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Meriem Djouhri

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Meriem Djouhri. Modélisation des processus de distribution et de dérive des pesticides lors de la pulvérisation et de leur contribution à l'exposition des passants : illustration en viticulture. Sciences de la Terre. Université de Montpellier, 2022. Français. NNT : 2022UMONG073 . tel-04087624

**HAL Id: tel-04087624**

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Submitted on 3 May 2023

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# THÈSE POUR OBTENIR LE GRADE DE DOCTEUR DE L'UNIVERSITÉ DE MONTPELLIER

En Sciences du Sol

École doctorale GAIA – Biodiversité, Agriculture, Alimentation, Environnement, Terre, Eau

Unité mixte de recherche LISAH

**Modélisation des processus de distribution et de  
dérive des pesticides lors de la pulvérisation et de leur  
contribution à l'exposition des passants : illustration  
en viticulture**

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**Le 16 Décembre 2022**

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**UNIVERSITÉ  
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## Remerciements

Ces mots seront les premiers à être lus, pourtant, ce sont les derniers que j'ai écrits.

Après quatre années, cette incroyable aventure touche à sa fin. Difficile d'écrire ces remerciements, mais au même temps, c'est un plaisir de remémorer les souvenirs me liant à chacune des personnes ayant scientifiquement contribué à ce travail ou bien émotionnellement à travers un soutien infaillible.

En premier lieu, je tiens à remercier mon directeur de thèse, Marc Voltz, pour sa confiance, sa rigueur, ses conseils toujours avisés et le savoir qu'il a su me transmettre au cours de ces quelques années. Merci également à mes encadrants, Carole Bedos et Jean-Paul Douzals, pour leurs connaissances, disponibilité et bienveillance. Un merci particulier à Carole qui m'a suivie depuis mon stage de master et qui a toujours cru en moi. Je remercie également Benjamin Loubet qui sans lui, j'aurais été perdue dans le monde de la dispersion atmosphérique (j'aurais dérivé !) et Cécile Dagès qui était là tant pour le soutien scientifique mais aussi psychologique. Un grand merci à David Crevoisier, pour son aide précieuse, sa patience pour répondre à chacune de mes questions et pour les veillées bénévoles pour m'aider dans mon travail.

J'en profite également pour remercier l'ensemble des membres de mon comité de suivi, Yves Brunet, Jean-Stéphane Bailly, Benjamin Loubet et Flavie Cernesson, pour avoir pris le temps d'échanger sur mes travaux, et dont l'aide m'a été précieuse. J'adresse aussi mes remerciements à l'ensemble des membres de mon jury de thèse et plus particulièrement à mes deux rapporteurs, Sylvain Dupont et Emilio Gil, pour avoir pris le temps d'évaluer mon travail.

Après ces quatre années passées au sein de l'UMR LISAH, je remercie l'ensemble du personnel pour leur accueil chaleureux et les échanges que nous avons pu avoir. Une pensée particulière pour l'ensemble du pôle administratif, et spécialement à Nadia et Azziza, pour m'avoir accueillie avec une bienveillance remarquable et pour m'avoir aiguillée dans les rouages de l'administration au cours de ma présence au sein du laboratoire. Mais aussi pour m'avoir soutenue, entendu râler ou bien conseillée. Je remercie également l'équipe de l'UMR Ecosys de m'avoir chaleureusement accueillie lors de mes missions à Paris.

Un remerciement particulier à l'équipe de choc des doctorants où d'une part, nous avons un défenseur des droits des friends « Pauline », un ange gardien, bien veillant « Guillaume », une baby-sitter sauveteuse « Mariem » et le petit rayant de soleil joyeux qui nous rend visite « Bruce ». Discret, mais toujours là « Abdel Rahman » et la voie de la sagesse. Un énorme merci à vous pour votre présence et tous les bons moments partagés.

Une petite pensée à Pauline qui m'a toujours écoutée et soutenue même dans les moments où j'étais la plus insupportable.

## Remerciements

---

Une petite pensée à Rostom, Amel, Hakan, Asma, Amin, Sousou, Ghania et ma belle-famille qui m'ont toujours soutenu.

À mes sœurs qui tout autant que ma regrettée mère ont attendu le jour où je ferais mon premier pas dans la recherche. Je les remercie d'avoir cru en moi dans les jours ou même moi, j'en doutais.

À mes trois autres amies et sœurs avec qui j'ai partagé mes moments de faiblesse, mais aussi mes moments de joies durant ces années.

À Mina qui me faisait des pause anti-stress même à distance, avec des anecdotes les unes plus folles que les autres. À Melya pour son soutien expérimenté et pour m'avoir pardonné d'avoir raté l'événement le plus important de sa vie pour que je finisse ce travail. À Amina qui m'a soutenue durant mon master et continue d'être présente pour moi.

À Tarik qui a su m'accompagner tout au long de ces années de thèse. Je le remercie pour avoir su absorber mon stress et l'apaiser, mais aussi pour les cours gratuits de programmation. À ma petite Myral qui a apporté une note de joie innocente à mes journées.

*“In memory of my mother who would have loved to see me one day as a great researcher, even though she is not present among us today, I dedicate this first step in research to her while being confident that from where she is observing us she will be proud of it”*

---

***If I have to choose words to cover this life experience, these  
statements would be a nice beginning***

*“Raise your words, not your voice. It’s the rain that makes the flowers grow, not the thunder”*

— Djalâl ad-Dîn Rûmixi

*“There is no painting without drawing and there is no shape without line”*

— El-Sahali

*“The aim of argument, or of discussion, should not be victory, but progress”*

— Joseph Joubert

*“Our fatigue is often caused not by work, but by worry, frustration and resentment”*

— Dale Carnegie

*“Shoot for the moon. Even if you miss, you’ll land among the stars”*

— Oscar Wilde



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## Résumé

La maîtrise de la contamination des différents compartiments environnementaux et de l'exposition des différents êtres vivants par les pesticides requiert une bonne compréhension des processus majeurs impliqués. C'est le cas pour optimiser la distribution de la pulvérisation entre la végétation, le sol et l'atmosphère et minimiser les pertes par dérive hors de la parcelle traitée ainsi que les risques d'exposition des riverains et des passants. La modélisation constitue à cet égard une approche pertinente pour étudier les processus majeurs et évaluer les modes de pulvérisation. L'objectif de ce travail de thèse a donc porté sur le développement d'un modèle original décrivant à la fois la distribution des pesticides entre l'air, le couvert végétal et le sol dans la parcelle traitée et la dispersion atmosphérique en aval lors de la pulvérisation pour une application en vignoble. Une étude bibliographique initiale a révélé que les outils de modélisation déjà disponibles étaient spécifiques de contextes de cultures donnés et/ ou d'un nombre de processus restreints, peu adaptés en vignoble.

Notre travail a donc eu pour objectifs i) de développer et d'évaluer un nouveau modèle mécaniste, ADDI-SprayDrift, et ii) d'illustrer son utilisation dans le cadre de l'évaluation de l'effet de différentes conditions de pulvérisation sur l'exposition des passants. La modélisation de la dispersion des gouttelettes de pesticides est décrite par une approche lagrangienne de type marche aléatoire. Un module simple de turbulence atmosphérique permet de représenter le champ de vitesse de vent dans et au-dessus de la végétation, dans différentes conditions de stabilité atmosphérique (neutre, stable et instable). L'approche choisie permet de représenter l'interception des particules par la végétation en fonction de la géométrie et densité de la canopée et l'évaporation de l'eau des gouttelettes durant le transport aérien. Prenant en compte les conditions d'émission (type de matériels), la concentration de la bouillie en matière active, le stade de développement de la végétation et les conditions météorologiques, ADDI-sprayDrift prédit la distribution des pesticides lors de l'application entre le sol, le couvert traité et l'air, les dépôts au sol en dehors de la parcelle traitée ainsi que la concentration de gouttelettes dans l'atmosphère à un temps, distance et hauteur donnée.

L'analyse de sensibilité du modèle a montré que le paramètre le plus influent sous toutes les conditions est l'angle d'éjection des particules et que l'influence de la vitesse d'éjection des gouttelettes, la proportion en matière active de la bouillie, la vitesse du vent et la hauteur du couvert est variable - entre forte à moyenne- selon la position du pulvérisateur par rapport à la bordure du champ et la sortie du modèle considérée.

La confrontation du modèle à des données de dérive sédimentaire et de distribution des pesticides lors de l'application en vignoble ainsi qu'à des données d'exposition de passants indique une bonne capacité prédictive du modèle. Le modèle a été appliqué à l'étude de l'influence du type de pulvérisateurs utilisés (pulvérisateur pneumatique, aéroconvecteur et face par face), de buses (standard ou antidérive), du stade de développement de la végétation, des conditions météorologiques et de la teneur de la matière active dans la bouillie, sur la distribution des pesticides lors de l'application et sur l'exposition des passants par inhalation et dépôt cutané. Les résultats confirment l'effet prépondérant des caractéristiques du dispositif de pulvérisation sur la distribution des pesticides lors de l'application, les pertes en dehors du champ et l'exposition des passants. Toutefois, du fait de la multiplicité des interactions existantes entre les facteurs testés et de la variabilité de leur effet, une hiérarchisation de la contribution relative de chacun des facteurs nécessiterait des tests complémentaires.

Ainsi, les travaux conduits au cours de cette thèse ont permis le développement d'un nouvel outil de modélisation mécaniste, simple et conceptuellement générique, pouvant, après adaptations nécessaires, être appliqué à d'autres contextes de pratiques phytosanitaires et pouvant être intégré dans des approches d'évaluation de l'exposition au sein d'un territoire.

**Mots clés :** *ADDI-SprayDrift*, dispersion et turbulence atmosphérique, modèle Lagrangien, dépôt, interception par le couvert, dérive, dépôt cutané, inhalation.

## Abstract

Controlling the contamination of different environmental compartments and the exposure of different living beings by pesticides requires a good understanding of the major processes involved. This is the case for optimising the distribution of sprays between vegetation, soil and the atmosphere and minimising losses by drift outside the treated area as well as the risks of exposure of local residents and bystanders. In this regard, modelling is a relevant approach to study the major processes and evaluate the spraying technics. The objective of this thesis work was therefore to develop an original model describing the distribution of the application between the air, plant cover and soil compartments during spraying and the downwind atmospheric dispersion for a vineyard application. An initial bibliographical survey revealed that the modelling tools already available were specific to given crop contexts and/or a limited number of processes and were not very well adapted to vineyard conditions.

The objectives of our work were therefore i) to develop and evaluate a new mechanistic model, ADDI-sprayDrift, and ii) to illustrate its use in assessing the influence of spraying conditions on bystander exposure. The approach chosen for modelling the dispersion of pesticide droplets is a Lagrangian random walk approach. A simple atmospheric turbulence module is used to represent the wind speed in and above the canopy, under neutral, stable and unstable atmospheric stability conditions. The chosen approach allows to represent the interception of particles by the canopy as a function of its geometry and density and the evaporation of water from droplets during air transport. Taking into account the emission conditions (type of spraying equipment), the concentration of active ingredient in the tank mixture, the canopy growth stage and the meteorological conditions, ADDI-sprayDrift predicts the distribution of pesticides during application between the soil, the treated canopy and the air, the deposition on the ground outside the treated plot as well as the concentration of droplets in the atmosphere at a given time, distance and height.

The sensitivity analysis of the model showed that the most influential parameter under all conditions is the particle ejection angle, while the influence of ejection velocity, active ingredient proportion, wind speed and canopy height is variable - from high to medium - depending on the position of the sprayer in relation to the field edge and the output considered.

Comparison of the model with sediment drift data in vineyards, pesticide distribution at application data and bystander exposure data indicates a good predictive capacity of the model. An application of the model was carried out to quantify the influence of the type of sprayers used (pneumatic, axial fan and side by side sprayers), nozzles (standard or anti-drift), canopy growth stage, meteorological conditions and content of the active ingredient in the tank mixture, on the distribution of pesticides during application and on the exposure of bystander by inhalation and dermal deposition. The latter has further emphasised the dominating effect of sprayer characteristics on pesticide distribution during application, off-field losses and bystander exposure. However, due to the multiplicity of interactions between the tested factors and the variability of their effect, establishing a hierarchy of the relative contribution of each factor to sedimentary spray drift and bystander exposure would require further testing.

Thus, the work carried out during this thesis has allowed the development of a new mechanistic modelling tool, simple and conceptually generic, which can, after the necessary adaptations, be applied to other contexts of phytosanitary practices and which can be integrated into approaches for assessing exposure within a territory.

**Key words:** *ADDI-SprayDrift*, dispersion and atmospheric turbulence, Lagrangian model, deposition, canopy interception, spray drift, dermal deposition, inhalation.

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## Liste des abréviations, acronymes et symboles

### Liste des abréviations et acronymes :

- AASQA : Associations Agréées de surveillance de la Qualité de l'Air
- SBL : Surface boundary layer
- BNV-D : Banque Nationale des Ventes – Distributeurs
- bw : Body weight
- CFD : Computational fluid dynamics
- DPF : discrete probability function
- EAJ : les produits homologués dans les jardins
- EF : efficiency factor
- EFSA : European Food Safety Authority
- $E_p$  : Ejection point
- LAD : leaf area density
- LD : Low-drift nozzles
- LDV : Laser Doppler Velocimetry
- LES : Large Eddy Simulation
- MEF : Maximum Entropy Formalism
- non EAJ : les produits non autorisés dans les jardins
- OAT : "one at a time"
- PDPA : Phase Doppler Particle Analyser
- PPP : Produits phytopharmaceutiques
- RANS : Reynolds-averaged Navier Stokes
- RMSE : root mean square error
- RRMSE : relative root mean square error
- $S$  : Standard nozzles
- SAU : surface agricole utilisée
- TGAP : taxe phytosanitaire

### Liste des symboles :

- $A$  % : Proportion of active matter within the tank mixture (%)
- $A_h, A_f$  : Horizontal and frontal leaf area densities (LAD), respectively
- $c$  : Constant
- $C_D$  : Dimensionless number representing the drag coefficient
- $Ch, h$  : Canopy(vine) height (m)

- $D_0$  : Initial droplet diameter ( $\mu\text{m}$ )
- $D$  : Droplet diameter (m)
- $\bar{D}$  : Logarithmic mean diameter of the droplets ( $\mu\text{m}$ )
- $dt$  : Time step (s)
- $E_a$  : Ejection angle ( $^\circ$ )
- $E_i$  : Impaction efficiency
- $E_v$  : Velocity ( $\text{m s}^{-1}$ )
- $g$  : Acceleration of the earth's gravity ( $\text{m s}^{-2}$ )
- $k$  : Turbulence kinetic energy
- $K_x, K_y, K_z$  : Horizontal, crosswind and vertical turbulent diffusivities, respectively ( $\text{m}^2 \text{s}^{-1}$ )
- $L$  : Monin–Obukhov length (m)
- $l$  : Vegetation element characteristic size (m)
- $L_a$  : Horizontal surface
- LAD : Leaf area density
- LAI : Leaf area index
- $L_w$  : Leaf width
- $L$  : Water latent heat of evaporation at  $20^\circ\text{C}$  ( $\text{kJ kg}^{-1}$ )
- $nf$  : Number of input parameters
- $R_e$  : Reynolds number
- $Re_p$  : Particle Reynolds number
- $Sh$  : Sherwood number (dimensionless)
- $S_{i_v}$  : Probability of deposition by inertial impaction on the vertical surface
- $S_p$  : Probability of pesticide droplet deposition into the canopy
- $S_s$  : Probability of sedimentation on the horizontal surface
- $St$  : Stokes number
- $t$  : Time (s)
- $T_s$  : Stokes time accounting for the inertia of the particle (s)
- $u, v, w$  : Horizontal, crosswind and vertical wind speeds, respectively ( $\text{m s}^{-1}$ )
- $U$  : Relative velocity of the droplet with respect to the surrounding air ( $\text{m s}^{-1}$ )
- $u$  : Wind velocity at a reference height ( $\text{m s}^{-1}$ )
- $u_*$  : Friction velocity ( $\text{m s}^{-1}$ )
- $u_{p_0}, v_{p_0}, w_{p_0}$  : Horizontal, lateral and vertical droplet initial velocities, respectively ( $\text{m s}^{-1}$ )
- $u_p, v_p, w_p$  : Horizontal, crosswind and vertical transient particle velocities following ejection, respectively ( $\text{m s}^{-1}$ )
- $\mu_{dp}$  : Mean diameter

- $v_e$  : Ejection velocity (m s<sup>-1</sup>)
- $v_s$  : Particle settling velocity (m s<sup>-1</sup>)
- $\kappa$  : Water thermal conductivity at 20°C (W m<sup>-1</sup> K<sup>-1</sup>)
- $(x_i, y_i, z_i)$  : Position of a particle at time  $t_i$
- $y_{\text{exp},i}$  : Ganzelmeier experimental values
- $\bar{y}_{\text{exp},i}$  : Mean of the Ganzelmeier experimental values
- $y_{\text{sim},i}$  : Simulated values
- $z_0$  : Roughness length (m)
- $z$  : Height above the ground (m)
- $\xi_{u_{i+1}}, \xi_{v_{i+1}}, \eta_{i+1}$  : Random numbers belonging to a Gaussian distribution with a mean equal to zero and a unitary standard deviation, respectively
- $\theta_d$  : Dry-bulb temperature (°C)
- $\theta_w$  : Wet-bulb temperature (°C)
- $\alpha_e$  : Ejection angle (°).
- $\rho_p$  : Particle density
- $\sigma_{LN}$  : Standard deviation of the droplet diameter ( $\mu\text{m}$ )
- $\sigma_{dp}$  : Standard deviation of droplet diameter
- $\sigma_u, \sigma_v, \sigma_w$  : Horizontal, crosswind and vertical standard deviations of the wind speed, respectively (m s<sup>-1</sup>)
- $\tau_e$  : Droplet evaporation time scale (s)
- $\gamma$  : Extinction coefficient
- $\varepsilon$  : Dissipation rate
- $\eta$  : Random number drawn from a uniform distribution between 0 and 1
- $\rho$  : Air density in kg m<sup>-3</sup>
- $\sigma_{\text{exp}}$  : Standard deviation of the experimental values
- $\Psi_M$  : Stability correction function
- $\Delta\theta$  : Wet-bulb temperature depression (°C)
- $\kappa$  : Von Karman constant
- $\lambda$  : Evaporation rate ( $\mu\text{m}^2 \text{ s}^{-1}$ )
- $\nu$  : Kinematic viscosity of the air (m<sup>2</sup> s<sup>-1</sup>)
- $\rho$  : Water density (g cm<sup>-3</sup>)
- $\tau$  : Turbulent Lagrangian time scale
- $V_{\text{cubic-inhalation}}$  : Cubic volume for inhalation exposure
- $V_{\text{cubic-dermal}}$  : Cubic volume for dermal exposure



## *Introduction générale*

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Dans le but de protéger les cultures de divers ravageurs pouvant nuire à leur productivité, des produits phytopharmaceutiques (PPP) sont épandus sous différentes formes sur les parcelles agricoles. Étant majoritairement issus d'une synthèse chimique, leur présence dans les milieux naturels et dans les tissus d'organismes vivants constitue une contamination chimique pouvant avoir des conséquences avérées sur ces derniers (Mamy et al., 2008). Lors de leur application, une partie de ces PPP n'atteint pas la cible, ce qui constitue non seulement une perte en termes d'efficacité de traitement mais aussi une contamination de la parcelle traitée et des autres milieux où ces PPP sont transférés. Ainsi, les mesures dans l'air, l'eau et le sol au niveau des zones non-ciblées par les traitements, révèlent une présence des pesticides épandus de nombreux jours voire des semaines après leur application (Aubertot et al., 2005, Leenhardt et al., 2022). Cette prise de conscience concernant les potentiels risques toxicologiques et écotoxicologiques en lien avec cette contamination a donné naissance à des questionnements concernant la durabilité des systèmes de cultures actuels que ce soit sur le plan économique, social ou environnemental (Tiryaki and Temur, 2010). Pour y répondre, il est nécessaire d'acquérir non seulement une vision globale du devenir des pesticides appliqués dans les différents compartiments environnementaux mais également de comprendre les processus impliqués dans cette contamination. Cela doit permettre d'évaluer l'étendue de la contamination et des risques qui lui sont liés, mais aussi la performance de certaines pratiques agricoles et l'identification de celles limitant les contaminations selon les conditions locales d'application (Charbonnier et al., 2015).

Le devenir des pesticides dans l'environnement est déterminé par de nombreux processus et facteurs agissant dans les différents compartiments de l'environnement (Özkara et al., 2016). Un facteur clef est la technique d'application des PPP dont l'efficacité en termes de fractions de produit pulvérisé qui atteint la cible est très variable selon la technique utilisée, la culture concernée et son stade de développement de la végétation (Queyrel, 2017). A cela s'ajoute l'effet des conditions météorologiques locales qui font que, pour la même culture et la même technique d'application, l'efficacité de traitement et les pertes peuvent-être significativement différentes (Queyrel, 2017). Lors d'une application par pulvérisation qui est la technique la plus courante (Aubertot et al., 2005), le produit épandu est distribué entre une fraction atteignant la cible (le sol ou la végétation quand cette dernière est ciblée), celle perdue au sol dans la parcelle si la cible est le couvert et celle dispersée dans l'atmosphère par dérive des gouttelettes sous l'action de leur vitesse d'éjection et du vent induisant ainsi une extension de la contamination vers les zones non-ciblées (rivières, lacs, parcelles non agricoles, atmosphère). La détermination de la distribution initiale des pesticides lors de l'application au sein de la parcelle traitée et hors parcelle via

les transferts atmosphériques est une étape primordiale pour la compréhension et l'évaluation du devenir des pesticides dans l'environnement. Elle l'est également au plan des impacts sanitaires directs de l'épandage de pesticides puisque la dérive hors parcelle des produits lors de l'épandage fait l'objet d'un questionnement social majeur sur la protection des riverains des zones agricoles (e.g. M. Valo, , Le Monde 5 mai 2020). En effet, des prélèvements sur des sites viticoles indiquent par exemple une contamination saisonnière de l'air avec jusqu'à 24 ng/m<sup>3</sup> en concentrations moyennes hebdomadaires cumulées toutes substances confondues en juin (ANSES, 2017). Une étude récente montre aussi une contamination de l'air ambiant près d'écoles situées à proximité de vignobles avec des concentrations instantanées pouvant atteindre plus de 20 ng/m<sup>3</sup> (Raheison et al., 2019). La question de la dimension de la zone de non traitement à proximité d'habitations est clairement posée (Guilpart et al., 2022; Yootest & Générations Futures, 2021). Les zones viticoles sont particulièrement concernées car le secteur viticole reste un gros consommateur de pesticides en France, avec un indice moyen de fréquence de traitement de 15,3 en 2016 à l'échelle nationale. Cette utilisation est bien supérieure à celle des grandes cultures et correspond en France à 20% de la valeur de la consommation totale de pesticides pour seulement 3% de la surface agricole (Aubertot et al., 2005).

Dans ce contexte, l'objectif général de ce travail de thèse est de développer un outil original de modélisation permettant de prédire la distribution d'un pesticide épandu par pulvérisation dans et en dehors de la parcelle traitée pour pouvoir évaluer l'efficacité du traitement pour différentes techniques de pulvérisation via l'estimation de l'interception par le couvert traité, les pertes engendrées au sol par dépôt dans la parcelle et en dehors de la parcelle par dérive de gouttelettes, ainsi que les conséquences de ces transferts en termes d'exposition des passants et riverains. Une application prioritaire envisagée pour le modèle est dans le cas viticole, en raison du questionnement particulière prégnant actuellement relatif aux phénomènes de contamination de riverains dans les zones de vignes.

Le premier chapitre consiste en une mise en contexte sur l'utilisation des pesticides, les processus impliqués dans le transfert des PPP dans l'environnement avec un focus sur le transfert atmosphérique à l'échelle locale des gouttelettes de pulvérisation et de l'exposition qui en découle, ce qui nous permet de définir la démarche de la thèse. Les 3 chapitres suivants rédigés sous forme d'article scientifiques, dont un soumis en cours de révision, portent sur les travaux obtenus en termes d'analyse critique des modèles de dispersion de pesticides existants, de présentation et évaluation du modèle ADDI-spray nouvellement développé et de l'application de ce modèle à l'évaluation de la contribution du processus de dérive à l'exposition de riverains en bordure d'une parcelle viticole.

## Chapitre 1

## Contexte et démarche de thèse

*Le présent chapitre expose le contexte général et, la démarche entreprise pour atteindre les objectifs visés. Ainsi, il sera d'abord présenté l'utilisation des pesticides en général et plus précisément en viticulture. Ensuite, les généralités sur la contamination des milieux et des êtres vivants ainsi que les facteurs définissant le devenir des pesticides dans l'environnement seront discutés avec un focus sur le transfert aérien. S'en suivra une présentation de la démarche entreprise.*





## I. Les enjeux concernant l'utilisation des pesticides

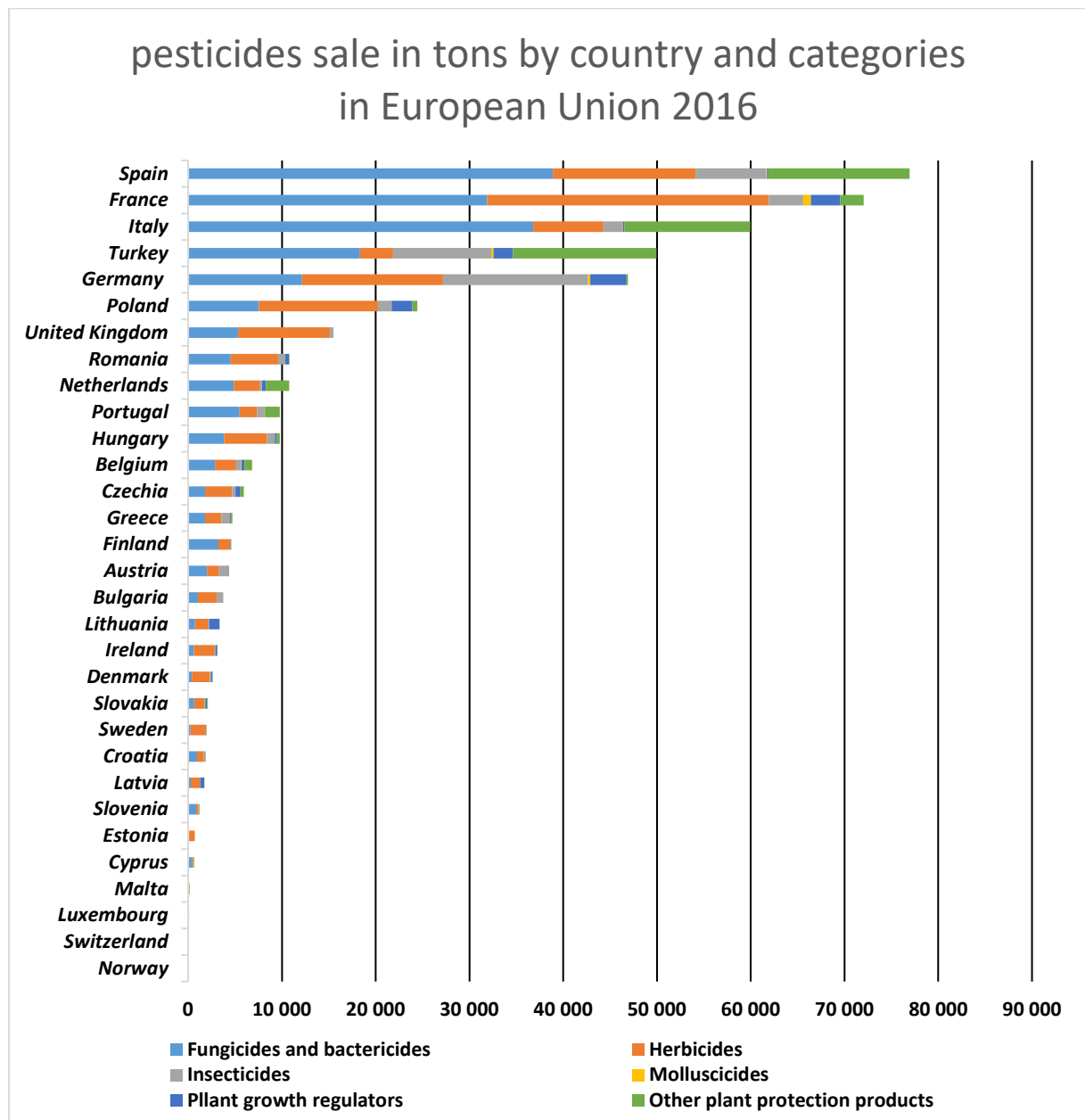
### I.1. Utilisation des pesticides en France

Dans la perspective de répondre à l'enjeu mondial de la sécurité alimentaire et avec l'apparition des premiers produits de synthèse, les systèmes de cultures intensifs permettant une plus grande productivité se sont répandus de par le monde en lien avec l'efficacité de ces produits pour la protection des cultures et leur facilité d'utilisation (Özkara et al., 2016).

En France et dans toute l'Europe, l'utilisation des PPP fluctue d'une année à une autre comme le montre l'analyse des chiffres de ventes des principales industries phytopharmaceutiques, analyse facilitée depuis la mise en place de la BNV-D 'Banque Nationale des Ventes – Distributeurs' créée par la loi n°2006-1772 du 30 décembre 2006 sur l'eau et les milieux aquatiques (un passage des données au code postal acheteur sont disponibles depuis 2015). Ces fluctuations peuvent être liées à plusieurs facteurs, allant de la variabilité de la pression parasitaire aux politiques agricoles en passant par l'évolution des produits de synthèse mis sur le marché (Lievens et al., 2012). Nous pouvons toutefois relever que ces données peuvent ne pas refléter complètement leur utilisation réelle pour l'année considérée, des variations pouvant être induites par le stockage ou le déstockage de produits. Une telle situation avait déjà été observée en 1990 où un pic de vente a été enregistré du fait d'un stockage en prévision de la taxe phytosanitaire (TGAP) appliquée à partir des années 2000 (Aubertot et al., 2005). De plus, ces chiffres intègrent l'utilisation non agricole de ces produits (par exemple par la SNCF), même si la BNV-D distingue les produits homologués dans les jardins (EAJ) de ceux non autorisés dans les jardins (non EAJ).

