son ensemble constitue un paramètre essentiel à la préservation et la restauration de la lagune de Biguglia.**

L'état qualitatif des eaux souterraines est donc une problématique essentielle autant pour l'alimentation en eau potable que pour la restauration écologique de la lagune. En accord avec l'objectif premier du SAGE, visant à « lutter contre toutes les pollutions, notamment diffuses, pouvant impacter le bon état des milieux aquatiques » (SAGE, 2012), les résultats présentés ici doivent interpeller les gestionnaires afin de prendre, ou plus exactement d'appliquer véritablement, des mesures de gestion permettant la pérennisation de la qualité de la ressource en eau. Le SAGE présente un ensemble de mesures pertinentes qu'il conviendrait de mettre en œuvre plus rigoureusement. Les actions menées pour l'amélioration et la réhabilitation des systèmes d'assainissement individuel et collectif (mesures n°10 et n°11 du SAGE) doivent être poursuivies. **La mise en place d'une urbanisation réfléchie et planifiée, permettant l'instauration d'un système de traitement optimisé des eaux usées doit constituer une des priorités à l'échelle du bassin versant.** Ce point n'est que très rapidement évoqué dans le SAGE (« les documents d'urbanisme seront compatibles ou rendus compatibles avec le SAGE ») et le mitage du territoire, entraînant l'extension toujours plus importante du réseau d'assainissement, ne fait l'objet d'aucune mention.

Une attention particulière doit être portée à l'optimisation de la gestion et du fonctionnement des stations de pompage. Mis en place dès 19ème siècle, suite à l'application de la loi de 1807, le réseau de drainage artificiel continue d'assécher la plaine pour limiter la prolifération des moustiques suivant un niveau seuil (environ 0,8 m) très proche du niveau seuil établi dans le années 1880 (0,6 m). Cependant, les décennies de drainage et de pompage ont considérablement contraint le fonctionnement hydrologique naturel du système. Compte tenu de l'avancée des connaissances concernant l'hydrosystème, il serait propice d'initier des réflexions quant à l'actualisation du système de drainage afin vérifier son adéquation avec l'état actuel de la ressource. Il conviendrait notamment de préciser la nature des flux drainés par les stations de pompage et d'adapter le seuil de pompage (niveau d'activation des poires) au cas par cas, en fonction du niveau des eaux souterraines environnantes. Il se pourrait notamment que les problèmes de salinisation des terres constatés à proximité des stations de pompage et de la lagune soient la résultante de pompage préférentiel des eaux lagunaires, facilitant leur avancée dans les terres.

Enfin, les eaux souterraines drainées par les canaux ceinturant la lagune et les stations de pompages sont rejetées vers la lagune. Cette ressource de bonne qualité, normalement utilisée pour l'alimentation en eau potable, est ainsi dégradée par salinisation. **Les volumes extraits constituent donc une perte directe de la ressource en eau douce.** La quantification des volumes d'eau extraits de l'aquifère via les stations de pompage permettrait d'évaluer cette perte. La réalisation de quelques travaux prospectifs au cours de cette thèse, via des mesures ponctuelles, a permis d'effleurer la complexité des mélanges des eaux pompées. L'étude des volumes d'eau souterraine extraits par les stations de pompage nécessite à minima la mise en place d'un suivi continu des conditions physico-chimiques voire des isotopes stables de l'eau.

Perspectives

Ces travaux ont également soulevé plusieurs interrogations et pistes de réflexion qu'il reste encore à éclaircir.

- **Précision de l'état qualitatif de la ressource**

Un nombre limité de polluants a été recherché au cours de ces travaux. Seul les micropolluants organiques d'origine domestique ont été analysés. Au vu du rôle actuellement prédominant des eaux usées dans la dégradation qualitative des ressources en eau, il serait approprié de procéder à la recherche complémentaire de micropolluants d'usage domestique courants, des hormones et des micro/nano plastiques. Ces polluants induisent une dégradation chronique des ressources en eau et impactent durablement l'ensemble des écosystèmes terrestres et aquatiques. Il serait également pertinent d'étendre la recherche aux micropolluants organiques d'origine agricole et industrielle, dont la présence est fortement suspectée.