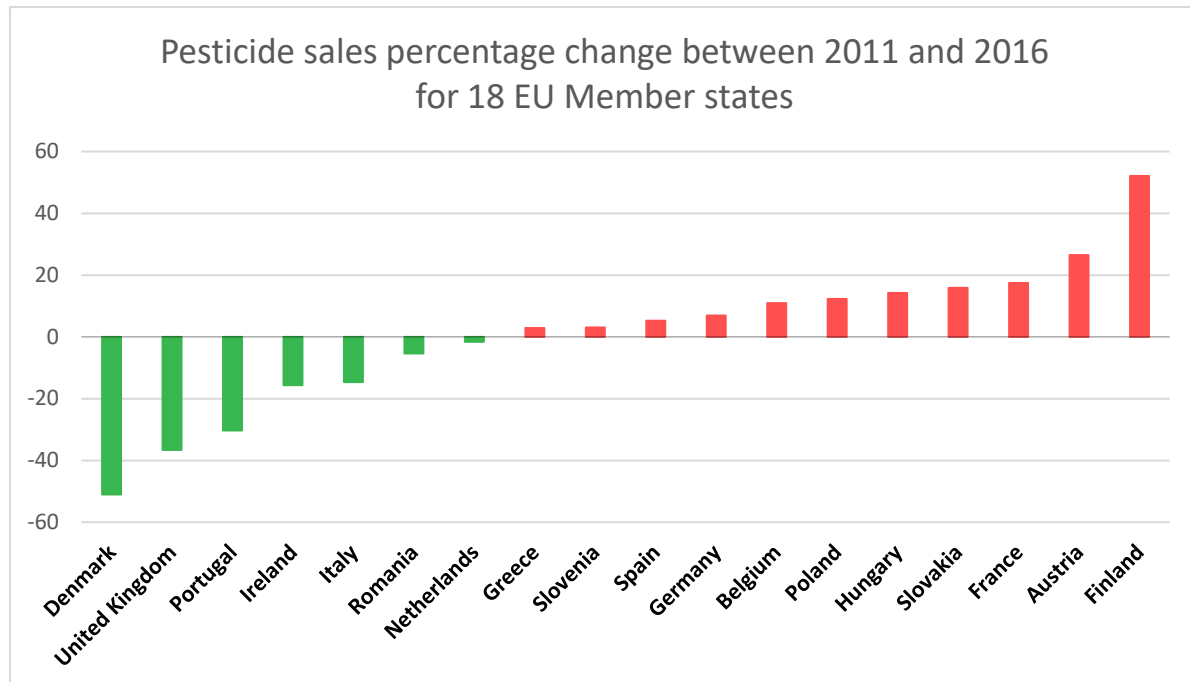
La France, avec une surface agricole utile parmi les plus importantes en Europe, fait partie des plus grands consommateurs de PPP au niveau européen: en 2016, elle est classée deuxième en Europe après l'Espagne avec 20 % des consommations de l'Europe des 18 grands pays agricoles consommateurs de pesticides (Fig. I.1). L'Espagne, la France, l'Italie et l'Allemagne à eux seuls représentent 70 % des utilisations. Parmi ces 18 pays, la France est le premier pays consommateur d'herbicides et de molluscicides et le troisième consommateur de fongicides après l'Espagne et l'Italie (Eurostat, 2016).

Figure I.1. Vente des pesticides en tonne par pays et catégories dans l'Union Européenne en 2016 (Données Eurostat, 2016)



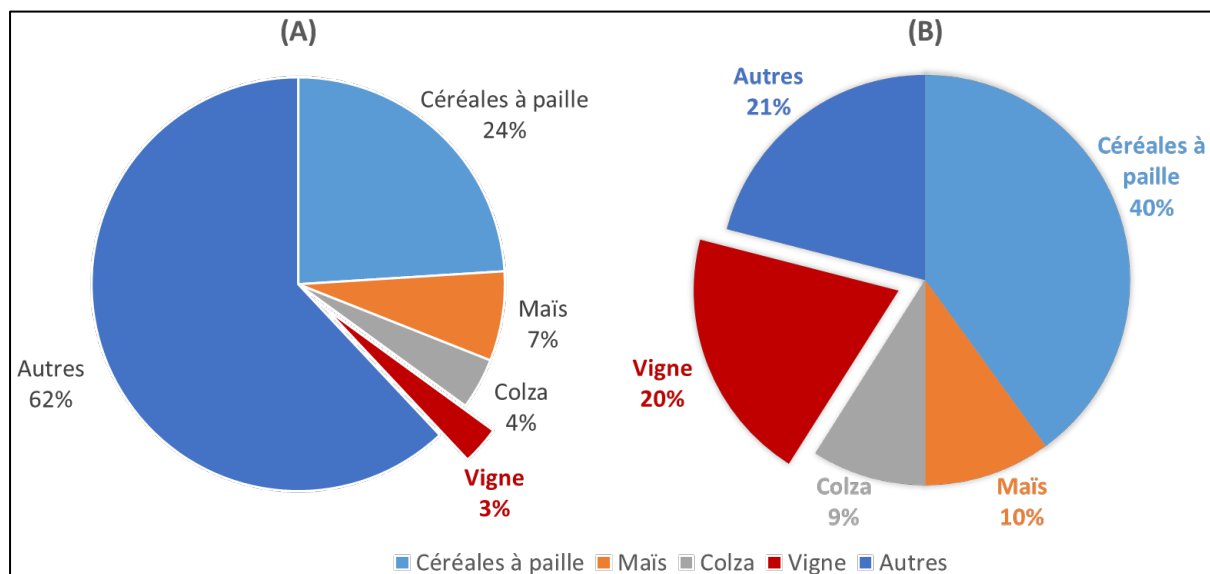
Toutefois, malgré les questionnements concernant la durabilité des systèmes de culture intensifs et de la prise de conscience concernant les impacts des pesticides sur la santé humaine, mais aussi sur les différents écosystèmes terrestres et aquatiques et les services écosystémiques qu'ils fournissent (Tilman et al., 2002), les quantités de pesticides utilisées en France ne sont pas réellement à la baisse, vu la dépendance systèmes de cultures actuels aux PPP. Entre 2011 et 2016, l'utilisation des pesticides en France a augmenté de 17 % (Eurostat, 2016) (Fig. I.2).

Figure I.2. Variation en pourcentage des ventes de pesticides entre 2011 et 2016 dans 18 États membres de l'UE (Données Eurostat, 2016)



En ce qui concerne l'utilisation des pesticides en termes de surface agricole utilisée (SAU), une grande hétérogénéité est notée selon les types de cultures. En France, près de 80 % des quantités sont utilisées seulement pour quatre types de cultures (céréales, maïs, colza et vigne) alors qu'elles occupent moins de 40 % de la surface agricole utile (Fig. I.3). Dans le cas des cultures pérennes, les pesticides sont fortement utilisés, alors qu'elles ne représentent qu'une faible surface par rapport à la surface agricole totale, comme c'est le cas de la vigne qui représente moins de 3 % de la SAU, mais qui consomme environ 20 % des pesticides utilisés en France (Aubertot et al., 2005; Cemagref, 2011; Inserm, 2013). Cette forte utilisation de pesticides en vigne, 30 kg/ha par rapport à la moyenne nationale qui est d'environ 5 kg/ha, constitue un risque sur l'environnement et implique une pression continue sur la viticulture pour la réduction de l'utilisation des PPP (Gaskin et al., 2002; Gil, 2007; Heidorn, 2002).

Figure I.3. Occupation du territoire et consommation de pesticides pour quelques cultures en France. Données 2000 source SCEES, UIPP. (A) représente le pourcentage de la Surface Agricole Utile (SAU) et (B) représente la consommation de produits phytosanitaires en pourcentage par rapport à la consommation total du pays.



## I.2. Contamination des milieux et impacts sur les organismes vivants

L'introduction des PPP de synthèse engendre une contamination de l'ensemble des compartiments écologiques (sol, eaux et atmosphère) des écosystèmes ciblés par le traitement, mais aussi ceux qui ne sont pas ciblés, cela résulte de la capacité des pesticides à se disperser au-delà de la zone d'épandage contribuant ainsi à l'expansion du périmètre de contamination (Aubertot et al., 2005).

Une vision globale de cette contamination et de son évolution spatio temporelle reste encore difficile à acquérir vu la variabilité spatiale et temporelle de son occurrence, du relativement faible nombre de données existantes selon le compartiment environnemental considéré (par exemple le sol) voire également de l'hétérogénéité des méthodes de quantification utilisées selon les campagnes, les années, etc. (Leenhardt et al., 2022). En fonction du compartiment étudié, de la proximité par rapport au lieu de traitement et de la nature du PPP utilisé, l'étendue spatiale de la contamination selon le moment de l'observation par rapport au moment où a eu lieu l'application est nettement variable. Lors de l'application et quelques jours après, les concentrations en PPP dans l'atmosphère à proximité de la parcelle traitée sont beaucoup plus significatives qu'à plus grande distance et après quelques semaines. Dans les autres compartiments aussi (eau et sol), la concentration des PPP diminue en fonction de la distance par rapport à la zone de traitement, mais par contre son évolution dans le temps et sa persistance dans la matrice n'est pas la même. À cela s'ajoutent les propriétés physico-chimiques du PPP utilisé qui favorisent la présence d'un pesticide dans un compartiment plus qu'un autre. Par exemple, les composés hydrophiles sont en général plus retrouvés dans l'eau, alors que les composés organiques les plus volatils sont plus retrouvés dans l'atmosphère (Leenhardt et al., 2022).

Dans les sols, une grande diversité de substances a été détectée dans différentes études. L'étude récente de Silva et al. (2019) sur des sols agricoles européens montrent que 83% des sols analysés contenaient

des résidus de PPP et 58 % de ces échantillons contenaient de multiples résidus de PPP avec une composition très variable donnant lieu au total à 166 combinaisons de PPP différents. Ces résultats sont en concordance avec les résultats d'analyse de l'étude de Pelosi et al. (2021) sur des sols Français, pour laquelle en moyenne 11 PPP ont été détectés dans des sols prélevés sur des parcelles de céréales d'hiver, contre 6 PPP en moyenne dans des sols de parcelles en agriculture biologique et entre 1 à 16 PPP dans des sols de prairies.

À travers le mouvement latéral (*e.g.* ruissellement) et vertical (*e.g.* infiltration) des PPP dans ou à la surface du sol, les eaux de surfaces et souterraines se retrouvent contaminées par divers types de PPP. En 2021, l'Agence environnementale Européenne a publié son rapport concernant la qualité des eaux de surfaces et souterraines en Europe (European Environment Agency, 2021). Ce rapport indique qu'entre 2013 et 2019, dans 13 à 30 % des sites surveillés, un à plusieurs pesticides ont été détectés au-delà de leur seuil d'effet sur les organismes vivants dans les eaux de surfaces contre 3 à 7 % des sites surveillés pour les eaux souterraines. En France en 2013, selon le rapport du commissariat général du développement durable sur les pesticides (Service de l'observation et des statistiques, 2015) dans les cours d'eau, au moins un pesticide est présent dans 92 % des points de surveillance avec dans la moitié des cas au moins 10 PPP différents. Les quelques exceptions restantes sont des points de surveillance en zone montagneuse où il y a peu d'agriculture. A noter toutefois que lors de campagnes de mesures spécifiques, une contamination par plusieurs pesticides a pu être démontrée dans des lacs de montagne (Machate et al., 2022), impliquant très certainement un transport atmosphérique.

En effet, sous l'effet du vent, les PPP se dispersent dans l'air sous forme de gouttelettes, d'aérosols ou de gaz. Selon leur forme (gouttelettes / gaz), leurs propriétés et la période d'application, leur persistance dans l'air et la distance à laquelle ils peuvent être transportés peuvent varier considérablement.

Compte tenu du rôle du compartiment atmosphérique dans la dissémination des polluants et de leur transfert vers des zones éloignées de la zone d'émission puis de leurs dépôts secs (*i.e.* gazeux et particulaires) et humides (*i.e.* associés aux pluies, *e.g.* Décuq et al. (2022)), la présence de PPP dans l'air constitue un potentiel risque d'exposition des écosystèmes non cibles et des populations, et ce, que ce soit dans les zones agricoles mais aussi urbaines, la présence dans l'atmosphère de différents types de PPP (herbicides, insecticides et fongicides) ayant été détectée dans les deux milieux (ANSES, 2020). Malgré les connaissances concernant la contamination de ce compartiment et l'importance de définir son étendue, la contamination de ce dernier reste moins bien documenté que les compartiments aquatiques notamment. D'anciennes mesures au Royaume-Uni avaient mis en évidence la présence d'un certain nombre d'insecticides dans l'air dans l'air tout au long de l'année (Tarrant & Tatton, 1968). Coscollà & Yusà (2016) identifient plusieurs études ayant montré la présence de près de 150 PPP différents dans l'air à travers le monde. Et Millet & Bedos (2016) en relèvent d'autres menées en régions polaires montrant la présence de PPP issus de retombés atmosphériques. En France, à travers la synthèse faite par Coscollà & Yusà (2016) sur les données de mesures des pesticides dans l'air, 6 PPP ont été

identifiés comme étant les plus fréquemment détectés dans l'air dont 2 fongicides (chlorothalonil et folpet) et 4 insecticides avec une concentration moyenne allant de 0.01 à 11 ng/m<sup>3</sup>. En France toujours, des données ont été acquises notamment grâce aux campagnes de mesures menées en région par les Associations Agréées de surveillance de la Qualité de l'Air (AASQA) depuis les années 2000 pour les premières en suivant des protocoles plus ou moins similaires. Les protocoles ont été harmonisés pour une campagne nationale menée en 2018/2019 sur 50 sites au niveau national (incluant les DOM, Hulin et al., 2020). Cette campagne a fourni une photo de la contamination de l'atmosphère (LCSQA, 2020) et des éléments pour une première interprétation des impacts sanitaires pour la population générale (Anses, 2020). Ces données ainsi que celles obtenues depuis 2000 sont bancarisées dans une base de données Phytatmo<sup>1</sup> disponible pour le public, fournissant ainsi un jeu de données des plus avancé au niveau européen. Une surveillance pérenne de la contamination de l'air aux pesticides a par ailleurs été décidée en France. S'est ajoutée à la campagne 2018/2019 organisée par l'Anses et l'Ineris, une nouvelle campagne dédiée à l'évaluation de la potentielle surexposition des riverains aux PPP en région viticole dont les résultats sont en cours d'analyse (étude PestiRiv, co-organisée par SpF et l'Anses avec l'appui des AASQA).

Le niveau de la contamination de l'atmosphère varie considérablement selon le composé utilisé, la période d'application (conditions météorologiques), la quantité appliquée et la fréquence d'application et selon la distance par rapport à la zone d'application (Millet & Bedos, 2016). Ainsi, proche des zones d'application, on peut s'attendre à des concentrations atmosphériques plus élevées que lorsque l'on s'éloigne de ces zones, et ce, en lien avec la capacité de dilution de l'atmosphère, les processus de dégradation ou de dépôts s'exprimant sur des temps de transferts plus longs.

### **I.3. Impacts sur les organismes vivants**

Étant destinés à éliminer des organismes vivants considérés néfastes aux plantes cultivées, les pesticides agissent sur leurs cibles en altérant leurs fonctions vitales ou reproductives, en perturbant la signalisation nerveuse ou hormonale, la respiration cellulaire, la division cellulaire ou la synthèse de protéines. Toutefois, il est connu que les organismes vivants des différents groupes taxonomiques partagent des mécanismes physiologiques communs et les substances utilisées ne sont pas totalement spécifiques à un bio-agresseur. Ainsi, les PPP présentent un risque potentiel sur tous les organismes vivants présents dans le compartiment contaminé qu'ils soient ciblés ou pas par le traitement (Inserm, 2013, 2021, Leenhardt et al., 2022).

Cette contamination a été la cause d'une dégradation de la qualité des écosystèmes et de la biodiversité. Plusieurs études ont mis en évidence l'effet des pesticides sur les organismes vivants. En Allemagne, une étude portant sur les insectes volants au niveau d'une zone protégée, a démontré un déclin dramatique de près de 76 % de leur biomasse en 27 ans, or les insectes jouent un rôle essentiel dans une

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<sup>1</sup> <https://www.atmo-france.org/article/phytatmo>

multitude de processus tel que la pollinisation, la restitution de la matière organique ainsi dans la chaîne trophique. Cette altération de la biodiversité affecte le fonctionnement des écosystèmes, mais aussi leur résilience (Hallmann et al., 2017).

De toute évidence, l'être humain est lui aussi concerné par ces effets toxicologiques. Plusieurs études ont démontré que l'exposition prolongée et chronique aux pesticides est susceptible d'entraîner des effets sur la santé, que ce soit sur les professionnels (Inserm, 2013) ou sur la population générale (Inserm, 2021). Parmi les pathologies recensées, il y a les maladies et troubles neurologiques et respiratoires (Simões et al., 2022), les altérations de la croissance, les problèmes dermatologiques et de fertilité et différentes formes de cancers (Fuentes et al., 2021).

En France comme dans la majorité des pays Européens, les terres agricoles occupent une grande partie du territoire (en France la SAU occupe 51.5 % de la superficie du pays contre 9 % de territoire artificialisé et 34.5 % de zones naturelles, forestières ou humides), ce qui fait que ces zones sont susceptibles de se retrouver à proximité directe des habitations. Certaines études épidémiologiques suggèrent l'existence d'une relation entre cette proximité des zones agricoles et des problèmes de santé suite à une exposition ponctuelle ou chronique à court et long terme des riverains et des passants (Dereumeaux et al., 2020; Insee, 2018).

## **II. Devenir des pesticides dans l'environnement et facteurs impliqués**

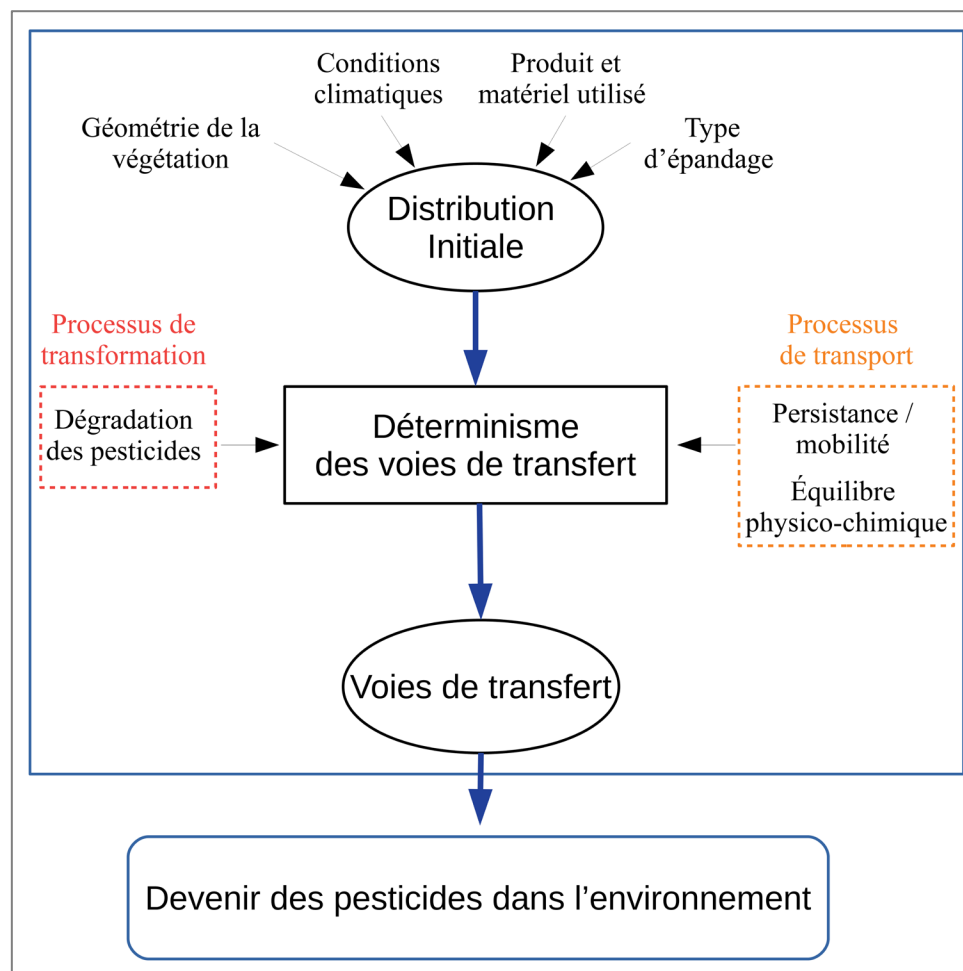
Le devenir des pesticides dans l'environnement est déterminé par de nombreux processus complexes agissant dans les différents compartiments de l'environnement. Une bonne compréhension de ces processus est la clé pour la détermination et l'évaluation des potentiels risques que constituent l'utilisation de ces produits sur les écosystèmes, les êtres vivants et la santé humaine. Elle permet aussi d'identifier les leviers d'amélioration pour la réduction de ces impacts (Özkara et al., 2016).

Pour une bonne compréhension des voies de transferts des pesticides et par la suite de leur devenir dans l'environnement, il est essentiel de connaître leur déterminisme ainsi que la dimension spatiale et temporelle qu'ils prennent. La détermination du devenir des pesticides dans l'environnement s'initie à l'échelle de la parcelle lors de l'épandage, puisque les pratiques liées à cette action ainsi que les conditions climatiques et la géométrie de la végétation définissent la distribution initiale des pesticides lors de l'application entre les différents compartiments environnementaux (Aubertot et al., 2005). Par la suite, le produit épandu prend divers chemins et formes selon la combinaison de différents facteurs qui représentent à chaque fois des conditions de terrain spécifiques, d'où la difficulté de représenter et de reproduire ces dernières. Cette combinaison de facteurs génère des processus de transport et de transformation des PPP qui déterminent leurs voies de transfert, mais aussi leur ampleur (Gavrilescu, 2005). Ainsi, des heures, des jours et voire même des mois après le traitement phytosanitaire, le produit épandu est entraîné en dehors de la parcelle traitée, causant ainsi l'expansion de la contamination depuis



l'échelle de la parcelle à celle du bassin-versant et du paysage, voire plus (cf. la contamination des régions polaires). C'est l'analyse de ces différents processus agissant à différentes échelles spatiales et temporelles qui permet de définir le devenir des pesticides dans l'environnement (Fig.I.4).

Figure I.4. Facteurs intervenant dans le devenir des pesticides dans l'environnement



## II.1. La distribution des pesticides lors de leur application

Lors de l'application des pesticides, la proportion du produit atteignant le sol, la végétation et l'air est très variable d'une application à une autre, et ce en lien avec divers facteurs dont les principaux sont : le type d'épandage, les conditions climatiques, la composition de la bouillie, le matériel utilisé et la végétation traitée (type, hauteur, LAI, ...). L'interaction et la combinaison de ces facteurs déterminent dans un premier temps la distribution du produit épandu lors de l'application entre les différents compartiments de l'environnement (sol/air) et dans et en dehors de la parcelle traitée et ce, selon la distance à la parcelle traitée (Queyrel, 2017).

- **Le type d'épandage :**

Le type d'épandage choisi définit la façon avec laquelle le produit utilisé va être appliqué, mais aussi l'efficacité de l'application et sa capacité à minimiser les pertes (PISC, 2002; Gil, 2007). Il existe quatre

types d'épandage à considérer : la fumigation, la pulvérisation, les granulés et micro-granulés et les semences enrobées.

La fumigation est utilisée pour stériliser le sol par incorporation, irrigation ou injection, la répartition initiale des pesticides suite à ce type d'épandage est fortement liée aux caractéristiques du sol et des composés. Cette pratique peut provoquer de fortes pertes dans l'air allant de 20 à 30 % et en cas d'épisodes pluvieux suivant l'application, les risques pour la contamination de l'eau sont très élevés.

L'utilisation des granulés, micro-granulés ou de semences enrobées sur ou dans le sol, peut causer l'émission de particules vers l'atmosphère notamment sous forme de poussières (Aubertot et al., 2005). Enfin, la pulvérisation est une application en phase liquide des pesticides par fragmentation en nuage de gouttelettes devant atteindre la cible (Gil, 2007). Lors de la pulvérisation sur le feuillage, une partie du produit est perdue vers le sol (10 à 70 %) et 30 à 50 % est émise vers l'atmosphère sous forme de gouttelettes ou de gaz (Van Den Berg et al., 1999 ; Aubertot et al., 2005) sous l'action du vent ou de la volatilisation depuis les gouttelettes – processus encore mal connu – ou bien depuis la fraction déposée au sol ou sur la végétation qui peut se volatiliser en post-application (Gil & Sinfort, 2005 ; Bahrouni, 2010 ; Queyrel, 2017). Cette technique permet une meilleure maîtrise de l'épandage du fait qu'elle soit plus facile à manipuler et d'un meilleur contrôle du dosage comparé aux autres méthodes (Gil, 2007). C'est pour cette raison que dans 95 % des cas, l'application en phase liquide est utilisée (Aubertot et al., 2005). De ce fait, dans la suite de cette introduction et du manuscrit, nous allons discuter exclusivement des processus liés à une application par pulvérisation.

- **Le produit utilisé :**

La distribution initiale des pesticides entre les différents compartiments de l'environnement dépend aussi de la solution apportée (la bouillie), qui comprend la matière active sélectionnée dans son produit commercial (mélange de la matière active avec des co-formulants et solvants), produit dilué dans un volume conséquent, le plus souvent d'eau.

Durant cette phase initiale, la proportion de matière active dans la bouillie influence significativement la taille des gouttes une fois l'eau évaporée depuis les gouttelettes. Cette variation en diamètre de gouttelettes influence leur transport, dépôt et donc leur répartition initiale. La composition de la bouillie peut elle aussi influencer la taille des gouttelettes. L'ajout de certains surfactants, permettant un meilleur recouvrement sur la cible, peut amener à la génération de plus de gouttes fines contrairement à certaines émulsions qui elles génèrent des gouttes d'un plus grand diamètre (Aubertot et al., 2005).

- **Les conditions climatiques :**

Lors de l'épandage, le facteur climatique principal pouvant influencer la distribution initiale des pesticides est le vent. Selon la direction et la vitesse du vent, le nuage de gouttelettes généré lors de la pulvérisation aura une direction et une forme différente. La trajectoire des gouttelettes varie donc selon ces deux paramètres qui peuvent induire une dispersion du nuage de gouttelettes jusqu'à des zones non ciblées à la base. Toutefois, la température et l'humidité relative de l'air influencent l'évaporation de

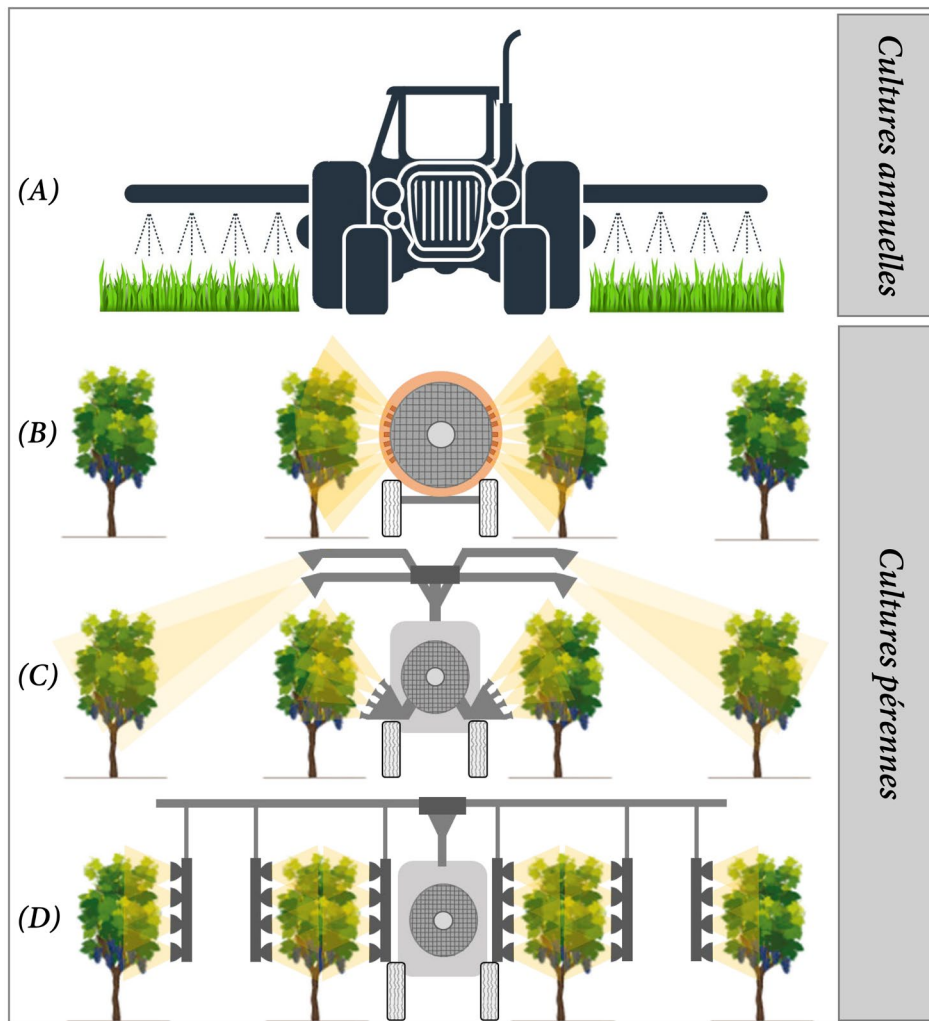
l'eau des gouttelettes, et donc leur diamètre qui à son tour impact leur dispersion. A noter que selon la réglementation en vigueur, les pulvérisations sont interdites à partir d'une vitesse de vent de 3 Beaufort correspondant à une vitesse au-delà de  $19 \text{ km h}^{-1}$  soit  $5,27 \text{ m s}^{-1}$  mesurée à une hauteur de 10 m.

- **Le matériel utilisé et l'architecture de la plante :**

Selon les objectifs du traitement et le type de culture traitée, les appareils de pulvérisation ainsi que leur technique de fragmentation de la phase liquide de la bouillie en nuage de gouttelettes varient considérablement (Gil, 2007). Ainsi, en fonction du type de ravageurs visés et de la structure du couvert, de son stade de développement et de sa densité, l'appareil utilisé est différent. Dans le cas des couverts à structure bidimensionnelle comme les cultures annuelles (*e.g.* blé), les pulvérisateurs à rampe sont les plus utilisés. Le jet de gouttelettes à la sortie des buses est vertical et dirigé vers le sol. Pour les cultures pérennes (*e.g.* vigne) qui ont une structure tridimensionnelle complexe, les pulvérisateurs les plus souvent utilisés sont le pulvérisateur pneumatique, l'aéroconvecteur et le face par face. Pour ces appareils, le jet est orienté à différents angles vers le couvert quand ce dernier est ciblé (Fig. I.5).

Pour minimiser les pertes et optimiser l'application, selon le développement du couvert, les points d'éjections sont adaptés sur chaque pulvérisateur afin de pulvériser essentiellement la végétation. Dans certains cas, le plus souvent en cultures pérennes, une assistance d'air est utilisée afin d'améliorer la pénétration du produit dans le couvert. Malgré l'efficacité avérée de cette méthode pour l'amélioration de l'application, elle augmente toutefois les risques de pertes par dérive (Cross et al., 2001, 2003; Gil, 2007).

Figure I.5. Les principaux types de pulvérisateurs utilisés en grande culture et en viticulture. (A) pulvérisateur à rampe, (B) l'aéroconvecteur, (C) le pneumatique et (D) le pulvérisateur face par face.



De cette diversité d'appareils résulte une variation dans les propriétés de la population de gouttelettes générées en termes de diamètre, de vitesse initiale d'éjection et d'orientation (angles d'éjection). Ces caractéristiques initiales du nuage de gouttelettes sont essentielles dans la définition de la distribution initiale des PPP au sein de la parcelle traitée.

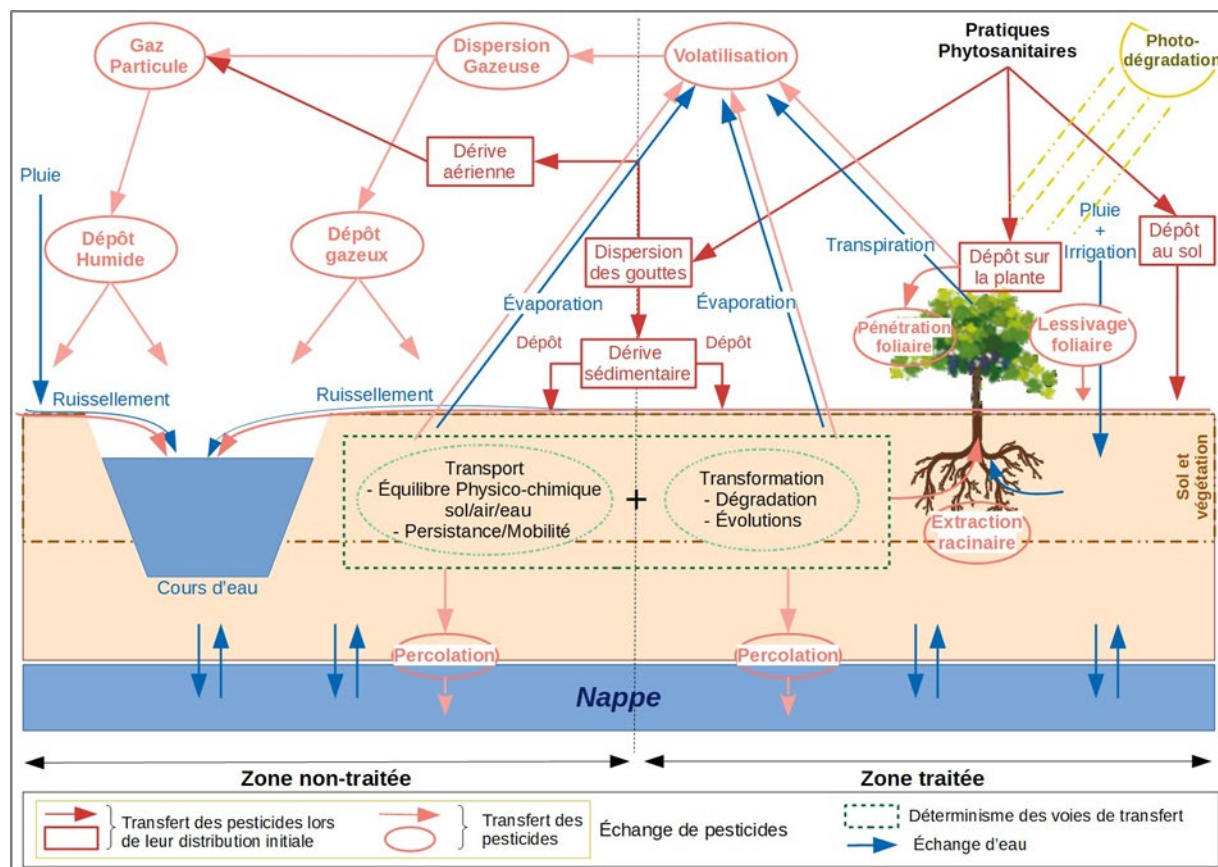
Dans la parcelle, la quantité de PPP interceptée par le feuillage dépend également de la capacité de filtrage du couvert. En fonction de la densité foliaire du couvert, de sa structure et de la taille du feuillage, les particules de taille différentes sont interceptées différemment par le couvert (Aubertot et al., 2005; Dupont et al., 2006).

## II.2. Les voies de transfert des pesticides en post-application et leur déterminisme

Le transfert des PPP dans l'environnement se fait durant mais aussi après l'application (Gil, 2007). Durant l'application, la fraction du produit appliqué n'atteignant pas la cible se dépose au sol dans la parcelle traitée ou bien est transportée dans l'atmosphère vers des zones non ciblées par la dérive de

gouttelettes de PPP par le vent. Le devenir des différentes fractions du produit appliqué dans l'environnement et les voies de transfert qu'elles entreprennent ensuite sont gouvernés par un ensemble de mécanismes en interaction qui sont les processus de transport et les processus de transformation (Queyrel, 2017) (Fig. I.6.).

Figure I.6. Les voies de transfert des PPP dans l'environnement durant et après l'application ainsi que les principaux processus impliqués.



Brièvement, les processus de transport sont ceux qui contrôlent le mouvement des PPP dans et entre les différents compartiments environnementaux. Ces processus dépendent des propriétés physico-chimiques des composés qui définissent la persistance et la mobilité des PPP et donc leur répartition entre la phase solide et liquide (eau et le sol) et la phase solide/liquide et gazeuse (eau, sol, air et végétation). Ainsi, les PPP les plus solubles tendent à suivre les transferts d'eau dans le sol voire percoler vers la nappe, les plus volatils peuvent se transférer vers l'atmosphère sous une forme gazeuse par volatilisation et ceux ayant une forte capacité d'adsorption au sol sont moins mobiles et restent plus au moins longtemps dans le sol (Tiryaki and Temur, 2010). Les composés peuvent également *ruisseler* en surface selon la pente du terrain, la perméabilité du sol et l'intensité du phénomène pluvieux (Aubertot et al., 2005). Les PPP peuvent être aussi transférés vers les eaux superficielles par les dépôts secs atmosphériques gazeux et particulaires, mais aussi par le dépôt humide des PPP suite à une pluie. A noter qu'une partie du composé intercepté par le couvert peut être transférée vers le sol via le lessivage foliaire à la suite à un épisode pluvieux ou bien une irrigation ainsi que via les résidus de récoltes. Par

ailleurs, selon ses propriétés physico-chimiques, sa mobilité et sa persistance, le PPP dans le sol est susceptible d'être **absorbé par la plante** via l'extraction racinaire. Les différentes voies de transfert ainsi que les processus qui leur sont liés sont schématisés et résumés dans la Fig. I.6.

Les processus de transformation sont les processus induisant un changement dans la structure et les propriétés du PPP épandu. Ces processus peuvent être biologiques, chimiques ou physiques et peuvent se produire dans tous les compartiments environnementaux. Cette transformation donne naissance à des produits de transformation ayant une structure et des propriétés différentes de la molécule mère, ce qui modifie le comportement des pesticides à court et long terme (Gavrilescu, 2005). La dangerosité des produits de transformation qui en résulte peut elle aussi être différente de celle de la molécule mère avec soit une augmentation ou une réduction de la toxicité pour les organismes vivants présents dans le compartiment impacté (Calvet et al., 2005).

À leur tour, les processus ayant lieu dans le sol sont influencés par certains facteurs pédoclimatiques à savoir : la température et l'humidité relative du sol et de l'air, la texture, la perméabilité, la profondeur, le pH et la teneur en matière organique du sol, mais aussi la pente du terrain (Vryzas, 2018).

La texture et la perméabilité du sol influencent le mouvement de l'eau et des PPP dans le sol, alors que la profondeur du sol définit la durée et la quantité de contact entre le sol et le PPP épandu. Le pH du sol agit sur la solubilité des PPP alors que la teneur en matière organique influence le potentiel de sorption du sol (Meftaul et al., 2020). Avec la température et l'humidité relative du sol et de l'air, ces derniers facteurs agissent sur l'activité biologique du sol et donc sur la biodégradation du produit appliqué. Pour finir, en fonction de la pente du terrain, l'infiltration ou bien le ruissellement est favorisé, ce qui induit l'implication de différents processus de transfert et de transformation (Gavrilescu, 2005).

Ainsi qu'évoqué précédemment, un transfert vers l'atmosphère peut avoir lieu par volatilisation depuis la surface traitée (sol/couvert) ou depuis le sol pour la fraction n'ayant pas été interceptée par le couvert quand ce dernier est la cible. L'ampleur et la durée de cette **volatilisation** dépendent des propriétés physico-chimique du produit épandu, des conditions pédoclimatiques ainsi que de la nature de la surface de dépôt (le sol ou la végétation) (Lichiheb, 2015). Ainsi, la volatilisation en post-application au niveau des cultures ou des sols pourrait représenter jusqu'à 60% de l'émission dans l'air et elle peut durer quelques jours voir plusieurs semaines (Bedos et al., 2002; Leenhardt et al., 2022). Une volatilisation de la matière active peut également avoir lieu durant la dispersion des gouttelettes avant leur dépôt, toutefois ce phénomène est moins caractérisé en vue des difficultés de mesures. A noter que l'intensité de la volatilisation dépend de la quantité de composé disponible aux transferts, disponibilité qui peut être atténuée selon par exemple l'adsorption du composé sur le sol ou la pénétration foliaire. À cela s'ajoute le transfert des PPP adsorbés aux fines particules du sol vers l'atmosphère par le vent (érosion éolienne). Ces fines particules de sol et de PPP restent dans l'atmosphère pendant une courte période a priori et finissent par se déposer plus loin. Il semblerait que ce transfert par érosion éolienne reste mineur par rapport à la volatilisation et à la dérive de gouttelettes. Sa contribution au transfert

vers les autres compartiments est estimée entre 1 et 5 % de la dose appliquée (Tiryaki and Temur, 2010) – il faut noter toutefois que pour certains composés, cela peut être une des explications de leur présence dans l’atmosphère – contre quelques dizaines de pourcent pour la volatilisation (Aubertot et al., 2005; Lichiheb et al., 2014) et jusqu’à 40 – 50 % pour la dérive des gouttelettes (Gil & Sinfort, 2005; Guiral et al., 2016).

Ainsi, la voie de transfert atmosphérique des PPP est l’une des principales voies de contamination des différents compartiments de l’environnement puisqu’elle intervient à différentes étapes du transport des PPP vers les autres compartiments, depuis la distribution des PPP lors de l’application par le biais de la dispersion des gouttelettes de PPP par dérive ou en post-application par dispersion atmosphérique de la fraction du composé volatilisée depuis la surface traitée, le sol ou la végétation (Lichiheb et al., 2014).

### III. Exposition environnementale par voie atmosphérique

Dans cette section nous abordant uniquement l’exposition environnementale des populations avec un focus sur la voie atmosphérique, nous ne présenterons donc pas l’exposition par voie alimentaire.

Les travailleurs, les opérateurs, les résidents et les passants sont exposés aux PPP par voie atmosphérique pendant l’application et juste après lors de la réintroduction dans le champ traité ou lors du contact avec des surfaces contaminées (Kuster et al., 2021; Mercier, 2020). Quatre voies d’exposition sont possibles et ces dernières sont directement ou indirectement liées au transfert des PPP par voie atmosphérique (Kuster et al., 2021). Durant l’application, deux voies d’exposition directes sont possibles, à savoir le dépôt dermique et l’inhalation de gouttelettes de PPP ou l’inhalation de gaz volatilisés depuis les gouttelettes durant leur transport. Après l’application, une exposition directe peut avoir lieu par inhalation des gaz issus de la volatilisation depuis les surfaces traitées mais également indirecte via le contact dermique avec les surfaces contaminées lors de l’entrée dans la parcelle traitée ou bien par contact dermique et oral avec les surfaces contaminées en dehors de la parcelle traitée (EFSA, 2022; Kuster et al., 2021).

Étant donné que les opérateurs et les ouvriers travaillent sur le terrain pendant et après l’application, respectivement, ils sont dans l’obligation de porter des équipements de protection adaptés pendant le traitement et lors du contact avec le couvert traité après application (EFSA, 2022).

Cependant, les passants localisés dans ou près de la parcelle traitée et les individus vivant ou présents dans quelconque type d’installations localisées à proximité du champ traité (résidents) et dont la présence est accidentelle et non liée à l’application des PPP, peuvent être respectivement exposés durant une courte période, mais d’une manière plus aiguë ou bien plus longtemps d’une manière plus diffuse respectivement (Kuster et al., 2021; Mercier, 2020). L’exposition de ces deux catégories n’a été abordée que récemment, notamment celle des riverains du fait qu’ils sont exposés sans porter ou avoir nécessairement les protections adaptées pour prévenir cette exposition ni de solution adaptable à leur échelle (Figueiredo et al., 2022). On peut citer l’exemple de deux projets qui ont vu le jour récemment



1) En Hollande, le projet OBO (“Research on exposure of residents to pesticides”) avec pour objectif de fournir des réponses concernant la concentration à laquelle sont exposés les résidents habitant à proximité des champs de cultures de fleurs à bulbes (<250 m), connues pour une forte utilisation de PPP, mais aussi concernant les sources et voies d'exposition contribuant à l'exposition environnementale et des individus aux PPP dans les zones de culture de fleurs à bulbes (Figueiredo et al., 2021) et 2) en Wallonie, où plusieurs campagnes de mesures ont été lancées dans le cadre du projet PROPULPPP (PROPULPPP, 2018) pour évaluer l'exposition des habitants vivant en bordure de champs (jusqu'à 100m) et l'efficacité des mesures mises en œuvre pour la réduction de cette exposition (buses anti-dérive et installation d'un écran de type filet de protection ‘anti-insectes’).

A noter que l'EFSA (the European Food Safety Authority) a établi un protocole spécifique pour l'évaluation de l'exposition en distinguant deux catégories d'âge (enfant et adulte) et en adaptant les caractéristiques de dépôt dermique et d'inhalation en fonction du poids, de la hauteur et de la surface du corps moyens et du taux d'inhalation selon l'effort pour chacune des deux catégories d'âge. Par la suite, leur exposition est basée respectivement sur le 75<sup>e</sup> et le 95<sup>e</sup> percentile pour les résidents et pour les passants (Mercier, 2020; EFSA, 2022).

## IV. Problématique et démarche de thèse

Une bonne quantification de la distribution initiale des PPP au moment de l'application au sein de la parcelle traitée est donc fondamentale pour étudier le devenir dans l'environnement des PPP. Des estimations à dire d'experts et à partir de données expérimentales de certains organismes de recherche ou bien d'instituts professionnels permettent de fournir des premiers ordres de grandeurs (Hulin et al., 2021) (Table S.B.1, Annexe B) mais les auteurs soulignent les limites en lien avec les simplifications qui ont dû être faites, et le manque de connaissance en termes de variation de cette distribution en fonction des diverses combinaisons de facteurs possible. Un outil de modélisation permettant de tester diverses situations est donc primordial pour mieux caractériser cette distribution, mais aussi pour évaluer la performance des appareils de pulvérisation et les pertes dans et à l'extérieur de la parcelle traitée dans différentes situations. Cet outil permettrait également l'évaluation de l'efficacité de certaines mesures de réduction des pertes et l'identification de certains leviers.

En termes de perte dans l'environnement, EFSA (2022) a encore souligné le manque de données relatives à l'exposition des riverains et des passants liée à la dérive des gouttelettes de pulvérisation de PPP et plus particulièrement en vigne. Le peu de données actuellement disponibles sont celles établis à partir des mesures ou de la modélisation faites majoritairement dans les vergers de pommiers (PestiRiv, 2021; EFSA, 2022; Mercier, 2020). Ainsi, un outil permettant l'estimation de la variation de cette exposition en fonction des différentes conditions de traitement en termes d'appareillage et de technique d'application, de stade de développement du couvert et des conditions météorologiques s'avère



également primordial pour comprendre le déterminisme de cette exposition et identifier les leviers de réduction tout en optimisant l'application (Mercier, 2020).

L'objectif de cette thèse est de développer un modèle mécaniste d'interception et de dérive des gouttelettes pour représenter la distribution initiale des PPP dans et en dehors de la parcelle. Le modèle développé doit permettre de représenter les principaux processus impliqués dans la dérive et l'interception par le couvert, être assez simple pour qu'il soit facilement intégré dans des modèles d'évaluation du devenir des PPP dans l'environnement à des échelles locales voire territoriales et être générique pour pouvoir être utilisé pour les différents types de matériel d'application et types de couvert. Il doit également permettre d'évaluer l'effet des différents paramètres sur la distribution initiale des PPP et l'exposition des riverains et des passants, la dérive des gouttelettes constituant l'une des principales voies d'exposition environnementale pour ces deux catégories d'individus (Figueiredo et al., 2022).

Nous avons choisi un premier contexte d'application en vigne, contexte pour lequel il y a une forte pression pour la réduction de l'utilisation des PPP.

Ces aspects seront présentés dans ce mémoire de thèse à travers les trois chapitres suivants, où tout d'abord une analyse bibliographique des modèles mécanistes les plus utilisés dans la communauté scientifique sera présentée dans le *chapitre 2*. Ce chapitre a permis d'identifier les possibilités de modélisation pour chaque processus clé dans la dérive des PPP et l'interception par le couvert ainsi que les avantages et inconvénients de chaque approche. Les conclusions de ce dernier ont permis le développement d'un modèle mécaniste de dérive et d'interception qui est décrit et évalué dans le *chapitre 3*. Pour finir, une application du modèle est présentée dans le *chapitre 4* où on évalue l'influence du matériel de pulvérisation, du stade de développement du couvert et des conditions météorologiques sur la dérive sédimentaire en dehors du champ et l'exposition des passants à différentes distances. Enfin, une conclusion générale sur les principaux résultats obtenus et les perspectives de ce travail de thèse est présentée.

**Chapitre 2**

***Synthèse bibliographique sur les principes de la dérive atmosphérique et les outils de modélisation existants***

*Ce chapitre présente les principaux processus impliqués dans la dérive des PPP et discute de la façon dont ces processus sont mis en œuvre dans une gamme de modèles qui couvre la plupart des situations en termes de cultures traitées et d'approches de modélisation. Huit des modèles les plus connus et utilisés dans la communauté scientifique sont revus, les forces et les faiblesses des approches de modélisation sont discutées et les points les plus pertinents pour la modélisation future sont mis en évidence. Ceci concerne les caractéristiques physiques de la gouttelette émise, mais aussi la description détaillée de la turbulence atmosphérique et le dépôt des gouttelettes dans le couvert et au sol. Les points les plus prépondérants à considérer pour la modélisation de la dérive selon le type de culture (pérenne ou annuelle) notamment en vigne, sont mis en avant. Ce chapitre est présenté sous la forme d'un article à soumettre.*



## ***Pesticide spray drift processes and current modelling tools: a survey***

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### **Abstract:**

During application, pesticides droplets are transported outside the treated field by airflow inducing a contamination of the atmosphere and non-targeted areas through sedimentary spray drift, both leading to exposure of living beings at a local scale. Due to the diversity of driving factors and processes, and their interactions, experimental studies and empirical models still need improvement for establishing a mass balance of sprayed pesticide at the time of application that covers the diversity of situations encountered in reality (e.g. different crop architecture, spraying equipment or atmospheric conditions). Process based models yet appear to be the most appropriate ones to address this issue. This paper spans the main processes and input information involved in spray drift and discusses how these processes are implemented in a range of models that covers most situations in terms of treated crops and modelling approaches. The strength and weaknesses of the modelling approaches are discussed and the most relevant points for future modelling are highlighted. These regard the physical characteristics of the emitted droplet (size and speed distribution), but also the detailed description of atmospheric turbulence including thermal stability and their effect on droplet transport, and the description of canopy structure effects on local atmospheric turbulence and droplet deposition. A major step forward would also be the development of a model able to represent most agricultural practices including crops, orchards and wine.

**Keywords:** Atmospheric dispersion, particle tracking, spray drift modelling, canopy flow, droplet interception, aerial spray drift, sedimentary spray drift.

## I. Introduction

Pesticides used in agriculture to protect crops dissipate within all environmental compartments including the atmosphere through different transfer pathways, which occur during application by the drift of spray droplets outside of the treated field (ISO 22866, 2005), and soon after the application by dispersion downwind of the treated field of the fraction of pesticide being volatilized from the treated surface (FOCUS, 2008; Van Den Berg et al., 1999).

Off target losses during application imply:

- (1) "**airborne drift**" defined as spray droplets carried by the wind outside the application area and still suspended in the air for a considered time interval, implying a contamination of the atmosphere locally and thus presenting a risk of exposure by inhalation of living beings (birds, insects, mammals, including humans *e.g.* residents). The gas resulting from the volatilization of the compound from the droplet could be included in this component of the drift (Figueiredo et al., 2021), although they are currently mostly not taken into account. In this study, this fraction will not be considered.
- (2) "**sedimentary drift**", defined as surface deposition of spray drops beyond the limits of the treated field, which leads to contamination of non-target ecosystems (Guicherit et al., 1999; FOCUS, 2008), in particular surface water, and which has given rise, as a risk management measure, to the establishment of vegetated strips along watercourses or ditches with a permanent presence of water. It may also concern the risk of exposure of local residents through contact or surrounding crops injuries (Bish et al., 2021).

Sedimentary drift has been studied for long and has been included in the authorization procedure of pesticides, for example to assess potential contamination of surface waters and identify, if necessary, mitigation solutions (FOCUS, 2008; Van De Zande et al., 2019). The impact of airborne drift being more difficult to assess and the exposure of residents and bystanders being only recently identified as a potential risk, the study of airborne drift is more recent (Jensen and Olesen, 2014). In addition, Jensen and Olesen (2014) also pointed out the need to establish a spray mass balance of pesticides during the application, including airborne, sedimentary drift, and deposition and interception by the canopy within the treated field.

Various experimental methods have been developed to assess off-target losses or the efficiency of mitigation measures to reduce them, from laboratory, wind tunnels (Butler Ellis et al., 2021; Nuytten et al., 2010, Alheidary et al., 2014) to field conditions (*e.g.* the synthesis of Van de Zande et al., 2019 for orchard conditions). However, the set-up of experiments is time, labor and cost consuming. Moreover, off-target losses result from a combination of processes that comprise the emission of spray droplets, their subsequent atmospheric dispersion, the droplets evaporation during their transport, and their

deposition to the ground as well as the interception by the canopy. As a consequence, different factors govern pesticide drift intensity: the used equipment (sprayers, nozzles, ...), the nature of the treated crops (annual or perennial crops), the properties of the spray liquid and the local weather conditions (Bonds & Leggett, 2015; Hong et al., 2021; Matthews et al., 2014). Because of the difficulty to gather datasets combining all factors, modelling is a useful complementary approach to quantify sedimentary and airborne drift.

Empirical relationships are the simplest modelling approach to estimate sedimentary drift. Several such models exist: Ganzelmeier et al. (1995) proposed sedimentary drift curves for various crops, based on measurements under various conditions. Van de Zande et al. (2014) established also probabilistic empirical model of bystander exposure by airborne and sedimentary drift based on field measurements in orchards. Holterman et al. (2017) developed the empirical SPEXUS model for sedimentary spray drift based on more than twenty years of measurements in apple orchards. Even if they have the advantage of a low computing time, limitations can however be stressed: their range of validity is restricted to the range of conditions of the experimental campaign. They have often been developed for distances lower than 30 m (Hong et al., 2021). Another limitation noted by Holterman et al. (2017)(SPEXUS) concerns the fact that considering average ambient conditions does not allow the reproduction of the experimental variability observed in relation to the variability of weather conditions, as well as a difficulty in reproducing the effect of temperature on deposition.

Process-based models overcome some limitations of empirical models which allow (1) studying complex interacting processes, (2) accounting for many factors, (3) representing a range of outputs - *e.g.* deposition on the ground, concentration in the air downwind of the treated plot, interception of the spray by the canopy within the treated plot, and (4) assessing the effectiveness of management practices in reducing drift.

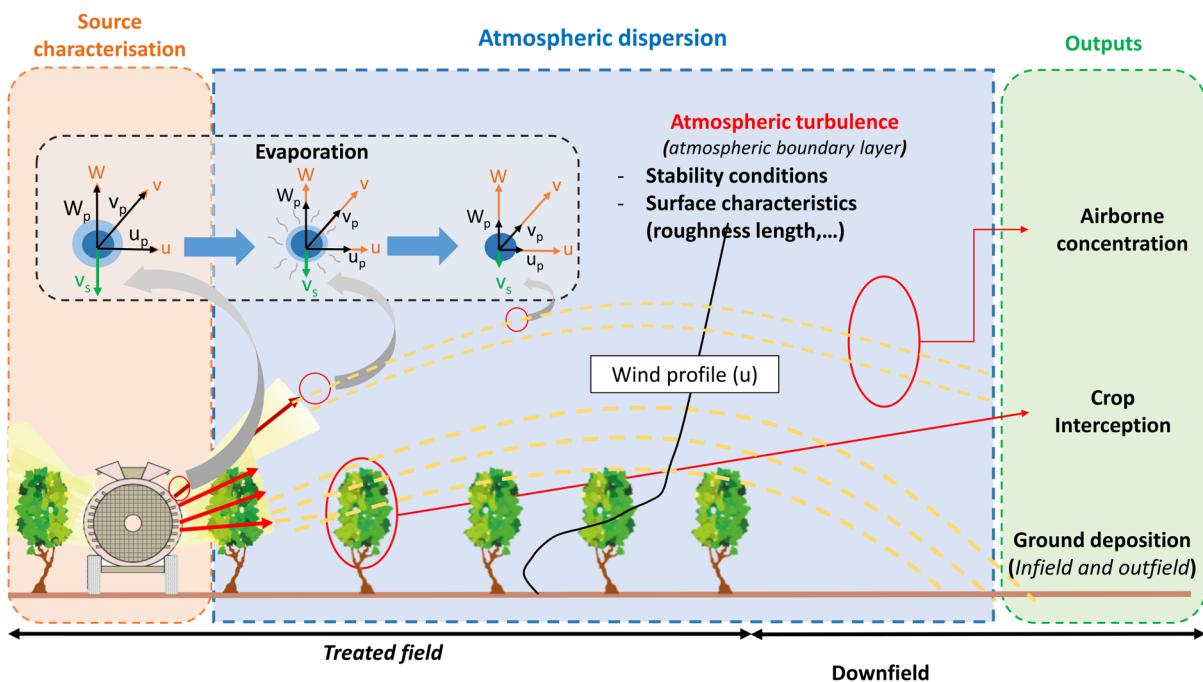
This paper review a selection of most-used process-based models of spray droplets dispersion in the atmosphere after pesticide application to identify their underlying assumptions, strengths and means of improvements. Each process is discussed one at a time: (1) spray droplet generation, (2) atmospheric dispersion including settling due to gravity, drag and inertia as well as evaporation, and (3) deposition on crops and at the ground. For each process, modelling approaches are first globally discussed and then detailed for each of the reviewed models. The reviewed models were chosen among the most used by the scientific community working on pesticide drift, in order to cover a range of situations in terms of treated crops (field crops, orchards, ...) and modelling approaches. As a consequence, this paper is not exhaustive in describing drift models but rather focuses on a selection of models: AGDISP (Nsibandé et al., 2015; Teske et al., 2002; Teske et al., 2009), ARPS (Chahine, 2011); CFD model used by Hong et al. (2018a,b), IDEFICS (Holterman et al., 1997; Vermeulen et al., 2019), OML-Spray Drift (Lofstrom

et al., 2013), RTDrift (Lebeau et al., 2011), Silsoe Spray drift model (Butler Ellis and Miller, 2010), and Thompson & Ley 1983 (Thompson and Ley, 1983). Finally, after a summary of the processes generally described in these models and those that would need improvement, an analysis of the application areas of the identified models and the areas for improvement is carried out.

## II. Pesticide droplet emission

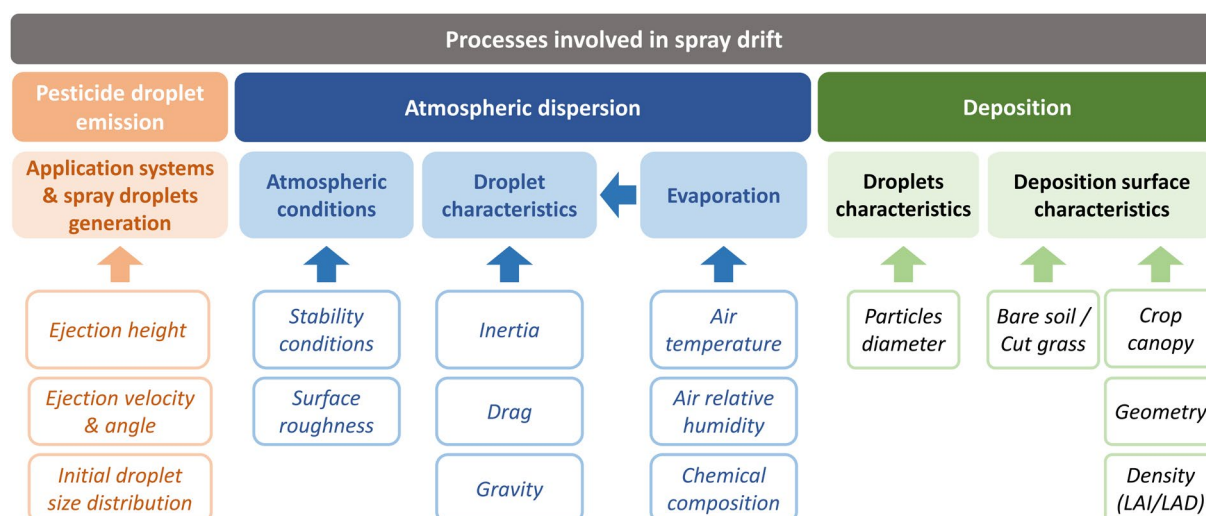
During pesticide application, the first deterministic driver of spray drift is the characteristics of the droplets released (Fig. II.1).