- **Amélioration de la connaissance quantitative**

La réalisation d'un bilan hydrique sur l'aquifère reste un véritable challenge, en grande partie à cause de l'absence de connaissances quant aux volumes extraits. Pourtant, **dans le contexte actuel de changement climatique, caractériser les flux entrants et sortants apparaît comme un élément clé, nécessaire à la gestion pérenne des ressources**. Il s'agit à la fois d'évaluer les volumes extraits pas les stations de pompage mais aussi de mieux quantifier les volumes extraits à partir des ouvrages privés. Il est important de préciser le nombre d'ouvrages présents sur la plaine mais surtout de mieux contraindre les dispositifs de pompage actuellement fonctionnels.

- **Quantification des flux de nutriments en provenance des eaux souterraines**

La lagune de Biguglia se caractérise par des concentrations en nitrate extrêmement élevées. Après avoir démontré le stockage des nitrates dans l'aquifère et la dépendance de la lagune aux eaux souterraines, **la quantification des flux de nutriments via les eaux souterraines permettrait de mieux contraindre les risques de dégradation futur de l'écosystème lagunaire**. La quantification des flux de nutriments est intimement liée à la quantification des volumes d'eau souterraine rejoignant la lagune.

Les méthodes courantes de quantification des décharges d'eau souterraine en milieux côtiers et lagunaires (notamment par le couple radon/radium) pourraient cependant s'avérer être en limite de fiabilité dans le contexte de l'aquifère de la Marana (décharge naturelle minoritaire face aux apports via les stations de pompage). Une réflexion quant aux meilleures stratégies de quantification doit donc être amorcée.

- **Modélisation numérique de l'hydrosystème de la lagune de Biguglia**

La modélisation numérique de l'hydrosystème apparaît comme une perspective de recherche importante. Bien que déjà appréhendée au cours de cette thèse, la complexité de l'hydrosystème et des conditions aux limites ainsi que la mauvaise connaissance de la géologie du secteur (épaisseur, extension et délimitation exacte des terrasses alluviales notamment) rend la modélisation particulièrement ardue et chronophage. Un important travail stratigraphique via l'étude des données disponibles (sondage, forages, puits…) doit être envisagé pour permettre l'élaboration d'un modèle en 3-dimension de qualité. Une fois ce modèle réalisé, les résultats de la modélisation numérique pourront ensuite être comparé aux résultats géochimiques. Bien que fastidieux, **ces travaux pourraient permettre une première quantification des flux au sein de l'hydrosystème mais également le traçage des zones d'infiltration des polluants azotés. Cette modélisation serait à la fois un excellent outil pour valider par le calcul la véracité des conclusions tirées grâce aux données géochimiques mais prodiguerait également des informations quantifiées utiles à l'instauration d'une gestion durable de la ressource**.

- **Etude prospective des effets de l'élévation du niveau marin**

D'après les projections réalisées, de nombreux littoraux pourraient être impactés par l'élévation du niveau marin. Si le risque d'inondation marine est souvent évoqué en premier, le risque d'inondation par les eaux souterraines serait probablement le premier à impacter les zones littorales (Rotzoll and Fletcher, 2013). En effet, pour les aquifères libres dont le niveau piézométrique est proche de la surface, comme c'est le cas pour l'aquifère de la Marana, l'augmentation du niveau marin induira une élévation simultanée de la surface piézométrique, pouvant dépasser la surface topographique. Ce processus pourrait alors induire des problèmes importants et couteux, liés à la gestion de l'eau (salinisation des aquifères) et des infrastructures (inondations). **La réalisation de plusieurs scénarii prospectifs d'élévation du niveau marin sur la plaine de la Marana pourrait alors permettre d'évaluer les risques et les potentielles mesures à mettre en place**.

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