Figure II.1. Main processes involved in pesticide spray drift



Depending on the targeted pest or weed, the treatment period and the treated crop, the used application system, pesticide, machinery, nozzle type and settings are fundamentally different. Such diversity in application techniques leads to differences in size distribution of pesticide droplets and also in their speed, angle and height of ejection (Fig. II.2), which affect the proportion of pesticide lost outside the targeted area and the distance at which the droplets can be carried away (Gulyas et al., 2012).

Figure II. 2. Main processes to be considered for sedimentary and aerial spray drift modelling



In the following, the volume median diameter (VMD or  $Dv_{0.5}$ ) will be used to characterise the median size of droplets. It is defined as the diameter such that 50% of the total liquid mass is contained in drops of smaller diameter.

## II.1. Application systems

Machinery (design and application issues) are divided in two main categories depending on the treated crop: annual or perennial crops (orchards and viticulture) (Gil, 2007). In **annual crops**, the spraying device is oriented vertically toward the ground as the target is almost below the application system independently from plant growth stages and targeted pest type. Generally, boom sprayers are used for these treatments. These sprayers are known as *projected jet devices*, they are widely used especially for weeds control and are considered to be the simplest spraying equipment (Van De Zande et al., 2003). For this type of equipment, droplets are formed using pressurised liquid in the sprayer that passes through a calibrated orifice characterised by its diameter and shape. The pressure range from 0.5 to 15 bar according to spraying objectives and chosen nozzles. These device generate droplet with VMD ranging from 100 to 500  $\mu\text{m}$ , and ejection velocity ranging from about 3 m/s when spraying is performed without air assistance to about 30 m/s when an air assistance is applied (Nuyttens, 2007). This latter kind of sprayer may be used even for perennial crops in weed control context, using a boom sprayer with drop-pipes.

In **perennial crops**, the targets have a complex three-dimensional structure that expand above the spraying system. Therefore, generally, the used systems are air blast or pneumatic sprayers. These devices are equipped with an additional airflow to enhance application efficiency by reaching targets at farther distances and by increasing product penetration inside the canopy (Gil, 2007). In air blast sprayers, droplets are formed exactly like in projected jet sprayers with a spraying pressure ranging from



5 to 40 bar depending on the needed droplet diameter. In addition, droplets are transported by a strong airflow (axial or tangential) induced by turbines or fans. Typically, the generated droplet diameter varies from 75 and 300  $\mu\text{m}$  (VMD) depending on nozzle type and ejection pressure, with smaller nozzle sizes and higher pressure generally producing smaller droplets (Cerruto et al., 2021; Gil, 2007; Nuyttens et al., 2007). For these devices, it is possible also to switch off the air assistance when it is estimated of no use in order to reduce losses by spray drift (case of early development stage of the canopy). When performing spraying with the air assistance, droplet ejection velocity ranges from 15 to 60  $\text{m s}^{-1}$  (Duga et al., 2015). In pneumatic sprayers, droplets are formed and transported by the mean of a high-speed airflow from 60 to 150  $\text{m s}^{-1}$  obtained with Venturi-like pipes. The airflow rate at the outlet of the device is however probably smaller but not well documented. Unlike other sprayers, the liquid fragmentation into droplets results from the shear induced by the high-speed airflow in which the liquid is injected. Consequently, generated droplets have smaller diameters (between 50 and 150  $\mu\text{m}$  VMD), while their ejection velocity corresponds to the sprayer outlet airflow velocity. This type of sprayer are of high application performance in terms of product penetration into the canopy and its distribution over it, which is an expected feature for smaller droplets. On the other hand, losses by spray drift and evaporation are considered to be more important due to the smaller diameter of generated droplets and to the high-speed ejection velocity (De Luca, 2007).

## **II.2. Atomization process**

Even if the approximate droplet size range is known for sprayers as shown above – e.g. experimentally, it is possible to describe droplet size distribution and velocity using various optical technics like LDV (Laser Doppler Velocimetry) and PDPA (Phase Doppler Particle Analyser)-, it is important to describe and represent well the size distribution within the whole spraying cloud. For this, a good understanding of the droplet formation mechanism is first required and then size distribution representation within the spray cloud can be addressed.

Atomization is the process by which droplets are generated. It represents the transition from a continuous volume of liquid to a set of droplets characterised by a size distribution and a velocity. This process occurs in two steps: (1) the primary atomization consisting in liquid fragment formation on jet surface due to the shear generated by the difference in speed between the liquid and the air around, and (2) the secondary atomization consisting in an additional break up of formed fragments into droplets (De Luca, 2007). The atomization process is dependent on the air-liquid shear, which implies a dependency to the square of their relative velocities and viscosities, but also on surface tension, that plays a role in the film and droplets breakdown processes.

Hence, essentially, atomization is obtained by large differences between air and liquid velocities. This leads to two main atomization methods used in agricultural application: (1) atomization by pressure

using hydraulic nozzles, commonly flat fan, cone and deflector nozzles, where atomization occurs by passing pressurized pesticides spray liquid through nozzles orifice. In that case the liquid velocity is very high while the air velocity is very small, which creates the required shear; (2) atomization by air-induced shear, where the liquid is injected into a high-speed airflow usually obtained in a Venturi section. In this latter case, the air velocity is high and the liquid velocity is low. Since atomization is primarily dependent on liquid-air relative velocity, it explains why droplet size distributions and velocities is highly dependent on the method, nozzle and pressure used (Nuyttens, 2007).

Liquid properties, and especially viscosity and surface tension, but also density are also key factors. Hence, the composition of the solution in additives and products can influence droplet size distribution. A positive correlation was observed between droplet size and spray liquid density, surface tension and viscosity, as when these three parameters decrease, droplet diameters decrease (Nuyttens, 2007; Matthews et al., 2014). However, active ingredient and additives as well as time after mixing may modify these properties and hence droplet size (Matthews et al., 2014). Butler Ellis et al. (1997) showed that in addition to the effect on droplet spectrum, spray formulation may influence droplet initial velocity but these effects are variable as they are also dependent on nozzle design. Additives are surfactants, adjuvants, oils and other compounds that are added to the mixture in order to increase the efficiency of pest treatment or reduce the driftability. These additives may have an impact on droplet spectrum (Holterman et al., 1997; Butler Ellis et al., 1997). However, Zhu et al. (1997) and Nuyttens (2007) reported that additives has minor effects on droplet size compared to nozzle type, spraying pressure or airflow, hence suggesting that these parameters are of second order.

As for size distribution, droplet initial velocity depends on the atomization method, and thus on the nozzle type, the spraying pressure or the airflow speed. Ejection height and direction depend on targeted pest, crop structure and growth stage. Droplet size distribution and spraying velocity, ejection height, direction, and angle have to be adjusted properly in order to get an efficient treatment and to reduce losses by spray drift. This should then guide the choice of material to be adapted to these needs.

### **II.3. Pesticide droplet emission representation in spray drift models**

No model represents perfectly atomization mechanisms and the resulting size distribution and velocity based on the liquid pressure, the used nozzle or sprayed liquid properties (Lee et al., 2012). Four methods are used to describe droplet size distribution: the empirical method, the maximum entropy formalism (MEF), the discrete probability function (DPF) and the stochastic method.

The **empirical method** includes a number of classical mathematical distribution functions like normal (Gaussian), log-normal or hyperbolic distributions and other specific distributions developed by some authors like Rosin-Rammler (Rosin and Rammler, 1933) or Nukiyama and Tanasawa (1984)

distributions. These functions often rely on a single representative parameter like the *VMD* or *Dv 0.5*, the Sauter mean diameter, the volume mean diameter or the arithmetic mean diameter (Déchelette et al., 2011). In addition to this single parameter, a size range must be added as an extra parameter (Stevenin, 2012; De Luca, 2007). In general, these distributions give satisfactory results when compared to data. They are easy to use and allow extrapolation to several application situations. However, none of these empirical models can represent the effect of liquid properties, nozzles characteristics or pressure regimes, if not tabulated from a database. In spite of existing differences between these empirical functions, the statistical study of (Paloposki, 1994) shows that none of these methods is more appropriate than another for a general application. In practice, the model should be chosen as the one fitting the best a particular dataset. Paloposki (1994) further showed that even if Nukiyama and Tanasawa and hyperbolic distributions give the best fits in general, they showed some stability problems. Instead, the log-normal distribution performed less, but showed a better stability. Rosin-Rammler showed poorer fits (Déchelette, 2011; Lefebvre and McDonell, 2017).

The maximum entropy formalism (MEF), the discrete probability function (DPF) and the stochastic method are able to describe fully (Stochastic method), or partially, the atomization processes (primary atomisation for maximum entropy formalism and secondary atomisation for discrete probability function). However, these methods are complex and need more input for their calibration and use. Therefore, even if in the spray retention model of Dorr et al., (2005; 2008; 2015) an atomization model has been used to generate the mean droplet diameter to calculate droplets size distribution, atomization models still less used in pesticide spray drift models (Lefebvre and McDonell, 2017), and are thus not detailed in this paper.

In the pesticide spray drift models selected in this paper, few models use distributions derived from empirical functions such as Rans-Rammler distribution (Hong et al., 2018a,b) or simple Gaussian distribution (ARPS, (Chahine, 2011)). None of them used the other three methods because of their complexity. In the end, for most of them, authors define droplet size classes based on their own field measurements like in IDEFICS (Holterman et al., 1997), AgDRIFT (Teske et al., 2003), RTDrift (Lebeau et al., 2011), Silsoe spray Drift Model (Butler Ellis and Miller, 2010) and OML-Spray drift model (Lofstrom et al., 2013).

For ejection height and angle, the used values in drift models are adjusted according to used sprayer and droplet ejection velocity is generally measured or issued from the literature. Nevertheless, an empirical equation (Bernoulli's equation) that converts pressure into initial liquid velocity is available for calculating droplet initial speed based on nozzle discharge coefficient (depends on nozzle orifice shape), used liquid density and exerted pressure (Lee et al., 2012; Al Heidary et al., 2014). However, this equation cannot be used for air assisted sprayers which need empirical values to be available.

### **III. Atmospheric dispersion of spray droplets**

The dispersion of particles and thus the distance travelled by them depends on the mean wind intensity and the turbulence in the surface boundary layer. However, the surface characteristics have a considerable effect on atmospheric turbulences and on wind speed profile and direction. As we are studying droplet movement, a third type of parameters has to be integrated: droplet inertia and settling velocity (Dupont et al., 2006) (Fig.II.1 and II.2). As droplets undergo evaporation during their transport, it is also necessary to integrate its effect on droplet inertia and settling velocity.

In this study, the term "short-range" represents distances of less than 10 metres from the edge of the treated plot and the term "medium-range" represents distances of more than 10 m up to a few hundred metres.

#### **III.1. Turbulence in the surface boundary layer**

As pesticide spray drift occurs within the lowest atmospheric layer (surface boundary layer or SBL), it is governed by the airflows within this layer where the wind flow and air temperature are characterised by a vertical gradient influenced by the earth surface and mainly driven by two physical mechanisms: mechanical shear and buoyancy.

**Mechanical shear** is the fundamental process driving the turbulence structure in the SBL under neutral conditions. Shear originates from the friction between the air and the earth surface and its constituting objects, which characteristics are hence essential parameters describing the SBL. The SBL under neutral conditions can be described by only a few parameters: the friction velocity ( $u_*$ ) which depends on the wind speed vertical gradient, the roughness length ( $z_0$ ) and the displacement height ( $d$ ) which expresses the height at which the flow wind speed follows a logarithmic profile. The roughness length itself depends on the canopy structure and leaf area density profile.

The presence of a canopy within a landscape affects considerably the airflow by creating a strong vertical velocity gradient at the canopy-atmosphere interface (Dupont and Brunet, 2008b). Through the drag effect, the vegetation adsorbs momentum, which induces a deceleration of the airflow in the canopy. This results in an inflexion point of the wind-speed profile at the top of the canopy, which is the origin of several fluid instabilities that generate turbulence. Kelvin-Helmholtz instabilities are transverse undulations in the flow that are transformed into transverse rolls in the wind direction (Raupach et al., 1996; Chahine, 2011). These transverse rolls are broken up by longitudinal rolls issued from the highly sheared air layers that separate the transverse rolls, which generates coherent three-dimensional structures having their front at the top of the canopy and their tail in the canopy. These structures play a major role in the exchange of mass and momentum between the canopy and the atmosphere. In the case of an heterogeneous canopy, like vineyard (row crops), turbulence structure depends on the wind direction relative to the orientation of the vine rows, since the average aerodynamic characteristics of

the vine canopy vary according to the wind direction. According to (Chahine, 2011), in the case of perpendicular and 45° winds in regard to vine rows, the coherent turbulent structures that develop at the canopy scale remain similar to those observed in a horizontally homogeneous canopy with greater displacement height and roughness length than in the case of a wind parallel to the rows. Whereas in the case of a wind parallel to the rows, the air is accelerated at the row spacing level and decelerated in the rows due to the channelling effect of the air in the row spacing.

**Buoyancy** forces result from a vertical temperature gradient either positive (surface colder than the atmosphere, stable conditions) or negative (surface hotter than the atmosphere, unstable conditions). Unstable conditions generates additional mixing by free convection as buoyancy forces amplify any vertical air movement. Stable conditions on the opposite will destroy turbulence as buoyancy forces reduce any vertical air movement. Stability conditions hence greatly influence turbulent mixing in the SBL. Vertical temperature gradients are governed by heat exchanges between the surface and the atmosphere. These are driven by solar radiation and surface radiative properties, but also atmospheric turbulence, soil heat conductivity and water evaporation at the surface. All these present diurnal and seasonal variations, influenced by cloud cover, water balance and ecosystem metabolism.

Stability regimes have been described by different methods: The Pasquill method gives a qualitative estimation of turbulence parameters from basic meteorological measurements (wind speed, sun elevation and cloud cover) from which six stability classes have been defined from the very unstable (A) to the moderately stable (F). However, this method is less accurate compared to those based on non-dimensional numbers: the Richardson number (Ri) (Richardson, 1920) and the Obukhov length (L), and its associated non-dimensional number  $z / L$  (Monin-Obukhov, 1954), where  $z$  is height. These two non-dimensional numbers give a measure of the relative strength of shear and buoyancy forces as the source of turbulence in the SBL. The atmosphere can be described as neutral when the turbulent forces are dominating and stable or unstable when buoyancy forces cannot be neglected (Stull, 1988). In practice, the Richardson number is the ratio of the vertical temperature gradient (expressing the buoyancy force) to the wind speed gradient (expressing the shear force). The Obukhov length is estimated from  $u^*$  and the sensible heat flux (H). The Monin-Obukhov description of the SBL is most used nowadays (Monin-Obukhov, 1954). In this approach the vertical profiles of wind speed and turbulent parameters include stability correction functions that express the influence of buoyancy as a function of  $z / L$  (Kaimal and Finnigan, 1996). A limitation of this approach is the inability to model extremely stable or unstable conditions that fall into non-stationary or free convection regimes (Kaimal and Finnigan, 1996).

### **III.2. Additional turbulence caused by the sprayer**

Additional local turbulence caused by the sprayer displacement itself may need to be considered in the case of pesticides application (Butler Ellis and Miller, 2010; Holterman et al., 1997). Even if the geometry of the sprayer may not affect significantly the airflow (Hong et al., 2018a,b), its movement may cause increased mixing and affect spraying rate (Matthews et al., 2014). However, in spray drift models, this effect is rather indirectly represented through an additional ejection velocity affecting droplet trajectory (Butler Ellis and Miller, 2010). Holterman et al. (1997) found only a slight effect of driving speed on outfield sedimentary drift, for low  $0.36 \text{ km.h}^{-1}$  to realistic  $5.4 \text{ km.h}^{-1}$  driving speeds. Experimental data reported by Matthews et al. (2014) and Van de Zande et al. (2005) found an increased outfield sedimentary drift by up to 51% when driving speed is increased by  $2 \text{ km.h}^{-1}$  from  $4 \text{ km.h}^{-1}$  and up to 144% for driving speed up to  $16 \text{ km.h}^{-1}$ , a speed being however less common (Holterman et al., 1997).

Additional local turbulence can also be caused by the air jet created by air assistance sprayers. Sprayer air jet create instabilities at the ejection orifice called Kelvin-Helmholtz, these instabilities generate eddy structures of a proportional size to the distance from the source propagating in the direction of the ejection. Then, next to the source and at very short distances, the trajectory and the velocity of droplets are affected by this air flow (Chahine, 2011; Gil, 2007).

### **III.3. Effects of droplet characteristics on dispersion**

Particles of different diameters and density will not behave similarly in a same turbulent environment. This is mainly due to the simultaneous effect of three factors: gravity, inertia and the drag force induced by the droplet velocity relative to the fluid.

A particle falling freely into a still fluid reach after a certain time a state where gravity and drag forces equilibrate. The Stokes' law (Stokes, 1851) provides a formulation for the terminal velocity of a settling particle  $V_s$  in a still fluid, proportional to the particle diameter squared and the particle density (Guha, 2008). Stokes' law is only valid for regimes of low particle Reynolds numbers, for which the drag force varies linearly with the particle velocity (Doychev, 2014). The particle Reynolds number is the dimensionless ratio between the inertial forces and the viscous forces of the fluid relative to the particle. In high-Reynolds number regimes, which correspond to large inertia and/or large relative velocity between the fluid and the particle, the Stokes' law does not hold and a more complex expression for the drag force has to be considered in the equation of motion (Morsi and Alexander, 1972; Aylor, 2002; Loubet et al., 2007). In turbulent flows, the consideration of all the forces acting on the particle, and of the fluid fluctuations, lead to a higher gravitational settling velocity that found in still or laminar flows and to clustering of particles in zones of low vorticity where the particle concentration is locally higher than in the surrounding (Wang and Maxey, 1993; Maxey, 1987).

Particle weight also induce inertial effects. Since the particle weight is proportional to the diameter cubed, the particle diameter ( $D$ ) is the key parameter of inertial effects. The inertial effect can be expressed through the relaxation time of the particle, or Stokes time, equal to the terminal velocity divided by the gravity acceleration  $g$ , and hence proportional to  $D^2$ . Particles with small diameter will hence have a small relaxation time and will follow the fluid, while larger particles will leave the fluid filaments due to inertia. This leads to the so-called crossing trajectory effect (Csanady, 1963), that leads to a reduced autocorrelation time of heavy particles compared to the fluid (Sawford and Guest, 1991). In the context of a droplet released by a sprayer, the Stokes time can be viewed as the time a particle needs to relax to the surrounding airflow, i.e. the time at which particle velocity becomes equal to air velocity.

Under the influence of settling and inertia, smallest particles (diameter less than 10  $\mu\text{m}$ ) behave almost like fluid particles while larger particles are influenced by their sedimentation, and less influenced by the speed of the surrounding airflow (Giardina and Buffa, 2018).

### **III.4. Droplets evaporation during their transport**

During their transport and under the influence of climatic and atmospheric conditions, spray droplets might undergo evaporation, affecting thus their size distribution and consequently their atmospheric dispersion and deposition. Indeed, it has been shown that, in some cases, evaporation might increase pesticide spray drift distance by a factor of 4, which can considerably expand the contamination perimeter (Lebeau et al., 2011). Numerous studies demonstrated that evaporation has a considerable effect on droplets having a diameter under 250  $\mu\text{m}$ , as substantial evaporation may occur while they are still airborne which increases significantly their driftability. In contrast, evaporation has a small effect on the drift of larger particles, as they reach the ground faster than they evaporate, their trajectory being hence negligibly affected (Lebeau et al., 2011).

Spray droplets are a mixture of water, active ingredient, co-formulants and potential additives such as surfactant, adjuvant or retardant, these additional compounds can also change droplet water evaporation process (Gil and Sinfort, 2005). However, experimental studies gave evidence that evaporation of pesticide droplets can generally be represented as a pure water evaporation process, for both still and moving droplets relative to the surrounding fluid (Thompson and Ley, 1983; Fuchs, 1959). Later, Wanner (1980) and Dennison and Wedding (1984) confirmed this assumption (Duan et al., 1992), while (Duan et al., 1994) showed that this assumption is only appropriate when droplet size is smaller than 600 $\mu\text{m}$ . For bigger droplets, the evaporation rate may be over-estimated under specific experimental conditions if pure water is considered. In the end, most studies adopt the hypothesis that pesticide droplets are merely water droplets, considering that, based on the theory of “solid core” of Elliott and Wilson (1983), all suspended material is grouped in the drop centre, so only water will stay at the surface

of the drop and therefore will be exposed to evaporation (Holterman et al., 1997). In addition, for water-based pesticides, the vapour pressure of water is much higher than that of dissolved pesticide, leading to a negligible pesticide evaporation (Holterman, 2003).

When considering a pure water droplet, isolated and immobile in a stagnant medium, two phenomena are involved in water evaporation: conduction and diffusion (Gil and Sinfort, 2005). As they leave the nozzles, droplets reach the wet bulb temperature, having a temperature lower than the ambient air temperature and a surface vapour pressure higher than the air vapour pressure. As a consequence, a sensible heat transfer from the surrounding air to the droplet takes place (Kincaid and Longley, 1989), while in mean time the vapour pressure gradient between the surface of the droplet and the environment leads to evaporation (Belkhef, 2008).

The variation of air temperature, relative humidity and atmospheric pressure change considerably the water evaporation from droplets (Asman et al., 2003). As the ambient temperature rises, the kinetic energy of the water molecules increases, resulting in an increased evaporation of water (Kincaid and Longley, 1989). In contrast, a decrease in air pressure and relative humidity increases evaporation (Hong et al., 2018a,b). As atmospheric pressure declines, molecules close to the surface of the droplet are more likely to break their intermolecular attraction, which generates an increase of evaporation (Tanner, 2011). When relative humidity decreases, the water pressure gradient increases increasing evaporation.

As pesticide droplets are sprayed with an initial speed in a moving fluid, the size of the viscous layer diminishes when the relative fluid velocity increases which increases considerably the mass transfer of vapour from a drop surface to the air (Belkhef, 2008). Thus, the consideration of convection effect on evaporation is key for pesticides sprays (Beard and Pruppacher, 1971). Droplets in assisted spray drift may however evaporate less than in a non-assisted spray as they have a small initial velocity relative to the surrounding air in the first milliseconds of their trajectories. Finally, the percentage of active ingredient in the mixture is also key in determining the final diameter of an evaporating pesticide droplet. Indeed, with the assumption that the water evaporates, the volume of a fully evaporated particle should hence be equal to the volume of active ingredient contained in the droplet. With active ingredients ranging from 0.02 to 2% of the mixture volume, this means that the evaporated particle diameter may range from  $10^{-2}$  to  $10^{-5}$  of the initial particle diameter.

### **III.5. Modelling approaches in spray drift models**

#### **III.5.1. Droplet dispersion models**

A range of models exist for predicting the trajectory of pesticide droplets under field conditions, with varying degrees of details and combinations in the implementation of the previously described processes. Therefore, the choice of a model will mainly depend on the objectives of a given study. Here we restrict the review to dispersion models used for pesticides only.



As a first approach, the **Gaussian models** provide a simplified, steady-state solution of the advection-diffusion equation (Chahine, 2011). They are used to describe the mean along-wind, vertical and across-wind distribution of particles concentration as Gaussian functions where the standard deviation in each direction changes with the distance downstream from the particle source (Reible, 1998). Gaussian models require the hypothesis of spatially homogenous atmospheric conditions and flat homogenous terrain. In other research areas some more elaborated Gaussian-like approaches account for vertical profiles in turbulence properties (Bouvet and Wilson, 2006). If these models allow a good description of dispersion for distances downstream of the source higher than 50 m (Craig, 2004), they poorly describe dispersion near the source. They also describe the source with a low accuracy due to the fact that droplet diameters are defined by size classes (the precision depends on the number of classes) and their evaporation is not or hardly taken into account (Nuyttens, 2007). These models assume that deposition does not change the particle concentration in the atmosphere, and the deposition can be partially considered by reducing the source term, as a function of distance (McCartney and Fitt, 1985 in Jarosz, 2003). Some of these models are able to consider the deposition by assuming it to be uniform over the plume area *i.e.* independent of surface nature or physical barriers (Lushi and Stockie, 2010). Among the selected models, this approach has been used in RTDrift and OML- Spray drift models.

Unlike Gaussian models, **Lagrangian stochastic models** allow a good description of dispersion in short and medium distances, with the ability to consider complex atmospheric conditions and heterogeneous and near surface turbulences, as well as in-canopy transfer. In addition, they are able to characterize in details the source with a distribution for particle size, ejection height, angle and speed. Furthermore, by essence, Lagrangian stochastic models are built to account for phenomena affecting droplet diameter along their trajectory such as evaporation. Lagrangian stochastic models simulate stochastically each individual trajectory of a group of particles that evolves over time along its path from emission to deposition (Jarosz, 2003; FOCUS, 2008) under the effect of aerodynamic and gravity forces and inertia (Smith, 1970; Reichard et al., 1992 b; Urip et al., 2002 in Nuyttens, 2007). The concentration is computed as the probability of presence of particles in a given volume (Chahine, 2011). The main theoretical limitation of Lagrangian stochastic models for heavy particles is that inertia leads to a decoupling of the fluid and particle trajectories, which requires either to neglect inertia, or to resolve a two-trajectories model for both the particles and the fluid. In any cases, some assumptions have to be made, and especially a reduced autocorrelation time scale for the fluid trajectories, to account for the cross-trajectory effect (Reynolds, 2000, 2018). The second limitation is that they require the turbulence statistics to be provided within the boundary layer (Bouvet and Wilson, 2006).

Lagrangian stochastic models are the most often used in pesticide droplet drift models, for example in the Hong model, AGDISP/AgDRIFT, ARPS, the Silsoe Spray Drift model, the Thompson and Ley model or IDEFICS. In AGDISP/ AgDRIFT, and ARPS, the Lagrangian stochastic model includes a

particle and a fluid trajectory model, while only ARPS includes the cross-trajectory effect (Teske et al., 2002; Chahine, 2011). The Hong model does not include a fluid trajectory model, but accounts for an eddy lifetime and a cross-trajectory time that plays a similar role (Hong et al., 2018a,b). The Thompson and Ley model and Silsoe Spray Drift Lagrangian dispersion models are essentially similar and adapted from Hall (1975): the model includes only the droplet dispersion equation, and the fluid Lagrangian time scale, though not corrected for cross-trajectory (Thompson and Ley, 1983; Miller and Hadfield, 1989). This model would tend to over disperse. The IDEFICS model uses a similar mathematical equation for particle trajectories than Thompson and Leys but with no stochasticity and using the particle Lagrangian time scale for the fluid, which will lead to an over reduced dispersion for large particles. The description of the turbulence is also unclear in IDEFICS (Holterman et al., 1997).

A combination of Gaussian and Lagrangian approaches is used in **puff models**, where plumes are moving along the mean Lagrangian trajectory, while the concentration is represented within each plume with a Gaussian diffusion approach (Hong et al., 2021).

**Eulerian models** describe the fluid turbulence and the distribution of particles in a fixed system as spatio-temporal field, discretizing and solving the complete advection-diffusion equation over time and on a 3D gridded domain representing the atmosphere (FOCUS, 2008). There is a diversity of Eulerian models, varying essentially in the computation of turbulence that can be done using a first or second order closure assumption or with an LES (Large Eddy Simulation). These models calculate the mean particle concentration in each grid cell of the computational domain and can account for various physicochemical processes (Dupont et al., 2006). Computational fluid dynamics (CFD) models solving the Navier-Stokes equation, belong to the Eulerian category with second order closure assumptions, that can be used to investigate small or large scale 3-D flows taking into account turbulence created by sprayers fans or canopies. Recent advances in computing power have allowed them to be deployed on a wide range of topics, including the description of spray droplet dispersal. Eulerian-Lagrangian CFD approaches couple Lagrangian stochastic dispersion models with the Eulerian computation of turbulence. These are especially powerful to analyse relatively fine but crucial effects such as spray dispersion near the nozzle, even for air-assisted systems, droplet break up, collisions and coalescence. They are also powerful in evaluating spray interception by vegetated and soil surfaces including local disturbance of the turbulence by the tractor and sprayer system. Salcedo et al. (2017) developed a 2D-model for the tree context (citrus) that includes a number of factors such as air temperature, humidity, turbulence, nozzle type and orientation, and tractor ground speed. However, the authors note the need for further sensitivity testing, against datasets and a move to a 3D version of the model. Duga et al. (2017) also proposed a model based on a CFD approach coupled with a Lagrangian particle transport model to describe sedimentary and airborne drift in orchards, which they have successfully confronted with a dataset and applied to test anti-drift nozzles effectiveness. Recently, Hong et al. (2018a,b)

developed a similar CFD model for air-assisted sprayers in apple orchards, taking into account the relative motion of the sprayer by use of the “sliding mesh” technique (Fluent, 2014).

In most models, the turbulence is parameterised above the canopy using the Monin and Obukhov profiles, but often stability is neglected, excepted in Thompson and Ley model and the OML Spraydrift model. Most CFD models do not include stability, as this requires solving in parallel the Navier Stokes equation, the full energy balance of the surface, as well as latent and sensible heat transfer equations, which is a further degree of complexity. In IDEFICS stability seems to be accounted for only on eddy diffusivity but not on wind speed. In ARPS, the Lagrangian approach is coupled to a Large Eddy Simulation model with no stability, allowing analysing the effect of the vine structure or air jet induced by the sprayer on spray drift (Chahine et al., 2014).

The effect of the canopy on local turbulence is also accounted in different manners. The CFD model used by Hong et al. (2018a,b) represents the canopy as a porous media that perturbs the pressure, velocity, and turbulent kinetic energy according to the Darcy-Forchheimer model (Whitaker, 1996, 1986; Fluent, 2014). In ARPS, vine canopy geometry is defined by its leaf area density considered to be heterogeneous vertically and uniform in the row direction. In IDEFICS and Silsoe Spray Drift models, the canopy is only characterised by its height. For the first model, an average wind speed inside the canopy is calculated based on the defined height following Thom (1971) while for the second a vertical profile of wind speed is calculated based on the empirical equation of Raupach and Thom (1981) and turbulence within it are defined according to Wilson et al. (1982). In Thompson and Ley and AGDRIFT, the displacement height  $d$  is considered the lower limit of the domain and drops are not followed inside.

### **III.5.2. Droplet evaporation**

Evaporation models can be classified into six categories according to their degree of complexity (Sirignano, 2010; Tanner, 2011) namely: constant droplet temperature, infinite liquid-conductivity, conduction limit, effective conductivity, vortex, and full solution of the Navier–Stokes equations. The first two categories consider simply the temperature inside the droplet, while the four later ones deal with more complex processes like the effect of the transient heating process, the internal drop recirculation on the conductivity or the internal flow and its effect on droplet heating, respectively. However, as they are integrated into a drift model, most of the pesticide evaporation models are within the two first classes that consider the temperature of the droplet to be uniform and either stay constant with time or vary uniformly with time. The processes taken into account in the four other categories are considered of minor concern compared to atmospheric transport over the short droplet lifetime, while significantly increasing computational costs (Tanner, 2011). Holterman (2003) argued that the transient phase of droplet cooling before the evaporation and the effect of surface tension can be neglected as their occurrence time is negligible compared to droplet lifetime (Sinfort, 2005).

Thus, the existing pesticide evaporation models are mainly based on three approaches: (1) Ranz and Marshall (1952a,b) model based on mass conservation (cited in Goering et al., 1972 and Williamson and Threadgill, 1974); (2) Trayford and Welch (1977) model based on “D-squared law” in which the evaporation time scale is calculated as a function of the initial droplet diameter squared; and (3) the Davies (1978) model based on energy conservation. Among the 8 reviewed model, only the two first approaches are used: the first one is used by Holterman et al. (1997) in IDEFICS, Hong et al. (2018a,b) model, OML-SprayDrift and Thompson and Ley model. The second one is used in AGDISP/AgDRIFT (Teske et al., 2016) with a modification of the evaporation rate to add convection effects (Belkhef, 2008) derived from Fuchs (1959). In Gaussian models that do not represent the droplet along its trajectory, the challenge is to represent evaporation through its average effect on drift. In RTDrift an averaged travel diameter is computed based on evaporation time and lifetime, which is substituted to the initial droplet diameter to represent evaporation effects on dispersion. In the OML-SprayDrift model the way evaporation is accounted for in the Gaussian model is unclear although it seems to be made in a similar way as in RTDrift. Water evaporation is neglected in ARPS.

## **IV. Dry deposition of spray droplets**

Pesticide droplets are removed from the atmosphere onto the earth surface by either dry or wet deposition (Mariraj Mohan, 2016), leading to a pesticide flux into waterbodies, crops, vegetation, soil, roads, houses or human bodies (Cherin, 2017). In this manuscript we only deal with short-range processes and therefore do not consider wet deposition which is prevailing over larger scales but has little effect very locally, especially because pesticides are not applied under rainy conditions. The factors influencing droplet dry deposition belong to three categories: (1) atmospheric properties and meteorological conditions, (2) the nature of surfaces, and (3) the compounds that are depositing (Fig.II.1). The particle transfer from the atmosphere to the surface can be summarized by three physical mechanisms: aerodynamic transport and settling, boundary layer transport, and surface uptake and interactions (Mariraj Mohan, 2016). The combination of the four mechanisms leads to the definition of the particle deposition velocity. Particle properties like diameter, density, shape and composition, are key for the three mechanisms (Davidson and Wu, 1990), such as the turbulence and the properties of the depositing surfaces.

### **IV.1. Processes involved in dry deposition**

**Aerodynamic transport and settling** of particles is the atmospheric dispersion process, which is essentially an eddy diffusion process combined with a settling process. This defines the transfer rate of particles towards the surface from a given height. All the underlying mechanisms have been discussed at length in the “atmospheric dispersion” section. The transfer can be expressed as an aerodynamic resistance which is inversely proportional to the transfer rate (Cherin, 2017; Seinfeld and Pandis, 1998).

In the case of stable atmospheric condition, atmospheric mixing is reduced, leading to particles having a lower transfer rate towards the surface. In contrast, under unstable conditions, the atmospheric mixing is stronger and particles have a higher transfer rate towards the surface. Sedimentation and particles inertia are always present. Sedimentation is essentially adding a settling velocity onto the aerodynamic transfer rate, which increases the deposition velocity for particles larger than 10  $\mu\text{m}$  (Seinfeld and Pandis, 1998). Inertia affects particles through the cross-trajectory effect, which diminishes the mixing of particles larger than  $\sim 100 \mu\text{m}$  (Bouvet et al., 2006). Relative humidity and air temperature influence droplets evaporation and hence deposition process through their effects on droplet diameters (Rouspard, 2013). Among these processes, stability regimes were shown to be essential by Davidson and Wu (1990).

Once particles reach the **quasi-laminar boundary layer** just above the canopy or the ground, its transfer to the surface depends on Brownian diffusion, sedimentation, and microphysical mechanisms such as interception and inertial impaction (Noll et al., 2001). Brownian diffusion is the transport of particles by diffusion along a concentration gradient due to the stochastic effect of air molecules impaction on particles under thermal agitation. The effect of this deposition mechanism decreases with the increase of particle diameter and it mainly concerns submicron particles (diameter less than 0.1  $\mu\text{m}$ ) (Davidson and Wu, 2012). As pesticide droplet diameter is much higher than the submicron (several  $\mu\text{m}$ ) even after water evaporation, the Brownian diffusion is not considered in most pesticide spray drift deposition studies. Sedimentation is only significant for horizontally oriented surfaces, and concerns particles larger than 10 $\mu\text{m}$  (Davidson and Wu, 2012).

When the air flows over a rough surface or around an obstacle that protrudes from the surface, particles that are carried by this airflow may not follow the abrupt changes in flow directions around the obstacle or surface. Due to their inertia and the rapid change in airflow direction, the particles will impact on the surface (Rouspard, 2013). This phenomenon is called inertial impaction, which is more important for obstacles protruding from the surface such as elements of a vegetation cover (Davidson and Wu, 2012). Like sedimentation, inertial impaction increases with the size and/or mass of the particle, and it concerns particles with a diameter greater than one micron (Cherin, 2017). Inertial impaction can be described by the impaction efficiency, which is the ratio of the number of impacted particles to the number of incoming particles. Impaction efficiency varies according to the studied surface, it is parameterised as a function of the Stokes number ( $St$ ) which is the product of the particle relaxation time with the fluid velocity divided by the characteristic dimension of the obstacle (Rouspard, 2013). A particle with a large Stokes number hence leads to a large impaction efficiency.

During their trajectories, if these particles come to pass next to an obstacle within a distance which is less than or equal to their radius, they are deposited over the obstacle by interception (Rouspard, 2013). As inertial impaction, deposition by interception is a function of the particle diameter and the characteristic dimensions of the obstacle, but it mainly concerns particles that do not deposit by the other

dominant processes of Brownian diffusion and settling, and hence particles with a diameter greater than the submicron and smaller than a few microns (Cherin, 2017).

Finally, after reaching the surface, particles go on a last step in their deposition process, which is the **surface uptake and interactions**. This step depends on particles and the deposition surface characteristics. Indeed, particles may adhere to the surface, react chemically with it, bounce off, splash, or be resuspended into the airflow (Davidson and Wu, 2012). Resuspension is a process that concerns inert particles that stays on the leaves or ground and are detached by the movement of leaves and wind gusts (Jarosz, 2003; Jacobson, 2005). It can hence be neglected for pesticides droplets. Particles bounce occurs generally when particles hit a smooth surfaces and when their incident deposition velocity exceeds a critical velocity, which is inversely proportional to its diameter (Dorr et al., 2015). In contrast, when particles come into contact with a humid surface, they do not experience a rebound due to the marked effect of moisture on deposition and particle uptake (Cherin, 2017). As pesticide droplets are considered to behave like pure water droplets and as the rebound is more significant for solid particle rather than for liquid particles, the bounceoff and the chemical interaction with the surface are generally neglected in the context of pesticide spray drift deposition. Droplets may rather splash onto a leaves as shown by Dorr et al. (2015).

## **IV.2. Modelling dry deposition in spray drift models**

Droplet deposition can be represented by a deposition velocity (Asman et al., 2003; Seinfeld and Pandis 1998) with methodologies varying considerably from one study to another, due to differences in approaches to consider particle properties, depositing surfaces, meteorological conditions, modelling scale, the needed degree of precision and the aim of each study. The representation of deposition is also dependent on the type of dispersion model/framework used (Gaussian, Lagrangian or Eulerian). Generally, in deposition velocity models, the horizontal concentration gradient is considered negligible, as sources are considered to be far away. In such cases particles deposition velocity is estimated through a resistive model where all previously described deposition processes are considered separately (Mariraj Mohan, 2016). In spray drift models, the aerodynamic transport is included in dispersion equations, which also determine the horizontal concentration gradients. However, supplementary processes must be considered to represent deposition in the canopy.

We can thus distinguish models that aim to describe the deposition of pesticide out of the treated field and models that aim to describe ground deposition inside the treated field and interception by the canopy. Often, in models that aim to represent exclusively pesticide spray drift outside the treated field, only deposition on downwind bare soil, grassland or water is considered, as in IDEFICS (Holterman et al., 1997), AgDRIFT (Teske et al., 2002) and Thompson and Ley models (Thompson and Ley, 1983). As these surfaces show small canopy elements, they only considered deposition by sedimentation, which they integrate directly into their dispersion equation (Dupont et al., 2006). In IDEFICS, deposition into

the canopy was solely differentiated by deposition height, which was set equal to the displacement height, whereas Thompson and Ley model rather tested the sensitivity of spray drift to the variation of deposition height and to the consideration of an absorption coefficient that depends on droplets size. In AgDRIFT model (Teske et al., 2002), deposition is assumed to be identical to a classical Gaussian deposition independently from the type of deposition surface (vegetation, water, bare soil). In RTDrift, the deposition velocity is set to the sedimentation velocity, which is justified as the concentration is considered at the ground (Lebeau et al., 2011). In OML-SprayDrift, the concentration is considered at 0.5 m height and hence the deposition velocity includes the aerodynamic transport from 0.5 m and the boundary layer transport (Lofstrom et al., 2013).

Models that aim to represent ground deposition and interception by the canopy inside the treated field, inertial impaction and interception is necessary to account for. In this category, two probabilistic approaches (ARPS, Hong model) and one based on the local concentration of pesticides close to the ground are found in the reviewed models (AgDrift). The probabilistic approaches are both based on a single probability of deposition that includes both the deposition and the retention of pesticides droplets within the canopy, that is itself calculated based on two probabilities: (i) the probability that pesticides droplet encounter vegetation and (ii) the probability that vegetation capture the droplets. The first probabilistic approach is derived from the empirical equation of Walklate (1992) and used for pesticide application in orchard by Hong et al. (2018a,b). The probability of droplets encountering canopy is an empirical function of leaf area density (LAD) and droplet velocity. However, this type of approach is not generic and its use is restricted to models that are dealing with pesticide application over the same type of canopy as the experimental data that leads to this empirical equation or needs a readjustment and a validation for other types of canopies. The second approach is a Lagrangian approach, where the deposition of pesticide droplets within the canopy during model time step can be calculated separately for all canopy elements, namely, leaves, branches and trunk (Aylor, 2005). The deposition probability on vegetation is expressed as a capture efficiency which is a function of canopy geometry, vegetation element inclination (for leaves), the characteristic size of the considered elements, settling velocity, vegetation density and deposition efficiency. This approach has been first described by (Aylor and Flesch, 2001) for pollen particles deposition and then used for pesticide spray drift droplet deposition by Chahine (2011) in ARPS. The capture efficiency developed in Aylor and Flesch (2001) is limited to particle diameters below 80  $\mu\text{m}$  and a maximum of 0.86. This should hence be extended for pesticides droplets that are much larger, and could reach a capture efficiency of 1 for large Stokes numbers (Mirzaee et al., 2019).

The third approach is adapted for the Eulerian models. Instead of expressing a probability of deposition of a single trajectory as in Lagrangian models, in Eulerian models the deposition is expressed by a deposition rate computed as the concentration close to the ground multiplied by a deposition velocity (Dupont et al., 2006).

## **V. Synthesis and discussion**

The previous sections show a range of tools of varying complexity to model sedimentary and more recently airborne pesticide drift and for some of them interception by the treated crop (Table II.1). We discuss here the key processes related to spray drift and the limitations concerning their modelling.



Table II.1. Reviewed process based spray drift models

Models *	Application and source representation				Droplet dispersion and related processes				Deposition		References
	Crop type	Source	Used sprayer	Droplet size distribution	Evaporation	Approach	Atm. stability	Canopy effect on atm. turbulence	Ground deposition	Canopy interception	
<b>AGDISP / AgDRIFT</b>	Annual (sorghum)	Punctual	Boom sprayer	Defined classes from measurements	Tayford and Welch model <sup>2</sup>	Lagrangian	Neutral	Not considered	Sedimentation (Stokes' Law + drag coefficient)	Not considered	Nsibande et al., 2015; Teske et al., 2009
<b>ARPS</b>	Perennial (vine)	Punctual	Axial fan sprayer	Gaussian distribution	Neglected	Lagrangian + LES <sup>3</sup>	Neutral	Yes + in canopy wind vertical profile	Sedimentation (Stokes' Law + drag coefficient)	Lagrangian approach – Probability equation <sup>4</sup>	Chahine, 2011 <sup>5</sup>
<b>Hong et al. (Fluent)</b>	Perennial (apple orchard / Vineyard)	Cylindrical volumes treating one row side	Air assisted sprayer	Rosin-Rammler <sup>6</sup> distribution	Ranz and Marshall model <sup>7</sup>	Lagrangian (random walk model)	Neutral	Yes <sup>8</sup>	Sedimentation (Stokes' Law + drag coefficient)	Empirical probability equation of (Walklate, 1992) <sup>9</sup>	Hong et al., 2018a,b; Fluent 2014
<b>IDEFICS</b>	Annual and cutgrass	Punctual	Boom sprayer	Defined classes from measurements	Ranz and Marshall model <sup>7</sup>	Lagrangian – (random walk model <sup>10</sup> )	Neutral - Stable - unstable <sup>11</sup>	Set an average wind speed inside the canopy <sup>12</sup>	Sedimentation (Stokes' Law + drag coefficient)	Considered <sup>13</sup>	Holterman et al., 1997; Holterman

<sup>2</sup> Trayford and Welch (1977) / Equation of (Amsden, 1962) used to compute droplets lifetime

<sup>3</sup> ARPS is based on Large Eddy Simulation (LES) and (Vinkovic, 2005) method

<sup>4</sup> Probabilistic equation of Aylor and Flesch (2001)

<sup>55</sup> Adapted to spray drift prediction by (Dupont and Brunet, 2008a, b)

<sup>6</sup> Rosin and Rammler (1933)

<sup>7</sup> As described by Williamson and Threadgill (1974), (Ranz and Marshall, 1952a, 1952b; Goering et al., 1972; Williamson and Threadgill, 1974)

<sup>8</sup> Based on Darcy-Forchheimer model (Whitaker et al., 1986, Whitaker et al., 1996, Fluent, 2014)

<sup>9</sup> Equation issued from experimental data of (May and Clifford, 1967) on orchard

<sup>10</sup> Thompson and ley (1983)

<sup>11</sup> Stable and unstable conditions considered (pers. com. H.J. Holterman)

<sup>12</sup> Average wind speed inside the canopy Following Thom (1971) and eddy recirculation effect is ignored as transition effect is not marked for short crops or cut grass

<sup>13</sup> Differentiated from sedimentation by a deposition height (canopy height) and depending on droplets size where small particles are allowed to penetrate the

Models *	Application and source representation				Droplet dispersion and related processes				Deposition		References
	Crop type	Source	Used sprayer	Droplet size distribution	Evaporation	Approach	Atm. stability	Canopy effect on atm. turbulence	Ground deposition	Canopy interception	
											2003; Pers.com. H.J. Holterman 2022
<b>OML-Spray Drift model</b>	Annual	Punctual	Boom sprayer	Defined classes from measurements	Ranz and Marshall model <sup>7</sup>	Gaussian	Neutral - Stable - unstable	Not considered	Sedimentation <sup>14</sup> (Stokes' Law + drag coefficient)	Not considered	Lofstrom et al., 2013
<b>RTDrift</b>	Annual (wheat and chicory)	Punctual	Boom sprayer	Defined classes from measurements	Tayford and Welch model <sup>2</sup>	Gaussian	Neutral	Not considered	Sedimentation (Stokes' Law + drag coefficient)	Not considered	Lebeau et al., 2011
<b>Silsoe Spray Drift Model</b>	Annual (wheat) and short grass	Punctual	Boom sprayer	Defined classes from measurements	Usually not activated <sup>15</sup>	Lagrangian (random walk model)	Neutral - Stable - unstable <sup>16</sup>	Yes (empirical equation <sup>17</sup> )	Sedimentation (Stokes' Law + drag coefficient)	Probabilistic approach <sup>18</sup>	Butler Ellis & Miller, 2010 <sup>19</sup> ; Butler Ellis et al. 2017; Pers.com. C. Butler Ellis 2022
<b>Thompson and Ley</b>	Annual	Punctual	Boom sprayer	A defined droplet size	Ranz and Marshall model <sup>7</sup>	Lagrangian (random walk model)	Neutral - unstable	Not considered	Sedimentation (empirical equation <sup>20</sup> )	Not considered <sup>21</sup>	Thompson & Ley, 1983

canopy – no further investigation is addressed- (pers. com. H.J. Holterman)

<sup>14</sup> Deposition outside the treated field is calculated according to resistive method

<sup>15</sup> Original model (Miller and Hadfield, 1989) consider the evaporation. In

*Silsoe model it is considered but usually not activated under UK conditions (pers. com. C. Butler Ellis 2022)*

<sup>16</sup> Take into account stable and unstable conditions, but usually applied under neutral conditions (pers. com. C. Butler Ellis 2022)

<sup>17</sup> Empirical equation of Raupach and Thom (1981) for wind profile and the empirical equation of Wilson et al. (1982) for turbulence inside the canopy

<sup>18</sup> (pers. com. C. Butler. Ellis)

<sup>19</sup> A modified and a validated version of (Miller and Hadfield, 1989) model

<sup>20</sup> Empirical equation of (Mason, 1971)

<sup>21</sup> Tested the sensitivity of pesticide outfield deposition to the variation of canopy interception by varying interception surface height and interception efficiency as function of droplets size

## **V.1. Droplet characteristics at emission are key parameters that needs determination**

None of the selected models describes the atomization mechanisms. The droplet size distribution is thus described empirically by giving droplet diameter classes based on measurements in IDEFICS, AgDrift, RTDrift, Silsoe Spray drift model and OML- Spray drift model, or by using the Rans-Rammler distribution in (Hong et al., 2018a,b) model, or a Gaussian distribution in ARPS. The ejection height and angles are prescribed depending on the type of used sprayer and the ejection velocity is either measured or taken from the literature. The source is generally assumed punctual except in Hong et al. (2018a,b) where the source is a collection of 20 moving cylindrical volumes representing the nozzles. We found that droplet diameter and ejection velocities relative to the surrounding air are key factors that determine dispersion distances and greatly influence also droplet evaporation and hence the potential for long range transport. Overall, we found that droplet emissions is a process highly dependent on the sprayer and operating conditions, but also on the mixture used. Most models use empirical droplet size distribution and velocity, which is a sound approach as it is the most robust. However, this stresses the need for good quality measurement of these parameters for the range of field operating conditions for future modelling. This includes the need for droplet diameter measurements down to a few  $\mu\text{m}$  to include droplets that may contribute to longer-range transport.

## **V.2. Droplet evaporation may lead to farther dispersion of small particles**

Water evaporation from the droplet is taken into account in most models with different approaches (Table II.1). Drift models usually consider evaporation of a single droplet, without interactions between droplets (OML, RTDrift, ...) although the evaporation of a droplet may be slowed by the evaporation of other droplets around. However, due to the high speed at which droplets are ejected, this phenomenon does not last long and therefore its overall impact can be neglected (Holterman, 2003; Sinfort, 2005). For the Silsoe model, even if in the original model of (Miller and Hadfield, 1989) the evaporation was taken into account, the authors indicated that water evaporation can be neglected as the model was intended to be applied at short distance and under climatic conditions prevailing in UK, but that, for larger distances or under different conditions, it should be activated. However, depending on weather conditions and droplets diameter, evaporation can take place over very short time scales ranging from sub-seconds ( $\approx 0.1\text{s}$ ) to about a minute (up to  $\approx 100\text{s}$ ), especially in air-assisted applications. Furthermore, this review shows that mostly water evaporates from the droplets. Consequently, the final diameter of an evaporating droplet is equal to the initial diameter multiplied by the cube root of the pesticide content in the mixture, a number ranging from  $10^{-2}$  to  $10^{-5}$ . This has profound implications as

for an initial distribution of droplets, say from 50 to 500  $\mu\text{m}$ , the final distribution would contain particles mostly smaller than 5  $\mu\text{m}$  and even smaller than 0.5  $\mu\text{m}$ . Of course, the high settling velocity and downward ejection velocity of most sprayers in annual crops would lead to a time too short for the larger particles to evaporate before reaching the ground or the plant. However, for applications in trees or vines where the ejection is not towards the ground and air assisted systems are present, the fraction of particles quickly evaporating may be large. This seems to be confirmed by reported application efficiency of pesticides application, which is the ratio of deposited to applied pesticides, ranging from 50% to 90%. Indeed, this means that from 10% to 50% of the applied mixture does not reach the target field. However, these large losses are not recovered in sedimentary drift, which suggests a significant loss happens through farther dispersion of evaporated particles. It is therefore essential for future models validation to provide measurements of droplets downwind from application covering the micron and submicron ranges to better constrain evaporation models and close the pesticide application mass balance.

### **V.3. Atmospheric dispersion needs to consider atmospheric stability, canopy structure effect, and sprayer characteristics and movements**

The approaches most used to describe atmospheric dispersion are the Gaussian or the Lagrangian approaches, the latter being often preferred (Table II.1). Moreover, the stability conditions are in general restricted to neutral conditions except in Thompson and Ley's model, OML-Spraydrift, IDEFICS and SSDM. As stability conditions influence considerably the transfer rate towards the surface, it is important to consider it in future modelling.

For row crops, the effect of the wind direction compared to row orientation may be significant as the aerodynamic characteristics of the canopy vary with the wind direction. The row canopy acts similarly to an homogenous crop when wind is perpendicular or at 45° comparing to row orientation while a wind blowing in parallel to row may create a flow channelling in row direction as shown by Chahine et al. (2014). Losses downwind from the treated field may consequently be higher when the wind direction is parallel to row orientation.

The sprayer itself seems to have a negligible effect on local turbulence (Butler Ellis and Miller, 2010; Holterman et al., 1997), but the movement of the sprayer relative to the air may be important for high driving velocity. Holterman et al. (1997) and Lebeau et al. (2011) further shows that the vertical movement of boom sprayer induced by ground irregularities during sprayer movement affects considerably outfield spray drift, thus must be considered in spray drift models for annual crops.

In perennial crops, the use of air jet is common in order to facilitate drop penetration into the crop canopy. The way to include this perturbation in the atmospheric turbulence is question not extensively treated yet. Indeed, in the literature, the effect of air jet on spraying performance is more characterised than its effect on local turbulence or its consequences on spray drift. Chahine (2011), using a Large Eddy

Simulation approach however shows that high velocity fields penetrating the field in the direction perpendicular to the rows are created corresponding to the sprayer air jets and these areas are convected in the direction of the sprayer movement. Few seconds after the sprayer passage, a residual velocity is observed a few meters away from the initial emission zone (up to 4m). This velocity is due to the advective movement of the air jet already emitted by the source. Accordingly, the dispersion of particles in vine rows next to the sprayer is dominated by the airflow generated by the sprayer air jet itself rather than meteorological conditions.

This effect on local turbulence and droplet trajectory may be addressed by determining zones where the sprayer air jet influences local turbulence around the sprayer. Some simplifications may then be applied by assuming that the droplets are released with a uniform velocity at the edges of this zone.

#### **V.4. Droplet deposition needs to consider canopy filtering capacity**

As the filtering effect of canopy and the recirculation area at the field edge are less marked on spray drift predictions when treating annual crops (short crops) than perennial crops (high crops), models dealing with annual crops, like Silsoe spray drift model and IDEFICS, consider only the height of the crop to compute interception. In contrast, ARPS and the CFD model of (Hong et al., 2018a,b), include the effect of the structure and the porosity of the canopy on turbulence and hence impaction and interception, which is key for deposition onto perennial crops since the droplet ejection velocities are directed horizontally and upwards. This analysis suggests that sedimentation is the key parameter to model for short annual crops, which does not depend so much on leaf area density (as is solely used in the Silsoe spraydrift and IDEFICS models). Whereas, in addition to sedimentation, impaction and interception are key for perennial crops which therefore requires a good description of the canopy density and geometry (as in Hong et al., 2018a,b and Chahine, 2011). However, even in annual crops, spray drift outfield deposition may be significantly impacted by the height at which the deposition is assumed to occur and the filtering capacity of crops according to droplets diameter (Thompson and Ley, 1983). This highlights the need of selecting the most appropriate deposition approach according to the crop type and its growth stage (Hong et al., 2018a,b).

Improvements can be made to models dealing with annual crops by integrating impaction in the deposition process. The probabilistic approach of Aylor and Flesch (2001) used by Chahine (2011) seems more relevant to calculate droplet interception by the canopy comparing to the empirical equation used in Hong et al. (2018a,b), as it accounts for the characteristics of the canopy and thus can be adapted to various crop types (annual and perennial). For a more realistic representation of the impaction process near the sprayer, an additional term can be added to the probabilistic equation used by Chahine (2011) in order to account for the impaction of particles on the horizontal surface of the canopy.

## **V.5. Models need to consider airborne concentrations and farther dispersion distances**

The reviewed spray drift models outputs comprise (1) the spray interception within the treated field, (2) the downwind deposition (soil and vegetation), and (3) the airborne concentration downwind. Only ARPS and the model of (Hong et al., 2018a,b) predict these three variables, in the case of vineyard and orchard treatments respectively. Deposition is generally computed only a few tens of metres downwind from the treated field, except in the case of the Silsoe Spray drift model or AGDISP (Nsibande et al., 2015) that explores distances up to 400 m (Butler Ellis et al., 2017) or up to 500 m for the Thompson and Ley (1983) model. The evaluation of aerial drift is often done at a single distance downstream of the treated plot because few data are available. Note that IDEFICS calculates airborne concentrations at a distance close to the treated plot (typically 5m) and must be coupled with an atmospheric dispersion model such as OPS (Van Jaarsveld, 2004) to predict concentrations further away.

## **V.6. Overall performance of existing models**

The performance of the models may depend on considered output (airborne concentrations vs sedimentary deposition) and on distances or height of the treated crop as noticed for example by Butler Ellis and Miller (2017) who found a good correlation at 20 m between predicted and measured airborne spray for a short crop, but an overestimation of the airborne concentration for a crop of 0.7 m height, whereas sedimentary deposition was generally well predicted for all crop heights (with however a small over-prediction at large distances and underprediction at short distances).

IDEFICS, ARPS, Silsoe spray drift and RTDrift were evaluated regarding sedimentary drift, at short distances up to 5 m and 7 m for IDEFICS and ARPS respectively and at longer distances for Silsoe spray drift and RTDrift, up to 20 m and 30 m, respectively. ARPS overestimated the outfield deposition by a factor of  $\sim 4$ , while IDEFICS modelled deposition quite well next to the field with a tendency to increasingly overestimate drift with distance. RTDrift tended to underestimate outfield sedimentary drift from 15 to 30 m, while the Silsoe spray drift model overestimated it at distances greater than 8 m and underestimated it at 2 m (up to 3 times). The evaluation of AgDRIFT in the case of aerial applications, showed systematic underestimation at distances smaller than 60 m and overestimations at distances larger than 90 m. AgDISP and OML-spray-drift were evaluated against airborne concentrations at different distances and heights, where AgDISP validations showed two orders of magnitude variation between modelled results and measured data. OML-spray-drift underestimate airborne at 4m next to the boom sprayer and overestimate it for further distances (up to 36m from the boom). In addition, authors indicate that the model is not suitable for predicting airborne and drift while spraying over canopy cover as it does not consider droplets impaction on canopy and the effect of canopy on the atmospheric turbulence. Silsoe spray drift model have also been evaluated against airborne measurement data.

Results showed an overestimation of airborne farther from sprayer, which were identified by the authors to be related to the non-consideration of canopy interception. As the model of (Hong et al., 2018a,b) focuses more on spraying efficiency and the repartition of spray deposition inside the canopy, outfield deposition is poorly estimated with an increasing overestimation with distance from the sprayer. For canopy interception, models prediction are acceptable with an average relative error equal to 22.1%. Thompson and Ley (1983) have not evaluated their model as they estimated that the random walk models have been already proven their efficiency regarding particles dispersion.

### **V.7. Recommendations to improve pesticide spray drift modelling**

This synthesis allows identifying avenues for improvement of models dedicated to pesticide losses to the environment at the time of application. This involves taking into account processes that are currently neglected, application technics or contexts not yet implemented, as well as acquire new datasets:

In models, it is key to improve the description of the droplet characteristics by, including atomisation in models, although this is a complex process. A useful alternative would be to improve knowledge on emitted droplets. Indeed, they are one of the main drivers of pesticide spray drift, including the finest particles that are still poorly characterised even if they are recognised to contribute significantly to population exposure by inhalation. The effect of canopy characteristics on the local atmospheric turbulence and its filtering capacity affect significantly the initial distribution of pesticides between the crop, the soil and the atmosphere, and thus losses by spray drift. Accordingly, improvement in describing the effect of treated canopy structure, density and horizontal organisation on the airflow and on the droplet interception are one of the key points for improving pesticide spray drift, especially in perennial crops. It is also necessary to take more systematically into account atmospheric stability effects on droplet transport, and consider specific conditions such as crop row orientation related to wind direction. The effect of obstacles, like hedges, on the local turbulence and thus on spray drift is important, and the evaluation of these obstacles as a lever for reducing pesticide transfer to non-targeted require models to account for them. Taking into account adjuvants effects on droplet size distribution and considering pesticide volatilisation from the droplet would complete the risk assessment linked with the active ingredient dispersion and population or non-targeted ecosystems exposure.

To limit pesticide spray drift, several solutions have been identified: they were based on specific materials (nozzles, sprayer types such as tunnel sprayer) or application strategies within the field (last row not sprayed, ...). Modelling tools should therefore be able to represent these configurations to allow their efficiency to be evaluated. Furthermore, a generic model that is adapted to annual and perennial crops would be very useful. A generic model may be time and resource consuming, and it may therefore also be useful to develop emulators as suggested by Butler Ellis et al. (2017), to enable easy multi-



simulations. Recent progress in computing capacity may however allow using directly complex models on wider spatio-temporal scales.

Finally, it remains essential to acquire datasets, both to provide input data for the model (characteristics of emitted drops, of the treated canopy), but also of course to analyse the model's performance. Although datasets exist for sedimentary drift, it is necessary to ensure that they are representative for the current and new application conditions and, if necessary, to complete them. The mass balance at application between the treated surface, the soil and the air remains poorly documented and few data have been acquired on airborne drift, namely droplet atmospheric concentration downwind from the source. It is therefore essential to strengthen our knowledge on these last two aspects. As far as airborne drift is concerned, it is important to consider dispersion heights and distances adapted to the crops treated. Another point to stress is the fact that experiments are usually conducted with dyes in place of commercial products. Experiments should instead be carried out with commercial products to check the representativeness of existing datasets and models. This is especially key since we know that dye content (around 20% of the mixture) is much higher than pesticide content (0.02% - 2%). We have stressed in the document that, consequently, the droplet size after evaporation may be several order of magnitude smaller with commercial product than with a dye. Last but not at all the least, measurements should be made by actively sampling the air, since passive sampling does not catch the smallest particles. Moreover, the range of measured diameter should extend to sub-micron ranges to measure droplet that have undergone evaporation.

Integrating consistently all processes involved in pesticide transport and fate in the environment over larger spatial and temporal scales is the way towards a better assessment of pesticides effects on health of humans and all living organisms. A good description of the pesticide fate over the first seconds following application is key in this integration as it determines the repartition between targeted surfaces, soil, the immediate surrounding and the atmosphere. This review shows that there remain large uncertainties in this initial dispersion process, that require efforts to be made on model improvements but also on field measurements over real system.





## Chapitre 3

### *Modélisation de la dérive de gouttelettes et de la distribution des pesticides lors de l'application*

*Suite à la revue bibliographique présentée dans le chapitre précédent, des lacunes en termes de modélisation de la dérive en contexte viticole mais aussi de la distribution initiale de PPP lors de l'application ont été soulignées. Ainsi un nouveau modèle 3D type marche aléatoire « ADDI-Spray Drift » de dérive de PPP et de leur distribution lors de l'application entre le sol, la végétation et ce qui sort de la parcelle a été développé pour répondre à ces lacunes. Ce chapitre présente le modèle développé ainsi que les approches de modélisation entreprises pour représenter les principaux processus impliqués dans la dérive et la distribution des PPP lors de l'application. Les différentes sorties du modèle et sa sensibilité vis-à-vis de huit principaux paramètres sont également présentées. Enfin, une confrontation du modèle aux données expérimentales de dérive sédimentaire de Ganzelmeier et al. (1995) est établie. Ce chapitre est présenté sous la forme d'un article soumis au journal Biosystems Engineering et qui est en révision.*



## ADDI-SprayDrift: A COMPREHENSIVE MODEL OF PESTICIDE SPRAY DRIFT WITH AN ASSESSMENT IN VINEYARDS

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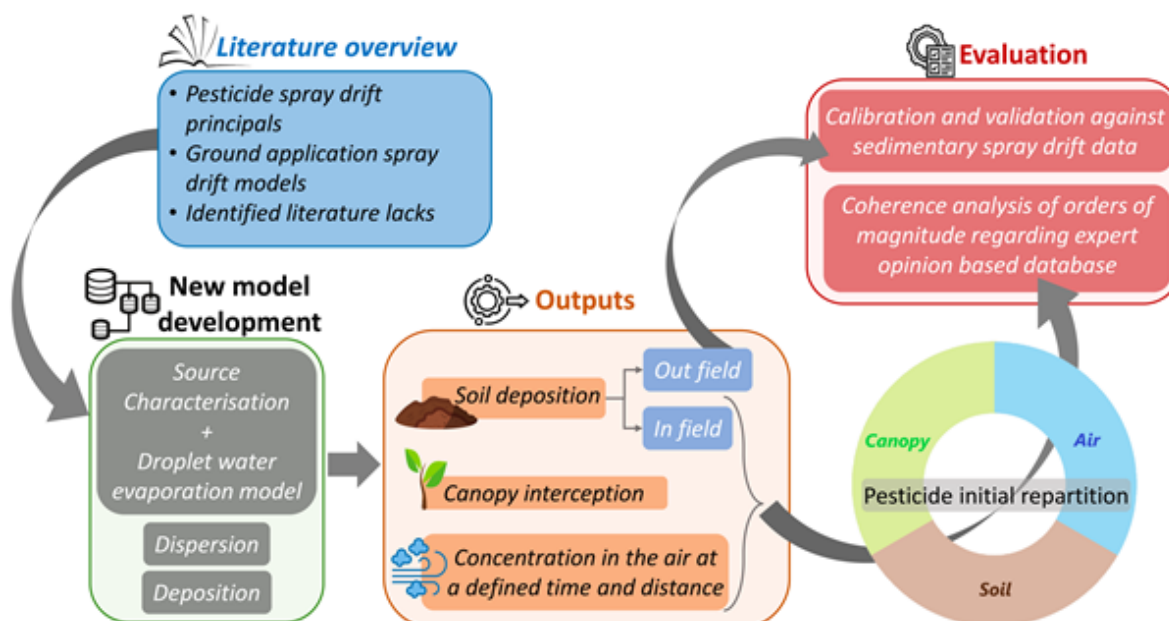
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### Graphical abstract:



### Highlights:

- The ADDI model is developed to estimate spray drift and crop interception for ground sprayers.
- ADDI is based on a random walk approach integrating spray drift key processes.
- Model predictions are highly sensitive to droplet ejection characteristics.
- Model predictions well represent the Ganzelmeier drift experiments on vineyards.
- ADDI is expected to be valuable for impact studies of spray drift for ground sprayers.

**ABSTRACT:**

Spray drift is a major contributor to losses in the atmosphere leading to nontargeted ecosystem exposure to pesticides. An overview of currently used ground-application spray drift models highlighted gaps in the description of the processes occurring during and after spraying. The ADDI-SprayDrift 3D model was developed to bridge some of the gaps and provide a comprehensive, yet parsimonious, description of the spray drift processes. The model is based on a random walk approach that describes droplet emission, dispersion and evaporation, ground deposition and canopy interception, and accounts for atmospheric stability regimes, and in-canopy turbulence. It predicts airborne concentration, canopy interception and deposition to the ground downwind from the sprayer. The sensitivity analysis to wind speed, active matter content, leaf angle, canopy height, leaf area index, ejection angle and velocity demonstrated the consistency of the model behaviour. The model was calibrated and evaluated against Ganzelmeier et al. (1995) sedimentary spray drift data in vineyard. The model satisfactorily predicts droplets deposition to the ground downwind from the field boundary with a mean deviation between modelled and measured deposition of 1.3%. A discrepancy is observed at 3 and 5 m downwind from the field boundary attributed to the sensitivity to spraying conditions for which several hypotheses are discussed. Bearing in mind the need to explicitly describe the sprayed droplet size and speed, the overall predictive performance of the model appeared to be sufficient for assessing and comparing application techniques efficiency, quantifying pesticide losses and bystanders or residents' exposure and evaluating mitigation measures' efficiency.

**Keywords:** Atmospheric dispersion, Lagrangian, ground application, atmospheric turbulence, deposition, droplet canopy interception.

## **I. Introduction**

Since the advent of intensive agriculture and the massive use of pesticides, soil and vegetation in agricultural areas have been highly contaminated by these products (Leenhardt et al., 2022). Moreover, many analyses in aquatic environments and later in the atmosphere have revealed the presence of pesticides even outside the treatment areas in air, water, soil and living organisms in adjacent environments (Aubertot et al., 2005; Hulin et al., 2021). The presence of pesticides in non-targeted areas demonstrates the propensity of pesticides to be dispersed in the environment that does not only lead to economic loss for the farmers and a reduction of their effectiveness against pests and to risks to the environment (Tiryaki & Temur, 2010; Waite, Cessna, Grover, Kerr, & Snihura, 2002) and to public health (Dereumeaux et al., 2020). Among the different transfer pathways of pesticides to nontargeted areas, spray drift is a major one, contributing to losses ranging from 20 to 60% of the applied amount of pesticides at the field scale (Hong et al., 2021; Zhang, Tang, Chen, Zhang, & Xu, 2018). Spray drift is defined as the fraction of pesticide droplets that does not reach the target and moves beyond it through atmospheric transport at the time of the application (ISO 22866, cited by Moltó et al., 2017). The contribution of atmospheric dispersion including spray drift and dispersion of the fraction of pesticide being volatilized in post-application to the contamination of non-targeted areas is raising worldwide concerns (Inserm, 2021; Moltó et al., 2017), especially regarding the toxicological and ecotoxicological risks to all living organisms (including humans) and the functioning of ecosystems and their resilience (Hallmann et al., 2017). This contribution to contamination is particularly true in vineyard areas. A recent study in France showed contamination of ambient air up to more than 20 ng/m<sup>3</sup> during the treatment periods near schools located close to vineyards and suggested possible links between the pesticide levels in air and respiratory and allergic symptoms such as rhinitis (Raherison et al., 2019). Indeed, the use of plant protection products is high in vineyards, much higher than that for field crops, and in France it corresponds to 20% of the total pesticide consumption for only 3% of the agricultural area (Aubertot et al., 2005). Moreover, the type of sprayers used in vineyard are particularly prone to spray drift losses (Warneke et al., 2021). In this context, improving the means for the evaluation of spray drift contributions to ecosystem contamination and residents or bystander exposure is a major challenge, particularly in vineyard areas.

The assessment and quantification of spray drift require a good understanding of the drift phenomenon and of the underlying processes (Özkara et al., 2016). This is also essential for identifying relevant mitigation actions. Many researchers have conducted laboratory or field experiments to address this issue, or have used wind tunnels (Al Heidary et al., 2014) to represent the potential spray drift of nozzles. However, due to the complexity of the processes involved in spray drift, the diversity of cases and scales (crop diversity, field characteristics, machinery, etc.), the challenging measurement conditions, and the high cost and time consumption, experiment-based assessment of drift has been

proven to be difficult (Hong et al., 2021). By contrast, computational models of spray drift may simulate a variety of situations. They are useful not only for the study of peculiar agricultural conditions (Salcedo et al., 2017; Nsibande et al., 2015) but also for regulatory purposes, as well as for the evaluation of treatment efficiency. Models can be used to estimate the exposure of nontarget organisms and of nearby residents and to assess the efficiency of spraying materials and mitigation actions (material used, buffer zones), for the different crops. Ideally, to allow such global evaluation, these models should be able to predict: (1) droplets deposits at different distances, (2) droplet concentrations in the atmosphere at different heights and distances, and (3) the fractions of the spray that is deposited to the canopy and to the ground within the treated field. These models should hence account for a range of key processes and their driving factors. Key processes are droplet emission from the sprayer, atmospheric dispersion, droplet evaporation, and deposition onto the canopy and onto the ground, whereas driving factors are (1) droplet emission characteristics (ejection and angle of ejection, droplet size distribution), (2) atmospheric conditions (wind speed, temperature, relative humidity, stability), and (3) crop characteristics (height, LAI, typical leaf size, canopy thickness) (Hong et al., 2021). Various models have been developed so far but, depending on their specific aims and application domains, they do not necessarily consider all the above processes and driving factors, their outputs may differ, and they are often devoted to one crop type (annual or perennial) and its associated sprayers (boom sprayers for the first, pneumatic/axial fan sprayers with or without air assistance for the latter).

In most models, particle dispersion in the atmosphere has been described based on a Lagrangian approach that simulates the individual trajectories of a group of particles that evolve over time along their path (FOCUS, 2008; Jarosz, 2003) under the effect of aerodynamic, inertial, and gravity forces (Urip, Yang, & Marek (2002) in Nuyttens 2007). Regarding pesticide spray drift, the Lagrangian approach was used in IDEFICS (Holterman, 2003; Holterman et al., 1997), AGDISP (Nsibande et al., 2015; Teske et al., 2009), ARPS (Chahine, 2011; Chahine et al., 2014; Dupont & Brunet, 2008a, 2008b), the Silsoe Spray Drift model - SSMD (Butler Ellis & Miller, 2010) or the model of Thompson & Ley (1983). Indeed, the Lagrangian approach has a strong ability to consider complex atmospheric conditions (heterogeneous turbulence, stability), precise source characteristics (particle size distribution, ejection height, angle and speed), but also the evaporation process. Moreover, it can reproduce near-field diffusion that does not follow standard diffusion laws and is therefore appropriate for in-canopy dispersion (Loubet et al., 2006). Other approaches exist, such as Gaussian models that are based on an analytical solution of the advection-diffusion approach (e.g. in OML-spray drift model (Lofstrom et al., 2013)), and the Eulerian models that are based on the resolution of the mass conservation equation in a turbulent flow (they often also include the Navier-Stokes equation). These latter approaches are used but they seem to be less appropriate than the Lagrangian one for predicting both infield deposition/interception and outfield deposition and airborne concentration (Craig, 2004). Moreover, the precision of Eulerian models depends on the defined grid cell size of the computational domain

(FOCUS, 2008), which can result in computationally expensive simulations and restricted spatial computational domains (Hong et al., 2021). There are also so-called mixed modelling approaches where the particle trajectory is calculated by means of a Lagrangian model while the atmospheric turbulence is described by an Eulerian model such as Computational Fluid Dynamics models (CFD) or Large Eddy Simulation models (LES). CFD models are particularly adapted for representing airflow through complex configurations but at small spatial scales, while LES models provide turbulence at medium scales. In their spray drift model, Hong, Zhao, & Zhu (2018a) used a CFD model to represent airflow in an apple orchard, and more recently, in vineyards (Hong, Zhao, & Zhu, 2018b). Chahine (2011) used an LES model in ARPS to describe airflow in vineyards. However, CFD or LES approaches require enormous computational resources (Hong et al., 2021) and are thus, until now, applied on rather small spatial domain (few tens of m). Moreover, accounting for stability conditions in CFD and LES models requires even more computational power and complexifies their parameterisation (Hong et al., 2021). Atmospheric stability conditions, indeed, strongly influence dispersion (Miller et al., 2000; Thompson & Ley, 1983; Hong et al., 2021), and affect especially greater distance concentration and deposition, and thus smaller droplets (Hong et al., 2021). They are however not always taken into account in pesticide spray drift models, *e.g.* only neutral conditions are considered in LES and CFD models, ARPS, Hong et al. (2018a) or Duga et al. (2017), but also in AGDISP.

Spray interception by the canopy and canopy effects on turbulence have both been shown to be essential processes for simulating drift, especially when crops are tall (Thompson & Ley, 1983). Accordingly, the models that deal with tall crops, like ARPS and the models of Hong et al. (2018a) and Duga et al. (2017), are describing both processes. Considering treatments on short crops, IDEFICS and SSMD also represent the two processes. Some models account for a detailed plant architecture to study the spray interception by the targeted canopy, but they do not predict the drift downwind (*e.g.* Dorr et al. (2008 ; 2016) or Endalew et al. (2010)).

Droplet water evaporation is usually taken into account in the models. It was neglected in ARPS and that assumption was identified by the authors as the source of ground deposition overestimation by ARPS when compared to measurements.

Eventually, it must be pointed out that most of process-based models are devoted to annual crops and boom sprayers. To the best of our knowledge, the only models developed for perennial crops are ARPS, that was developed for vineyards, and the models of Hong et al. (2018a, b) or Duga et al. (2017) developed for orchards with an application in vineyard in Hong et al. (2018b).

This overview of existing models thus showed an interest in developing a modelling approach for high crops that represents all processes and outputs assumed to be essential for studying spray drift, but that is not too complex and time-consuming in order to be applicable on spatial domains with multiple treated plots for studying ecosystem or population exposures.

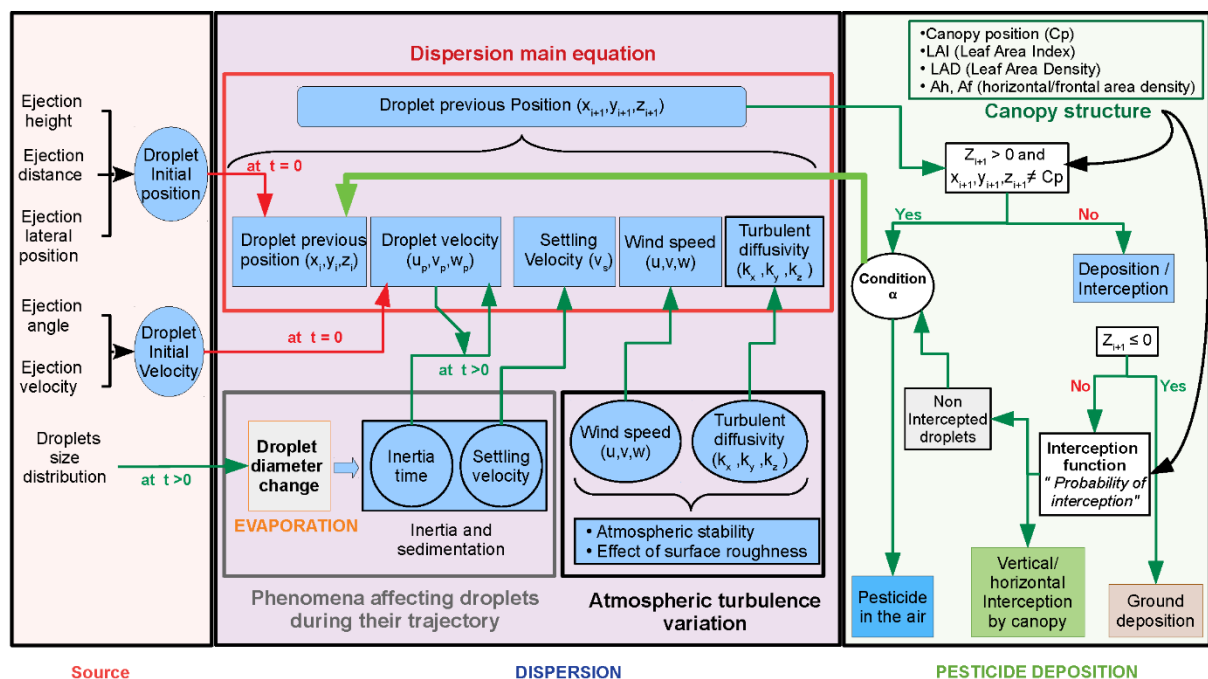


In this paper, we present and evaluate a new ground-application process-based model, ADDI-SprayDrift. The model is intended to be coupled later on with models describing pesticide dissipation in the environment at the landscape scale, and was therefore developed so to remain parsimonious in processes and parameters. The model is evaluated here for an application in a vineyard. The model predicts drift in addition to interception by the targeted vegetation and addresses outfield dispersion and deposition in the very short (< 20 m) and medium (< 500 m) ranges. This is required to estimate contamination of nearby surface waters (rivers or ditches), but also resident exposure. Below, we first present ADDI-SprayDrift and the methods used for its evaluation in detail. Then, we illustrate the behaviour of the model in an application, describe the results of its sensitivity analysis and finally proceed to the first evaluation of the model performance in the simulation of the experimental sedimentary drift data from Ganzelmeier et al. (1995) in vineyards.

## II. Description of the ADDI-SprayDrift model

The new pesticide spray drift model, called the ADDI-SprayDrift model (*Atmospheric Dispersion, Deposition and Interception of Spray Drift*), is subdivided into three components (Fig. III.1). The source component gathers all of the parameters and equations to describe the droplet size, velocity and pesticide content distribution at application. The dispersion component groups together the processes of particle dispersion and all of the phenomena influencing the trajectory of the particles, such as droplet evaporation and atmospheric turbulence. The deposition component describes particle deposition onto the ground and the canopy. In this section, we present in detail the chosen approaches and the operation of the model.

Figure III. 1. Scheme of the model architecture, composition and functioning



## II.1. Droplets emission

The source is modelled as a set of point sources spatially located to represent droplet emission by the sprayer and its nozzles. Each point source represents a single nozzle characterised by its height, location (x, y, z), ejection angle and velocity, and droplet size distribution. These parameters are defined based on the spraying equipment and pesticide used. The pesticide droplet emission model does not consider atomisation and thus the initial phase of droplet formation close to the nozzles. This is considered to be too complex for our purpose. We therefore assume that the diameter, ejection velocity, height and angle of the droplets are known.

Droplets are assumed to be spherical with a log-normal diameter distribution (Eq. (1)), which fits the actual distribution for different types of sprayers and has been identified by Paloposki, (1994) as the most mathematically stable distribution (Déchelette et al., 2011) :

$$f_0(D) = \frac{1}{D (\ln \sigma_{LN}) \sqrt{2\pi}} \exp \left\{ -\frac{1}{2} \left[ \frac{\ln(D/\bar{D})}{\ln \sigma_{LN}} \right]^2 \right\} \quad (1)$$

where  $\bar{D}$  is the logarithmic mean diameter of the droplets ( $\mu\text{m}$ ) and  $\sigma_{LN}$  is their standard deviation ( $\mu\text{m}$ ). These two parameters are defined according to the spraying device used. The droplet initial velocity components  $u_{p_0}$ ,  $v_{p_0}$ ,  $w_{p_0}$  depend on the type of material (atomization device and presence of air driver) and are computed considering the ejection velocity  $v_e$  ( $\text{m s}^{-1}$ ) and the ejection angle  $\alpha_e$  ( $^\circ$ ):  $u_{p_0} = v_e \times \sin(\alpha_e \pi/180)$  and  $w_{p_0} = -v_e \times \cos(\alpha_e \pi/180)$ , while  $v_{p_0} = 0$ . While using the model, the set of parameters describing the source must be adequately given according to existing experimental data for the concerned sprayer. As the first step and the first application of the model is in the case of vineyards, where an air jet is usually performed, no fluid entrainment is considered as opposed to boom sprayer models (e.g., Holterman et al. (1997) and Butler Ellis & Miller (2010) models).

## II.2. Droplets evaporation

We chose the model of Trayford & Welch (1977), where the droplet diameter changes over time following Eq. (2) (Teske et al., 2011, 2016, 2018). This model assumes that only water evaporates from the droplets and that when all water is evaporated, the droplet diameter reaches the minimum value corresponding to a particle containing only the active ingredient (all water and solvent are assumed to have evaporated). The effects of ambient temperature, relative humidity and atmospheric pressure are integrated using the wet-bulb depression concept (Teske et al., 2016). Furthermore, this model calculates a prescribed droplet evaporation time and diameter evolution based on initial conditions. The droplet diameter  $D$  (m) is obtained from a differential equation (Annexe A Eq.A1):

$$D = D_0 \left(1 - \frac{t}{\tau_e}\right)^{1/2} \quad (2)$$

where  $D_0$  is the initial droplet diameter ( $\mu\text{m}$ ) and

$\tau_e$  is the evaporation time. Here,

$$\tau_e = \frac{2 D_0^2}{\lambda \Delta\theta Sh} \quad (3)$$

where  $\lambda$  is the evaporation rate ( $\mu\text{m}^2 \text{s}^{-1}$ ),  $\Delta\theta$  is the wet-bulb temperature depression ( $^\circ\text{C}$ ) (Annexe A Eqs. A2 and A3, respectively) and  $Sh = 2 (1 + 0.27 R_e^{1/2})$  is the Sherwood number (dimensionless), where  $R_e = \frac{D_0 U}{\nu}$  is the Reynolds number,  $U$  is the relative velocity of the droplet with respect to the surrounding air ( $\text{m s}^{-1}$ ) and  $\nu$  is the kinematic viscosity of the air ( $\text{m}^2 \text{s}^{-1}$ ) calculated as a function of temperature (see Eqs. A5 and A6 in Annexe A, respectively). The initial diameter is set based on the distribution in Eq. (1). The results of the evaporation model compared well with those obtained by Bahrouni (2010) applying the Ranz & Marshall (1952a, b) evaporation model included in IDEFICS spray drift model (Figs. S.A.1.a and b in Supplementary Materials).

### II.3. Atmospheric dispersion

We chose a random walk dispersion model that allows the evaluation of spray drift at short- and medium-range distances and remains parsimonious. This model does not explicitly describe the inertia of the particles during their travel. However, inertia is accounted for “on average” by reducing the particle diffusivity by a factor  $k_p$  relative to the gas diffusivity, as in Bouvet & Wilson, (2006). This model allows an explicit description of the droplet size over a wide range (Craig, 2004) and explicitly includes the effect of evaporation on the droplet diameter of each individual droplet over its travelling time (Nuyttens, 2007). The model describes the motion of each droplet at each time step as influenced by the mean wind speed components  $u, v, w$  ( $\text{m s}^{-1}$ ) and the turbulent diffusivity components  $K_x, K_y, K_z$  ( $\text{m}^2 \text{s}^{-1}$ ), according to the principle of the Markov chain-relationship (Hall, 1975; Hashem & Parkin, 1991). Individual droplet trajectories are represented by a series of discrete displacements that are randomly selected from the appropriate Gaussian distributions of turbulent air velocity, where the prediction of the future droplet trajectory depends only on the current velocity of the droplet and not on its previous velocity (Thompson & Ley, 1983). Thousands of particles are selected at random according to the probability distributions of diameter and initial velocity. The position of a particle at time  $t_i$ ,  $(x_i, y_i, z_i)$ , where  $x_i$  is the horizontal,  $y_i$  is the crosswind and  $z_i$  is the vertical position (m), is governed by the following equation:

$$\left. \begin{aligned} x_{i+1} &= x_i + (u + u_p) \times dt + (2k_p K_x)^{1/2} \times \xi_{u_{i+1}} \\ y_{i+1} &= y_i + (v + v_p) \times dt + (2k_p K_y)^{1/2} \times \xi_{v_{i+1}} \\ z_{i+1} &= z_i + (w + w_p - v_s) \times dt + (2k_p K_z)^{1/2} \times \eta_{i+1} \end{aligned} \right\} \quad (4)$$

where  $dt$  is the time step (s);  $u, v, w$  are the horizontal, crosswind and vertical wind speeds ( $\text{m s}^{-1}$ ), respectively;  $u_p, v_p, w_p$  are the horizontal, crosswind and vertical transient particle velocities following ejection ( $\text{m s}^{-1}$ ), respectively;  $x_{i+1}, y_{i+1}, z_{i+1}$  are the particle horizontal, crosswind and vertical positions at time  $t+1$ , respectively ;  $v_s$  is the particle settling velocity ( $\text{m s}^{-1}$ );  $K_x, K_y, K_z$  are the horizontal, crosswind and vertical turbulent diffusivities ( $\text{m}^2 \text{ s}^{-1}$ ), respectively; and  $\xi_{u_{i+1}}, \xi_{v_{i+1}}, \eta_{i+1}$  are random numbers belonging to a Gaussian distribution with a mean equal to zero and a unitary standard deviation.

The effect of droplet ejection into the atmosphere is embedded in the random walk model as a transient particle velocity relative to the fluid velocity  $u_p, v_p, w_p$  ( $\text{m s}^{-1}$ ). This velocity is equal to the ejection velocity at  $t = 0$  and decreases according to the Stokes' law towards zero as  $u_p = u_{p0} \exp(-t/T_s)$ , and similarly for  $v_p, w_p$ . Here  $T_s$  (s) is the Stokes time accounting for the inertia of the particle. The inertial effect due to injection is thus superimposed onto the random walk equation (Eq. 4) as a pure drift term. This is justified if the jet effect on the turbulence is neglected. The Stokes number calculation is described in detail in Annexe A (see Eqs. A8 and A9).

## II.4. Wind speed and turbulence profiles

In our approach, for simplification, the turbulent characteristics are considered to be homogeneous in the x and y directions and the effect of air jet on the local turbulence is neglected. For the description of atmospheric stability conditions, we used the well-established Monin–Obukhov atmospheric surface layer parameterisation that allows the simulation of dispersion in various atmospheric stability conditions near the surface (Kaimal & Finnigan, 1994). The mean wind speed and turbulence profiles are described in detail in Annexe A (Eqs. A10 to A28). Therefore, the turbulence homogeneity assumption does not account for the roughness change at the edge of the treated field. This roughness change is expected to lead to a zone of increased turbulent mixing and downwards wind speed that may increase the droplet deposition at the field edge as well as mixing in the boundary layer. The limitations concerning both the homogeneity of the turbulence and the effect of air jet can be overcome in an upcoming development of the model by coupling the model with a more complex turbulence model or through an alternative approach by parameterising the roughness change using spline interpolation of turbulence profiles, as in Jarosz et al. (2004). Anyway, Bouvet et al. (2006) showed that accounting for a windbreak flow disturbance has a limited effect on heavy particle deposition. We could hence consider that this effect is of second order.

## II.5. Droplet deposition

The deposition model takes into consideration the fractions intercepted by the plant elements and deposited onto the ground. For ground deposition, all particles that reach the surface ( $z_i = 0$ ) are considered to be deposited with the assumption that bounce-off does not occur for pesticide droplets. Within the Lagrangian stochastic framework, a statistical approach is introduced to determine whether a particle is deposited due to interception by a plant canopy (e.g., Aylor (2005)). In Aylor (2005), this interception probability ( $S_p$ ) is calculated using two probabilities, namely the probability of sedimentation on the horizontal surface and the probability of deposition by inertial impaction on the vertical surface. This equation was developed by Aylor (2005) for simulating pollen interception and is used in our model but with an additional term to account for impaction over horizontal leaf surfaces (last term of Eq. (5)). Indeed, pesticide particles can be ejected with a large vertical velocity, which is in contrast to pollens or spores that are essentially transported by wind.

$$S_p = Ah v_s dt + Af Ei \sqrt{\langle u + u_p \rangle^2 + \langle v + v_p \rangle^2} dt + Ah Ei (w + w_p) dt \quad (5)$$

where  $S_p$  is the probability of pesticide droplet deposition into the canopy and  $Ah$  and  $Af$  are the horizontal and frontal leaf area densities (LAD), respectively. These areas result from the horizontal and vertical projection of the canopy elements, and  $Ei$  is the impaction efficiency given by (Aylor, 1982; Aylor & Flesch, 2001; May & Clifford, 1967) and explained in Annexe A (Eq. A29). To determine whether a pesticide droplet is intercepted by the canopy over time,  $S_p$  is compared to a random number  $\eta$  drawn from a uniform distribution between 0 and 1. The droplet is considered to be deposited onto the canopy if  $\eta < S_p$ . We note that the current model is limited to particles larger than  $\sim 1 \mu\text{m}$  because no Brownian diffusivity is taken into account, whereas this process may be an efficient deposition process for small particles (Reynolds, 1999).

## II.6. Model implementation

The developed model is a 3D model coded in R (version 4.1.1, (R Core Team, 2021)) with a time step set to 0.01 s that is much smaller than the Lagrangian turbulence time scale. Decreasing the time step to the 5<sup>th</sup> percentile of the Stokes time is recommended when dealing with smaller droplets or higher ejection speeds. The simulation domain can be adjusted according to the study requirements. For this, a stop condition can be set for the x, y and z dimensions (represented by the component “*Condition a*” in Fig.III.1). In this study, since we are seeking to determine the extent of spray drift, no spatial limit is defined; however, for simplicity, a mesh of 10 cm is set for representing the concentrations. A detailed list of the model’s inputs and outputs is given in the supplementary material (Table S.A.1). It is important

to note that calculation time depends on the number of released particles and the simulated configuration. For example, the simulation of spray drift from one nozzle with 20000 particles for all simulated configurations took on average 13 min by simulation on a dual-processor computer (Intel Xeon Gold 5120 2.2 3.7 GHz Turbo 25MB, 10 cores, hyper-threading activated and 256 GB of RAM). The simulations have been launched simultaneously on the available cores in order to optimized computing time.

### **III. Case study and model evaluation**

#### **III.1. Case study**

To evaluate the capability of our model to represent actual drift processes, we used the experimental setup and results of Ganzelmeier et al. (1995) as a benchmark. The Ganzelmeier dataset was chosen because it is an average of 11 trials set up on grapevines in a late growth stage and thereby encompasses the limitation of using individual trials whose wide variations can make model validation difficult (Holterman et al., 1997; Butler Ellis & Miller, 2010). Moreover, Ganzelmeier dataset is used in the European Union for deriving drift tables (Rautmann et al., 2001) that serve in the pesticide- exposure assessment of operators, workers, residents and bystanders (EFSA, 2014) and is therefore recognised as a reference dataset to represent drift deposits. Each Ganzelmeier trial consisted of an application of a copper formulation by an axial fan sprayer delivering an application volume of 400 to 600 l ha<sup>-1</sup> at driving speeds between 6 and 6.1 km h<sup>-1</sup> and pressures between 8 and 8.5 bar. For each trial, spraying was performed when the wind direction was perpendicular to the rows and was limited to the six vine rows adjacent to the field edge along the wind direction. According to Ganzelmeier et al. (1995), these rows were chosen to represent the maximum area from the field edge that contributes to the direct spray drift. For the first row, only one row side was treated (opposite side to wind direction), whereas for the other rows, the two sides were treated (Fig. S.A.2 in Supplementary Materials and Fig. 1 in Ganzelmeier et al. (1995)). The deposited drift material was observed beyond the field edge at seven distances, namely, at 3, 5, 7.5, 10, 15, 20, and 30 m, with at least 5 replicates per distance. Since all 11 trials are very close in terms of spraying and weather conditions (Table 4 in Ganzelmeier et al. (1995)), we use the mean values of temperature, air relative humidity, wind speed and dosage over all trials that are equal to 20.1°C, 59%, and 2.8 m s<sup>-1</sup> at a height of 3 m and 0.4%, respectively. In the absence of information concerning the atmospheric thermal stability condition, this was set as neutral. The average deposited drift material of the late growth stage and the standard deviations of the mean values are taken from Table 13 of Ganzelmeier et al. (1995).

### **III.2. Model setup to compare to the case study**

Model parameters that were not explicitly given by Ganzelmeier et al. (1995) were set based on the available information in the report of Ganzelmeier, the literature, and expert knowledge of canopy structure and droplet ejection velocity and size distributions. The model input parameters are compiled in Table III.1. A plantation density of approximately 4,000 plants ha<sup>-1</sup> was assumed. Leaves were assumed to be present only at the heights above 0.6 m above ground, as observed by (Hebrard, 2004). The LAI of the grapevine was considered to potentially vary between 12.5 and 15 (Sinfort, 2006) corresponding to a late growth stage. The spraying characteristics were assumed to be those of an axial fan sprayer, with 6 nozzles as described in the literature as a mid-range of existing practices (Gil & Badiola, 2007; Mion et al. 2012). For simplicity, all nozzles were positioned 0.5 m from the vine row edges at mid-height of vine foliage, and their vertical ejection angles were chosen to vary from 45° to 135° with an increment of 15° (Pergher, 2004). The droplet diameter was assumed to follow a log-normal distribution with a mean diameter of 100 µm and a standard deviation of 50 µm, which is within the range of reported values of 75-300 µm (Gil 2007; De Luca 2007) (Fig. S.A.3). The droplet ejection velocity was considered in the typical range of an axial fan sprayer of 30-65 m s<sup>-1</sup> (Duga et al., 2015), the leaf width was between 5 and 7 cm (Eftekhari et al., 2011) and the inclination angle varied from 45° to 90° (Mabrouk, et al., 1997). The field was a set of 6 vine rows perpendicular to the wind direction. To illustrate the model behaviour, we considered two source-spraying configurations: a point source positioned at mid-height of the vine leaves wall and spraying at six different angles (Table III.1) on the upwind side of either the second row (ROW-2) or the 6<sup>th</sup> row (ROW-6) (Fig. S.A.2).



Table III. 1. Parameters used for model behaviour illustration, sensitivity analyses and calibration

Model parameters	Measured or plausible range <sup>#</sup> of variation in parameter values in the Ganzelmeier trials	Fixed values for ROW-2 and ROW-6 simulations	Values considered in the sensitivity analysis
LAI <sup>a</sup> vineyard (-)	15 <sup>#</sup>	15	7.5, 10, 12.5, 15
Row spacing (m)	2.5 <sup>#</sup>	2.5	NC
Canopy height (m)	2 <sup>#</sup>	2	0.8, 1.2, 1.6, 2.0
Leaves starting height (m)	0.6 <sup>#</sup>	0.6	NC
Canopy width	1 <sup>#</sup>	1	NC
Leaf width (m)	0.07 <sup>#</sup>	0.07	0.05, 0.07, 0.09, 0.11
Leaf inclination angle (°)	45 <sup>#</sup>	45	45, 60, 75, 90
Ejection angle (°)	40, 55, 70, 85, 100, 115 <sup>#(b)</sup>	40, 55, 70, 85, 100, 115	45, 75, 105, 135
Ejection velocity (m s <sup>-1</sup> )	60 <sup>#(b)</sup>	55	35, 50, 65, 80
Active substance proportion (%)	0.4	2	0.02, 0.09, 0.43, 2
Wind velocity at 3 m height (m s <sup>-1</sup> )	2.8	2.8	0.5, 2.7, 4.8, 7.0

### III.3. Model sensitivity analysis

To determine the model response to key parameters, we set up a sensitivity analysis based on the Ganzelmeier case study. The sensitivity analysis was performed on the eight main parameters (Table III.1) and the four main output variables. Wind velocity was used as a proxy for all turbulent parameters. The four chosen output variables are infield and outfield pesticide deposition on the ground, pesticide infield interception by the canopy and the maximal deposition distance. Deposition is expressed as the ratio to the total applied mass of the pesticide and the maximal deposition distance at which 95% of the applied mass was deposited.

The sensitivity analysis was performed using the Morris method which is a "One at a time" (OAT) method, where only one parameter is changed at each simulation to assess the elementary effect of each factor on the outputs (Faivre et al., 2013). It is known as an effective screening method to identify the most important parameters in large models with only a few simulations (e.g., Campolongo et al., 2007). Here, we test the sensitivity of the model to 8 parameters, considering 4 levels per parameters (see Table III.1) and 20 repetitions which has been shown to be enough to provide suitable results (Herman et al.,



2013). The sensitivity of the model is expressed by the mean ( $\mu^*$ ) and standard deviation ( $\sigma$ ) of the absolute elementary effects associated with each input parameter. Where the larger is  $\mu^*$  the more a variation in the parameter induces a variation on the output. For  $\sigma$  smaller than  $\mu^*$ , the effect is monotone and interactions with other input parameters are negligible.

### **III.4. Model evaluation**

The model was evaluated against the Ganzelmeier grapevine late growth stage dataset. The source was set as a grid of 11 by 41 (along x- and y-directions) point sources, spaced every 1 m in the y direction, hence assuming a spraying over 40 m in the y-direction and at each side of the rows in the x-direction (Fig. S.A.2). The deposition along the x-axis at  $y = 0$  was used for comparison to the Ganzelmeier dataset. Preliminary tests showed that spraying at a distance larger than 20 m from this central line no longer contributed significantly to the drift along the central line.

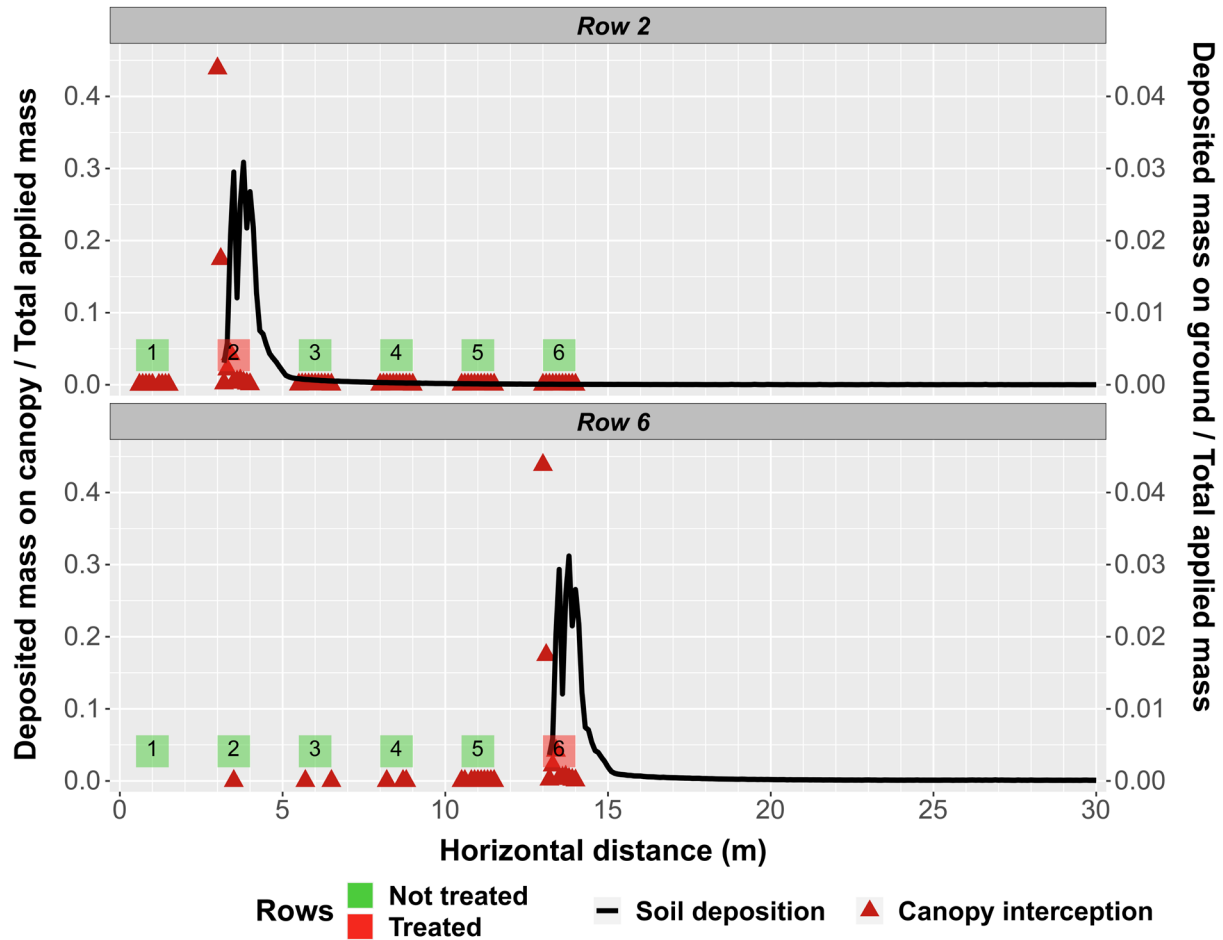
The model performance was assessed by computing the efficiency factor (EF), the root mean square error (RMSE), and the relative root mean square error (RRMSE) of the deposition out of the application field (Eqs. A30–A32 in Annexe A). EF was used to assess the model's ability to correctly represent the variability in droplet deposition. An EF closer to 1 indicates better model performance and explanation of the variability of the available observations. RMSE and RRMSE values close to 0 indicate greater accuracy of the model.

## **IV. Results**

### **IV.1. Model behaviour**

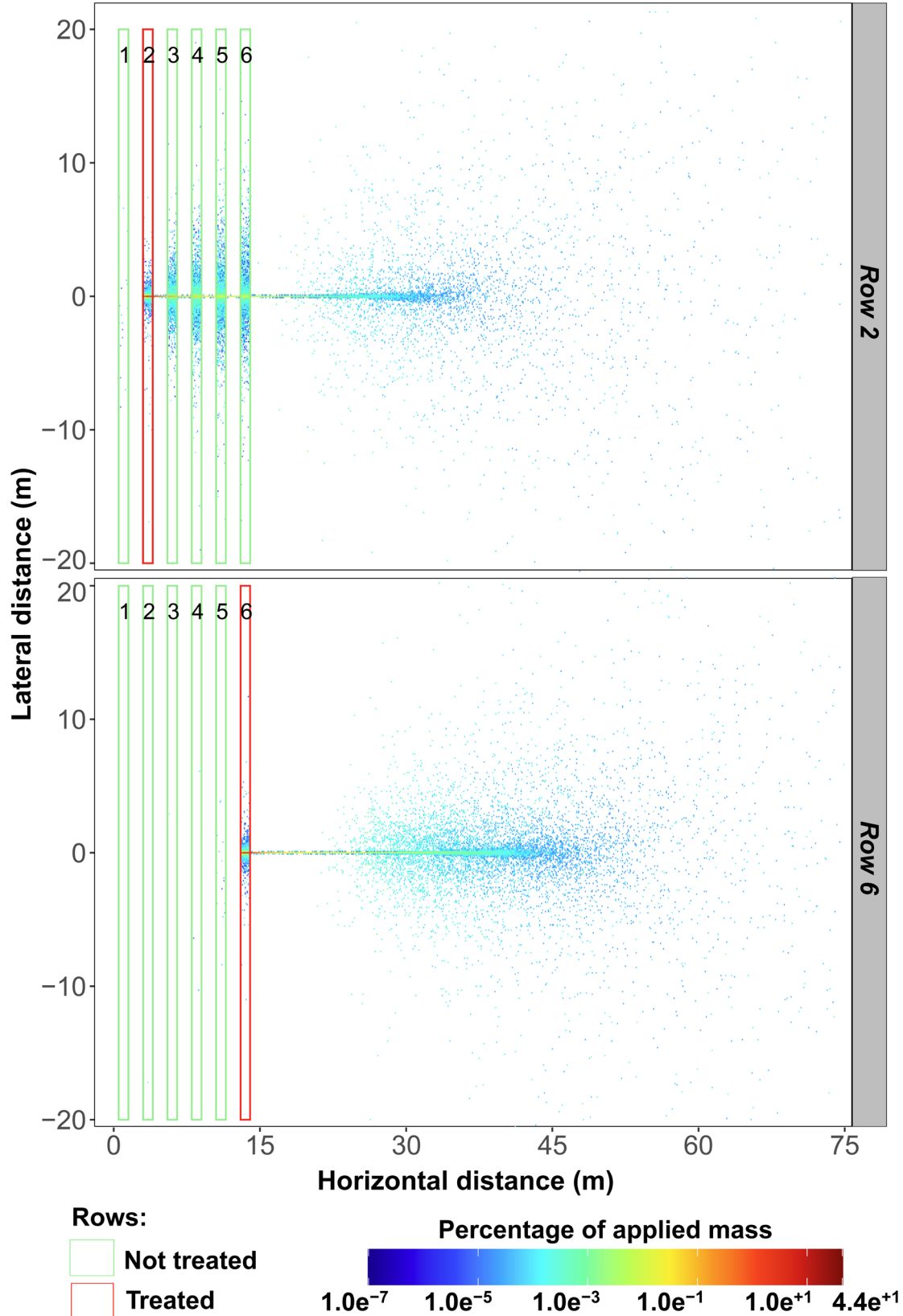
In both ROW-2 and ROW-6 simulations, the major part of the spray mass is intercepted or deposited in the treated field (Fig. III. 2), with a maximal deposition and interception at the downwind edge of the sprayed row. Up to 98.5% for ROW-2 and 87.4% for ROW-6 are deposited in the field. Canopy interception is maximal at the upwind edge of each row and then decreases exponentially. Deposition becomes much smaller 4 rows after the sprayed row (ROW-2) or 6 m after the field limit (ROW-6). Notably, some particles are intercepted and deposited on the rows that are upwind of the spraying source due to turbulence effects. As expected, in ROW-6, the deposition outfield is much larger than that in ROW-2 because there are no more vine rows after the treated row that can intercept the plume. Accordingly, the fraction of the pesticide exiting the field is 1.5% and 12.6% for ROW-2 and ROW-6, respectively, showing that spraying the last row at the field boundary is likely to be predominantly responsible for outfield drift issues.

Figure III. 2. Representation of the pesticide mass ratio deposited on soil and intercepted by canopy, where (ROW-2) refers to spraying on the second vine row and (ROW-6) refers to spraying on the sixth vine row. Numbers represented in the rectangles represent row positions relative to the field edge (going from inside the field to its edge), green rectangles represent nontreated rows and the red rectangle represents the treated row.



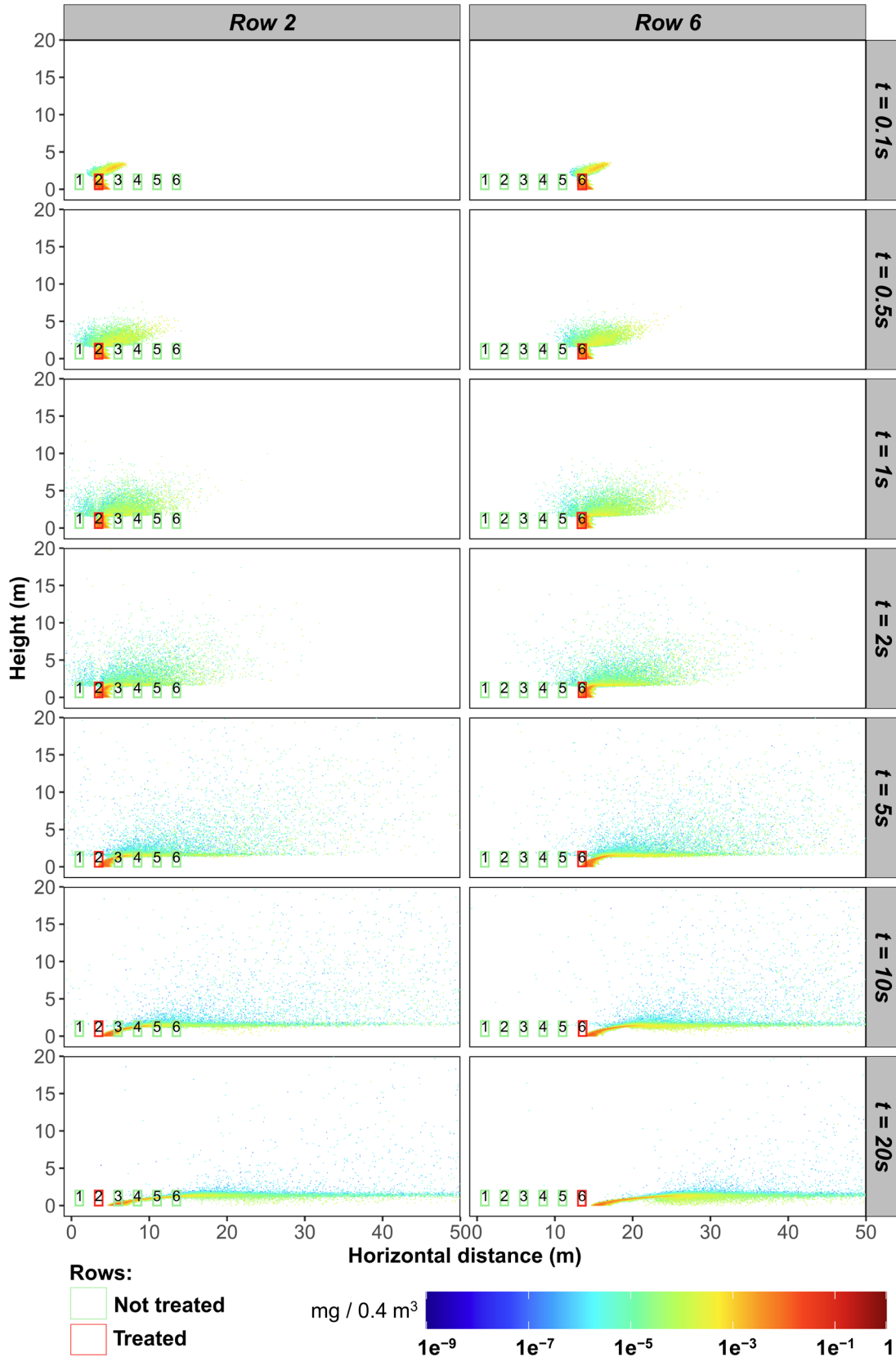
In the horizontal plane ( $x, y$ ), Fig.III. 3 shows that most of the intercepted and deposited particle mass is concentrated along the nozzle spraying direction ( $x$  direction), with both a marked lateral dispersion ( $y$  direction) and a large interception by the canopy rows within the first few metres after the source. For ROW-2, after the 3<sup>rd</sup> row, the deposition area widens laterally as the distance from the source increases, although the deposited mass decreases (Fig.III. 3). This can be explained by the fact that within the first few metres from the source, larger particles maintain the inertia given by their ejection, making their deposition concentrated near the source and along the  $x$ -axis, whereas for larger distances from the source, the smaller particles remain because they lost their inertia and therefore are influenced by the atmospheric dispersion. The same tendency is observed for the ROW-6 case. However, since there are no other rows after the treated one to intercept particles, the area where the pesticide deposition mass percentage is concentrated along the nozzle spraying direction is larger than that for ROW-2.

Figure III. 3. 2D representation (x,y) of pesticide deposited on soil and intercepted by the canopy 2 min after the spraying event, expressed in the mass percentage of the applied mass in the case of (ROW-2) spraying of the second vine row and (ROW-6) spraying of the sixth vine row. Numbers represented in the rectangles represent row positions relative to the field edge (going from inside the field to its edge). Green rectangles represent nontreated rows, and the red rectangle represents the treated row.



Finally, the simulated dynamics of the particle plume after spraying are almost similar for ROW-2 and ROW-6 (Fig.III. 4). As expected, the pesticide mass present in the air is highest at  $t = 0.1$  s and then decreases with time due to the deposition phenomenon. At  $t = 0.1$  s, the plume is small and condensed (4 m wide horizontally and vertically); at this stage, it still consists of coarse and fine particles (Fig. S.A.4). At  $t = 0.5$  s, a small part of the plume, mainly composed of fine particles with a low mass, is transported in the direction opposite to that of the spraying due to the turbulence effects mentioned above. Then, until  $t = 5$  s, the pesticide plume extends vertically and horizontally as finer particles follow the surrounding atmospheric flow. The plume reaches its maximum extent between 5 and 10 s. Then, from 10 to 20 s, the plume flattens progressively until almost all particles have fallen out. The main difference between ROW-2 and ROW-6 is that, as expected, a smaller mass reaches the field edge when the 2<sup>nd</sup> row is treated, whereas in the case of the treatment of the 6<sup>th</sup> row, the pesticide plume is already at 2 m from the edge of the last row since  $t = 0.1$  s, and a significant part of the spray plume lies outside the field.

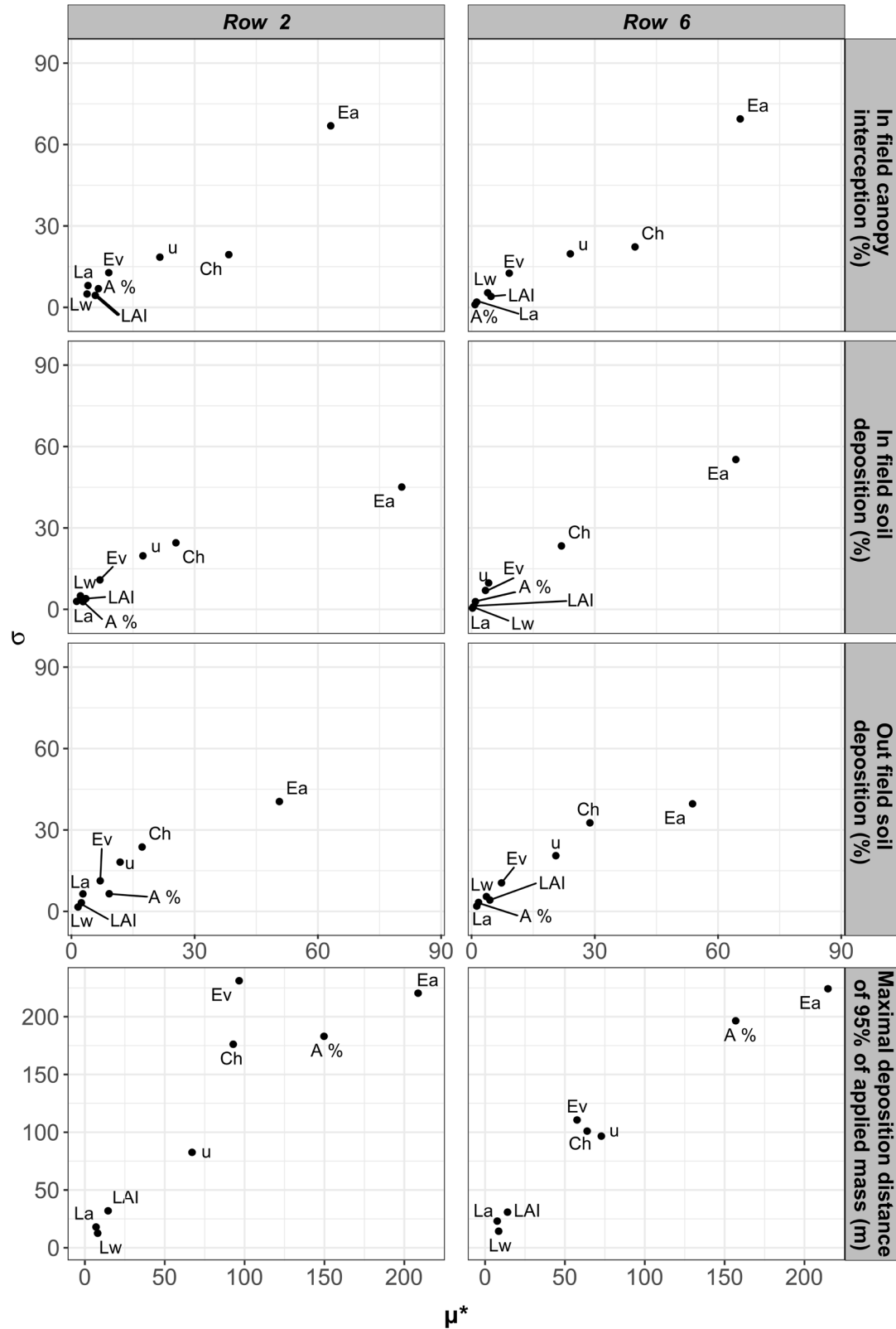
Figure III. 4. 2D representation of the vertical and horizontal evolution of pesticide clouds expressed in mass per 0.4 m<sup>3</sup> for the first few seconds following the spraying of the 2nd (ROW-2) and 6th vine rows (ROW-6). Numbers represented in the rectangles represent row positions relative to the field edge (going from inside the field to its edge). Green rectangles represent nontreated rows, and the red rectangle represents the treated row.



## **IV.2. Sensitivity analysis**

Fig.III. 5 compares the Morris index plots obtained for the two treatment cases ROW-2 and ROW-6. The median and ranges of each output variable for the most influential parameters are shown for the four defined values of each parameter in Figure S.A.5.

Figure III. 5. Comparison between Morris index plots of four output variables resulting from the treatment of the second and sixth vine rows. A% = active matter percentage; Lw = leaf width; Ch = canopy height; Ev = ejection velocity; Ea = ejection angle; u = wind velocity; La = leaf angle; LAI = leaf area index. For information, the parameters whose effect on an output variable is mostly linear are located on the right domain of the x-axis, and those having a non-linear effect or exhibiting interactions with other parameters are located near the top domain of the y-axis. Parameters whose effects are negligible lie close to the origin (0,0)



For the four selected outputs and for both spraying positions, the ejection angle is always a very sensitive factor. The strong impact of the ejection angle may be related to the fact that it determines whether the spray is directed more or less upwards/downwards and intersects with the canopy; this strongly influences the probability of interception, deposition on the ground and the possibility that the particle leaves the field. Accordingly, when the ejection angle increases, the *canopy interception*, *outfield deposition* and *maximal deposition distance* increase, whereas the *in field ground deposition* decreases because the nozzle is increasingly oriented towards the atmosphere (Fig. S.A.5). However, this trend has a limit for *infield canopy interception* because at 135° the nozzle is no longer directed to the canopy for some canopy heights.

Compared to the ejection angle, the effects of active matter proportion, ejection velocity, wind velocity and canopy height are also significant but more variable following the treated row and the selected output. The active matter proportion mainly influences the *maximal deposition distance*. Indeed, it governs the minimum diameter of the droplets after evaporation. Therefore, for small active matter content, the final diameter of droplets can be very small ( $< 20 \mu\text{m}$ ), favouring transport over long distances. The ejection velocity has a significant influence on the *maximal deposition distance* and a moderate effect on *infield canopy interception* and *outfield ground deposition* in both cases ROW-6 and ROW-2. This can be related to its effect on the trajectory and velocity of the particles during the first few seconds following their ejection until they lose their inertia at  $t=T_s$ . This effect is more pronounced for large particles because they have a large inertia and lose it later than small particles given their higher Stokes time. Consequently, high ejection velocities (when ejection is not oriented downwards) decrease infield ground deposition, increase outfield deposition and favour long-distance transport (Fig. S.A.5). The sensitivity to wind velocity is nearly similar to that of the ejection velocity with more effect on *infield canopy interception* in both cases ROW-6 and ROW-2 and moderate effect on the *maximal deposition distance* for ROW-2 since both velocities positively influence the probability of canopy interception. However, differences between the effects of wind velocity and ejection velocity are observed in the ROW-6 case, mainly for outfield ground deposition, whose variation is almost inversely proportional to wind velocity, whereas it is proportional to ejection velocity. This result can be related to the fact that for ROW-6, higher ejection velocities strongly limited the *infield canopy interception* along the last row, whereas increasing wind velocity increases the *infield canopy interception* and decrease outfield ground deposition mainly due to stronger canopy interception. The sensitivity of the model to canopy height is explained by the increased probability that particles impact leaves when the canopy is higher. Indeed, a larger canopy height leads on average to an increase *in-canopy interception* and in turn to a decrease in *infield ground deposition*.

Finally, the leaf angle, leaf width and LAI exhibit the smallest influence among the model parameters. For LAI, an overall weak linear effect is observed which may however be related to the



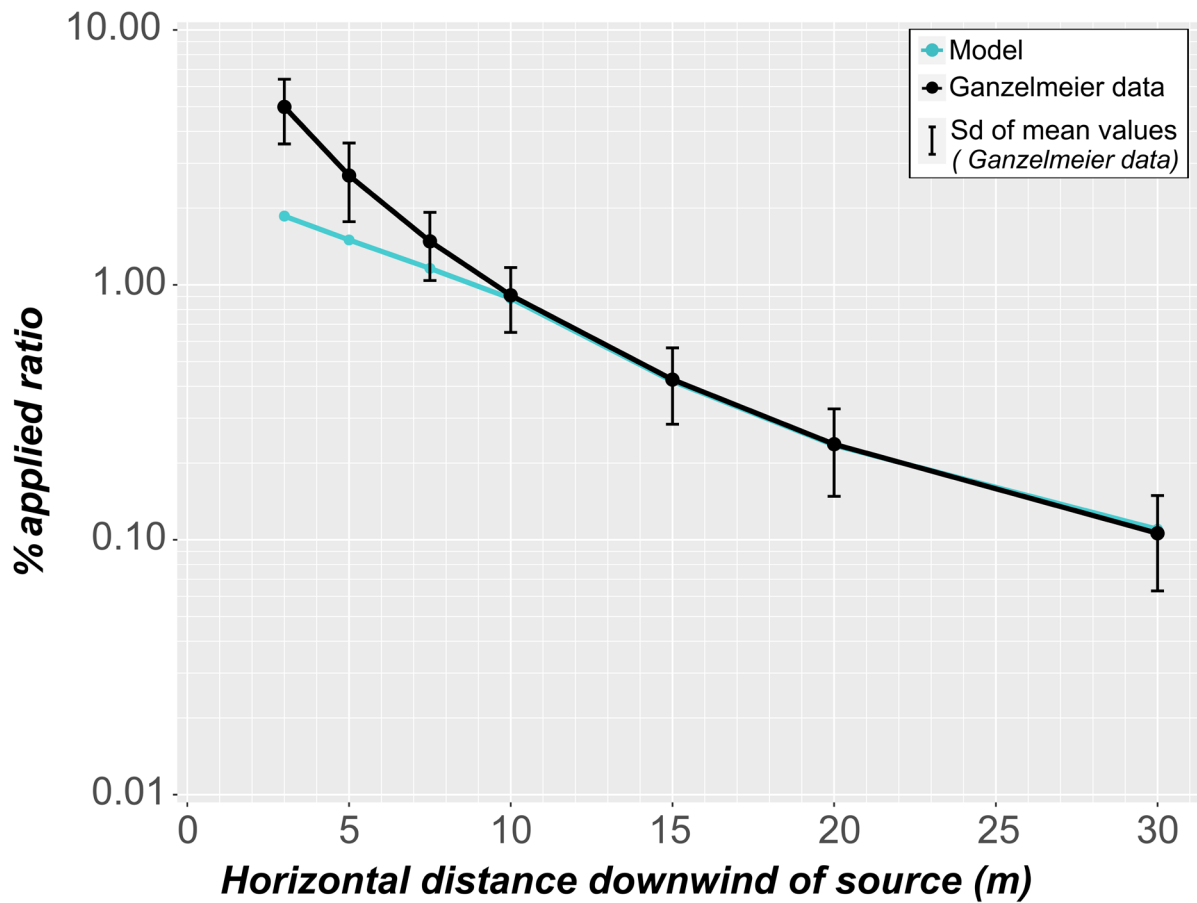
limited range of variation tested for the LAI that did not include extreme values such as those occurring during the early vine growth stage. For leaf angle, a slight effect is observed on *infield canopy interception* and *outfield ground deposition* in the case of ROW-2. As the porosity of the canopy varies according to leaf angle, its change from horizontal to vertical direction leads to the variation in the probability of particle impact on leaves. For leaf width, almost no influence is observed except for slight effects on *infield canopy interception and outfield deposition* in the case of ROW-6.

In sum, 5 main driving parameters are identified for both cases when the 2<sup>nd</sup> (ROW-2) or the last row (ROW-6) is treated, namely, ejection angle and velocity, active matter proportion, wind speed and canopy height.

### IV.3. Model evaluation

Fig.III. 6 compares the measured drift deposits of the Ganzelmeier et al. (1995) experiments with the best simulated combination of the drift deposits obtained from the calibration of our model regarding two of the unknown parameters of the Ganzelmeier experiment (ejection angle and velocity) that were identified as the most sensitive parameters in the sensitivity analysis. The model prediction in terms of outfield deposition shows rather good agreement with the experimental data, with a regular decrease in the outfield deposit with distance from the field edge. For short- and medium-range drift deposition (7–30 m), the model predicts outfield deposition well, and the simulated values are within the standard deviation of the observed mean deposit, whereas at very short distances (up to 5 m), the model significantly underestimates deposition.

Figure III. 6. Comparison between Ganzelmeier measurement data and modelled results of deposition percentage relative to the application rate along the horizontal distance from the source



A good prediction performance is found with an EF equal to 0.4 and a mean deviation between the modelled and measured deposition percentage (RMSE) equal to 1.3% of the application rate (Table III.2). This result shows that the model can satisfactorily reproduce the spray drift deposition in vineyards observed in the Ganzelmeier experiments at distances greater than 5 m, provided that the unknown velocity and ejection angles are calibrated. Detailed results are given in Supplementary Materials (Table S.A.2).

Table III. 2. The best combination of parameters used in model calibration and the respective EF, RMSE and RRMSE.

EF, RMSE and RRMSE results from the comparison between the Ganzelmeier data and modelled results of the percentage of deposited mass in relation to that applied.

Combinations	Calibration parameters		EF	RMSE	RRMSE
	Ejection velocity (m)	Ejection angles (°)			
<b>Best combination</b>	60	40, 55, 70, 85, 100, 115	0.4	1.3	2.66

## **V. Discussion and conclusion**

This section summarises the performance of the model in simulating outfield spray drift deposition, outlines the benefits and limitations of the current model by identifying areas for improvement and finally presents the potential applications of the model.

### **V.1. Accuracy of model predictions**

The model evaluation against the sedimentary spray drift experimental data of Ganzelmeier et al. (1995) showed a very satisfactory agreement between modelled and measured ground deposition with distance from the field edge (Fig.III. 6). Model predictions were within the uncertainty intervals of observations for all distances larger than 5 m, and only underestimated the observed mean values at the edge of the field (up to 5 m). The discrepancy observed at short distances can be primarily assumed to stem from the influence of local turbulence generated by the step change in the canopy height at the limit of the field and by the disturbance generated by the air jet. For simplicity these two features were neglected in our approach that considers a homogeneous turbulence in x and y and considers no effect of sprayer's air jet.

Indeed, concerning the effect of the step change, k-epsilon turbulence models that belong to the Reynolds-averaged Navier Stokes (RANS) family of turbulence models and are based on a relationship between the turbulence kinetic energy (k) and its dissipation rate ( $\epsilon$ ) (Foudhil et al., 2005), show a region of increased turbulent kinetic energy in the region  $x / h_c < 10$  downwind of the canopy (Dupont et al., 2006). However, neglecting the increase in turbulence caused by the step change may not be the sole reason for underestimating deposition near the field edge. Dupont et al. (2006) still found an underestimation in modelled deposition for maize pollen even with the use of a k-epsilon model. We note that the maize pollen had an aerodynamic diameter of 80  $\mu\text{m}$  that is representative of the mode of the droplet distribution studied here (Fig. S.A.3). The increased deposition may also be due to a complementary process that is not described in any drift model and corresponds to the preferential sweeping and clustering of particles with a Stokes number of  $\sim 1$  (inertial time scale equals the turbulent mixing time scale). Under such conditions, the particles sweep out of the smallest eddies on the downwards side of the eddies, increasing the deposition velocity (Aliseda et al., 2002). For certain particle sizes and turbulent conditions, an increase in the deposition velocity by 20 to 50% may occur (Wang & Maxey 1993; Dupont et al., 2006). Thus, improving deposition modelling at the field edge by incorporating preferential sweeping and clustering processes may be more beneficial than increasing the complexity of the description of the turbulence in this area.

Concerning the air jet misrepresentation, the adopted simplification may induce an underestimation of the particle velocity away from the nozzle. Indeed, measurements of Gu, Zhu, & Ding (2012) and modelling results of Hong, Zhao, & Zhu (2018a) show a reduction of sprayer air jet

velocity by ~30% on average from the nozzle outlet to 0.5 m from the nozzle. For the simulated droplet diameter distribution, the droplet travel time over that distance is smaller than the Stokes time (see Fig. S.A.6.a and b in Supplementary Materials) which means that the droplets will have a velocity close to their initial velocity at that distance. Droplet velocity values simulated by ADDI-SprayDrift showed an average reduction of droplet initial velocity by ~60% (see Fig. S.A.6.c in Supplementary Materials.). Therefore, we can state that our model would underestimate the particle velocity by ~40% on average at 0.5 m from the sprayer due to air jet misrepresentation. Underestimating particle velocity may also in principle modify droplet interception and droplet evaporation. Particle velocity influences impaction efficiency ( $E_i$ ) mainly for Stokes times lower than 4 as can be seen in equation A29, in the Annexe A, taken from Aylor (1982) and Aylor and Flesch (2001) (see Fig. S.A.7. a in Supplementary Materials). However, for the involved range of droplet diameter and velocities, the Stokes number remains larger than 4, the impaction efficiency does thus barely change (Fig.S7.b Supplementary Materials). Moreover, particle velocity influences evaporation time  $\tau_e$  (see equation 3 with Sherwood and Reynolds numbers). However, here again, for the range of expected bias in particle velocities, the variation in simulated evaporation will be limited (see Table S.A.3 in Supplementary Materials). The air jet misrepresentation also neglects the additional effect of the sprayer's air jet on the local turbulence, which leads to underestimate the droplet lateral dispersion near the nozzle that would mainly affect droplets smaller than 100  $\mu\text{m}$  in diameter (see Fig. S.A.8 in Supplementary Materials). Overall, the misrepresentation of the air jet is therefore expected to mainly affect particles in the smallest range of droplet sizes and, in turn, to contribute to the reduction of the accuracy of the model predictions at distances larger than ~50m. Near the sprayer, the largest particle velocity would also be underestimated leading to a possible reduced accuracy in the location of deposition at a distance that is typically smaller than 5 m.

Comparison of the present performance of ADDI-spray drift to that of other Lagrangian models is difficult because there are large differences between how the models were evaluated (crops, spraying material, performance criteria), and because ADDI-spray drift was only evaluated for airblast on vineyard. This comparison can only be made with ARPS, which is applied in vineyards within a distance of less than 8 m from the nozzle. At these distances, ARPS model showed a severe overestimation that was interpreted by Chahine (2011) as the consequence of neglecting droplet evaporation mechanism.

It is important to emphasise some limits of the present model evaluation. A first limitation is that the evaluation was restricted to the prediction of outfield deposits. Further evaluation is clearly needed to test the performance of the ADDI-Spray drift in simulating air concentrations and canopy interception. However, this requires the collection of relevant data with observed air concentrations and ground and canopy deposition in field trials of spray drift in vineyard fields because our primary objective was to develop a drift model that can simulate the case of grapevines. A second limitation is that a part of the discrepancies between simulated and measured deposits may be due to the fact that the simulated values refer to deposits on a flat surface whereas Ganzelmeier measurements refer to deposits

on petri dishes. The last limitation is that we evaluated the model performance against this dataset while calibrating some parameters. Indeed, given the lack of information in the Ganzelmeier report about the exact emission characteristics of the sprayers used, it was necessary to calibrate two model parameters, ejection angles and velocity, out of 11. However, calibration of these parameters was restricted to a very narrow range of values, corresponding to the uncertainty range of the values of the ejection velocity and the angles of an axial fan sprayer. Accordingly, we assume that the calibration of the two parameters was very focused and was not able to compensate for the main conceptual weaknesses of the model, as may happen when calibration is performed for a large number of parameters with large permitted ranges of parameter values.

## **V.2. Advances and limitations of the model**

The ADDI-SprayDrift model predicts pesticide distribution between the ground, the canopy and the atmosphere at the time of application by modelling surface deposition at short and medium distances and airborne concentration at any distance, time and height. It integrates the key processes expected to govern the behaviour of the spray droplets after their emission, namely initial inertia of the particles, droplet size changes due to water evaporation, atmospheric dispersion accounting for atmospheric stability (neutral, stable and unstable), and canopy geometry effects on atmospheric turbulence, as well as deposition processes to ground and the canopy. While water evaporation is generally considered in the drift models, not all of them consider the atmospheric stability conditions, the canopy effects on atmospheric turbulence and the distinction between deposition processes to ground and interception by the canopy within the treated field together.

A key strength of the model is thus that it incorporates particle interception by the canopy. Thompson & Ley (1983) already demonstrated using simulations the importance of droplet interception by the canopy on outfield soil deposit drift in the case of low canopies. This effect is even more important in the case of high canopies, such as in vineyards and orchards. The Lagrangian approach selected in this model is particularly relevant for the calculation of droplet interception by canopy leaves, as it allows to account easily for initial particle inertia and impaction processes. The ADDI-SprayDrift model considers the inertial effect due to high initial velocity that can be as high as hundreds of metres per second. Similar adjustments were introduced to the random walk models in the literature to address the effect of air entrainment and the initial spray sheet on the droplet trajectory near nozzles in Walklate (1986) while Dorr et al. (2005; 2008) and Miller & Hadfield (1989) addressed this issue by combining the model with a ballistic model. Another important feature introduced in the model is the impaction over horizontal surfaces that we felt necessary to represent the impactions of droplets whose ejection direction includes a vertical component. This factor has been added in Eq. (5) of the model as an additional term to the equation of Aylor (2005). Some models consider in addition the bounce-off of the particles or their breakup and the subsequent probable secondary interception such as Dorr et al. (2005;

2008; 2016) and Hong et al. (2018a). But, for simplicity, we choose to not consider these two phenomena because they increase the complexity of the model, resulting in more computationally costly simulations.

In ADDI-SprayDrift, the turbulence is considered to be horizontally homogeneous over the computational domain. Most reviewed models use the same assumption, which is justified because they consider annual crops (short crops < 1 m height). This assumption of homogeneity may however be considered a major limitation of the model in the case of high crops, but in practice, it did not appear to affect the quality of model predictions at distances larger than 5 m. Furthermore, the vertical profiles of turbulence in ADDI-SprayDrift are fully described within the canopy and above, accounting for both the atmospheric stability and canopy geometry effects, which is an original feature of our model and is important for simulating dispersion, particularly in vineyards and orchards.

A limit of the model is that it does not take into account the effect on the local airflow and turbulence of the air jet that is used to increase spraying efficiency in case of tall crops. As discussed above, this simplification mainly influences the simulation of drift near the sprayer (<5m). In ARPS and Fluent that are designed for vineyard and apple orchards, a Large Eddy Simulation (LES) method and a k-epsilon CFD model were used to represent the turbulence at the local scale. This may be beneficial, particularly to capture deposition for distances <5 m. For a better accuracy at short distances and to overcome the effect of the sprayer air jet, more complex consideration of the local turbulence should be used, which can be addressed by coupling the model to more elaborated turbulence models like k-epsilon models or Large Eddy Simulation models (LES).

The source is a key parameter of pesticide drift dispersion modelling. Indeed, our sensitivity analysis highlighted the predominant influence of ejection angle and velocity on model outputs, including outfield deposition. Similar findings have been identified in other models (e.g. in Thompson & Ley model or IDEFICS) regarding their sensitivity to source parameters, such as effective release height and droplet size distribution. In ADDI-SprayDrift a simple approach was adopted by describing these parameters according to available experimental data. Information regarding source characteristics is however often uncertain, and questions still arise concerning the lower tails of the droplet size distribution and the precision of the ejection velocity at the outlet of the nozzles. The use of atomisation models, as applied in the CFD model of Endalew et al. (2010), can help to address droplet size distribution variation according to spraying conditions. However, due to their complexity, they are rarely used in spray drift models.

It must be emphasised that the current version of ADDI-SprayDrift is not adapted for complex spraying systems, such as a tunnel sprayer. Indeed, this latter type of sprayer requires the characterisation of additional processes such as the turbulence induced by the air assistance between the two retrieval panels, the suction phenomenon that allows the recovery of the non-intercepted drops and the drag effect generated by the forwards speed of the sprayer. Moreover, the chosen Lagrangian random walk

modelling approach provides precise information about individual pesticide droplet positions at different time steps, distances and heights. However, even if these models prove to give good predictions at field and supra-field levels, their prediction quality at distances larger than 500 m requires a large number of released particles (Craig, 2004), increasing the computational cost. This may be a limitation to implementing this model in large-scale studies.

### **V.3. Potential applications of the model**

The ADDI-SprayDrift model is conceptually a generic model that can be used to simulate pesticide infield distribution, short/medium-range deposition and the position of spray plumes at different distances and times. This paper illustrates its application in the case of vineyard situations. However, we expect that the model can be further adapted for application to drift simulation in orchards and arable crops applications with boom-sprayers, provided the appropriate droplet emission characteristics are known. ADDI-SprayDrift will then be particularly useful for assessing the effectiveness of application techniques by quantifying off-target deposition (soil or outfield) and contamination by pesticide spray drift. Prediction of the evolution of the drift plume would also allow accurate estimation of bystander or resident exposure to pesticides by inhalation or skin contact. The model can also provide information concerning the origin of pesticide droplets and thus the relative contribution of each row to airborne concentrations and soil deposits over a given time. This information may then be used in exposure models to evaluate pesticide drift impacts on ecosystem functioning and bystanders. Overall, this would help identify leverage for minimising losses by optimising application techniques and machinery and provide recommendations on best practices regarding weather conditions and development stage.

## Chapitre 4

# *Évaluation de la distribution des pesticides lors de l'application et de l'exposition des passants à la dérive sédimentaire et aérienne à l'aide d'une approche de modélisation*

*Dans ce chapitre, le modèle ADDI-Spray drift est déployé pour évaluer l'exposition directe des passants lors des pulvérisations par inhalation et dépôts cutanés dans un contexte viticole. Pour se faire, une version multi-sources du modèle a été développée. Sont décrits (sous la forme d'un article à soumettre) les principaux résultats obtenus sur les scénarios identifiés en croisant divers types de pulvérisateurs/buses/concentrations de bouillie ou conditions météorologiques typiques de traitements en matinée ou en milieu de journée ainsi que des confrontations avec un jeu de données et des modélisations existantes. En perspectives, une analyse plus approfondie de l'ensemble de ces résultats permettra de dégager une vision globale de l'impact de ces différents facteurs sur l'exposition des passants.*





## *Assessment of pesticide distribution at application and bystander exposure to pesticide spray drift using a mechanistic modelling approach*

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### **Abstract:**

Bystanders and residents may be exposed to pesticide spray drift during their application on agricultural fields, either directly by inhalation or by dermal contact or indirectly, by contact with contaminated surfaces. Assessing these exposure pathways requires a good understanding of the underlying processes and of their drivers. The ADDI-spray Drift model allows assessing the effect of the main drivers - sprayer and nozzle types, active ingredient concentration in the spraying solution, canopy growth stage or meteorological conditions- on pesticide distribution at application time within the treated field, sedimentary drift and airborne concentration downwind of the treated field. Selecting three main sprayers used in vineyards, two nozzle types (standard and low drift), two growth stages, three active ingredient concentrations and two meteorological conditions representing morning and midday applications, 60 scenarios were run. Model's predictions for pesticide initial repartition and bystanders exposure via inhalation and dermal deposition showed good agreements with experimental data for same sprayers with however a slight overestimation of exposure of bystanders through dermal deposition. Additional validation of the model for exposure assessment and for pesticide outfield vertical profile at different distances remains however necessary by using more exhaustive experimental data.

**Keywords:** *ADDI-SprayDrift*, vineyard, sedimentary spray drift, airborne spray drift, dermal deposition, inhalation.

## **I. Introduction**

Due to the increasing use of pesticides, various ecosystem compartments undergo a contamination leading to an exposure of all living organisms including human beings (Zaller et al., 2022). Several epidemiological studies in human health show a link between the proximity of households or workplace to agricultural lands and the increase of various forms of cancer (Leukaemia, respiratory cancers), neurodegenerative diseases (Parkinson's, Alzheimer's), fertility and dermatological problems (Fuentes et al., 2021), and of respiratory problems going from a simple disturbance to a chronic obstructive pulmonary disease (Simões et al., 2022). The exposure of humans may result from 1) a direct exposure via dermal contact and via inhalation during pesticide application or 2) indirect exposure through dermal and oral routes while coming into contact with contaminated surfaces, through hand-to-mouth transfer, through ingestion of contaminated food and water or inhalation of vapour after application (EFSA, 2022). Based on the acuteness of the exposure, its duration and the location of the individuals regarding the treated field, the exposed population is generally classified into four groups, namely, operators, workers, bystanders and residents (Hulin et al., 2021). Operators and workers may be exposed acutely and for a long period when dealing with products mixture preparation and spraying or while working within the treated plot after application (EFSA, 2014; 2022). However, their exposure can be strongly limited by using personal protective equipment that are currently recommended during all treatment process or for the re-entry into the treated field. In contrast, bystanders during application periods and residents in the neighbourhood of treated fields are also submitted to acute (bystanders) or chronic (residents) exposure to pesticides, but are without any protective equipment (Matthews & Hamey, 2003). Besides, the population of bystanders and residents is very heterogenous and may include vulnerable individuals like children, elderly people and people with severe illnesses (Figueiredo et al., 2021; Simões et al., 2022). The risk of human exposure, especially of residents and bystanders, is particularly in wine growing regions given the high use of pesticides in permanent crops (Aubertot et al., 2005) and the larger spray drifts when spraying high canopies (Hong et al., 2018c). To protect bystanders and residents, acting upstream on the different exposure routes is essential since it is difficult to recommend them to wear protective equipments. In this respect, improving spraying methods that reduce drift and defining non treatment zones in the neighbourhood of households are often mentioned as levers.

Indeed, atmospheric dispersion of pesticides is one of the main exposure route of residents and bystanders (Zaller et al., 2022). During application, residents and bystanders are exposed through dermal contact and inhalation due to spray drift and volatilization. After application, they are still exposed to vapour (inhalation) for several days depending on compound volatilization. Even if the possible exposure routes through atmosphere are known, the assessment, quantification and reduction of their contribution to the exposure of bystanders and residents remain insufficiently documented

(ANSES, 2017; Hulin et al., 2021). Several studies explored the variation of resident and bystander exposure and the efficiency of anti-drift nozzles or spraying technics to limit this exposure, but general conclusions remain difficult if not impossible since application technics and field conditions vary considerably between experiments (Nuyttens et al., 2010). Furthermore, a lack of data has been highlighted by EFSA (2014; 2022) and Mercier (2020) concerning exposure related to pesticide application in orchards and even more in vineyards. The high phytosanitary pressure for these crops leads to a significant amount of pesticide use and a potentially high probability of exposure due to high droplet atmospheric dispersion related to the type of used machinery based on horizontal diffusion and air assistance (Matthews & Hamey, 2003; PestiRiv, 2021). To our knowledge, the available data estimating exposure in vineyards are those measured by Mercier (2020) and those predicted by the BROWSE model (Butler Ellis et al., 2017) and the German model (Martin et al., 2008). These data refer to spraying conditions that vary largely in terms of types of sprayers, anti-drift technics, canopy development stages and structures, and meteorological conditions. It is thus not possible to identify from these data the separate effect of each of these factors on the exposure of bystanders or residents, which would be of great help for analysing the factors that are the most influential and, in turn, for identifying the levers that can reduce exposure. The recent availability of the ADDI-sprayDrift model, (Djoughri et al., -under review) that represents the main processes influencing spray drift in vineyards, namely emission, dispersion, deposition and canopy interception and considers all mentioned factors, allows to analyze in a more systematic approach by simulation the theoretical influence of these factors.

In this paper, we therefore present a simulation study of the variation of bystanders exposure by inhalation and direct dermal contact related to spray drift during the treatment of a vineyard plot following the type of used spraying material, the vineyard development stage, the spray mix concentration and the meteorological conditions. In the following we describe briefly the ADDI-spraydrift model, how we built the simulation scenarios and checked the consistency of the simulations, then present the sensitivity of simulated within-field pesticide distribution, outfield pesticide deposits and bystander exposure to the considered factor, and the consistency of the simulations with available data from the literature, and finally discuss and summarize the main results.

## **II. Materials and methods**

### **II.1. Presentation of used spray drift and exposure models**

The Atmospheric Dispersion Deposition and Interception of Spray Drift model “*ADDI-SprayDrift*” (Djoughri et al., under review) is used in this study to model pesticides distribution during application, spray drift and the following exposure of bystanders. Few improvements have been integrated into the previous version of *ADDI-SprayDrift* model in terms of source representation and bystander exposure assessment. Indeed, in the first version of the model, the source of droplets was described as a point

source, taking into account ejection heights, angles and velocities of the generated droplets together with their size distribution and the outputs were : the interception of the solution by the treated canopy, the deposits on the soil within the treated field and downwind, the vertical profile of airborne concentrations downwind as a function of the distance. Subsequently, the novelty of the new version is 1) its multi-source version *i.e.* the possibility to treat the whole field at the same time, and 2) predefined outputs in order to quantify exposure of bystanders (or residents) through dermal deposition and inhalation at various distances.

For the source representation, the spraying of the entire field is represented by a set of discrete and equidistant source points in the field. The spacing between the source points can be adjusted according to the type of used sprayer or the required degree of precision regarding the representation of the source. Each source point may encompass one or various ejection points, representing nozzles. Depending on the used sprayer and its characteristics, the number of nozzles per source point and their respective height and distance regarding the treated canopy are adjustable. In the context of vineyard treatment, this version allows to specify the number of treated rows and rows sides.

For the assessment of bystander exposure, automatic predefinition of outputs have been implemented, allowing the quantification at defined distances and heights of all particles passing through a spherical or cubic volume surrounding the bystander. Indeed, ADDI-Spray Drift model is based on a lagrangian approach tracking all droplets on their trajectory. Thus, the mass of active ingredient (a.i.) in a given volume is calculated by summing the mass of all the droplets passing through this volume and taking into account the a.i. concentration within the spraying solution. Regarding the volumes to be considered, exposure through inhalation can be represented by a cube ( $V_{cubic-inhalation}$ ) by centring it at inhalation height. For dermal exposure, a cubic representation ( $V_{cubic-dermal}$ ) has been designed in order to represent the human body. For both cubic volumes ( $V_{cubic-inhalation}$  and  $V_{cubic-dermal}$ ), it is possible to fix the x, y and z dimensions depending on which category of individual exposure is studied (*e.g.* adults, children..).

To calculate the exposure of an adult bystander, we used the recommended protocol and reference values described in the EFSA guidance document (EFSA, 2022) for inhalation rate, body weight and exposure duration. First, the inhalation rate of a bystander is set equal to  $3.18 \text{ m}^3 \cdot \text{h}^{-1}$  for an adult having a body weight equal to 60 kg. The bystander exposure to spray drift droplets by inhalation is considered occurring during the 15 min following the application. Accordingly, given the air inhalation volume during 15 min (corresponding to  $0.795 \text{ m}^3$ ) and the concentration of active substance calculated by the model at the defined inhalation volume ( $V_{cubic-inhalation}$ ), we assess the exposure of bystanders by inhalation by calculating the mass of inhaled a.i. as presented above. Inhaled a.i. is considered to be fully absorbed (100%) by the human body. Then, for the exposure through dermal deposition, we maximised deposition as a worst scenario by considering that all particles passing through the cubic volume representing the human body ( $V_{cubic-dermal}$ ) are deposited, thus deviation of the airflow around the body is ignored. A reduction of 18% is applied to deposited mass, corresponding to the percentage

of reduction of exposure due to the semi permeation of clothes (EFSA, 2022). Note that not all deposited mass is considered to be absorbed by the body, since only up to 10% of the deposited mass is absorbed depending on the applied a.i.. Thus, the exposure by dermal deposition is evaluated through the calculation of the mass that penetrate into the volume representing the body, which results from the multiplication of the deposited mass within the defined body volume ( $V_{cubic-dermal}$ ) after clothes filtering and taking into account appropriate absorption rate.

## **II.2. Scenarios and simulations**

The source characteristics including spraying material design and settings and the fraction of active ingredient within the tank mixture, the vineyard development stage and the meteorological conditions are drivers of spray drift (Djoughri et al., under review). To assess the effect of these drivers on pesticide spray drift deposition and on the exposition of bystanders as a function of distance downwind of the treated field, we elaborated contrasted scenarios presented in this section, together with the approach used to assess the relative influence of each factor on representative outputs and comparison with available data in the literature.

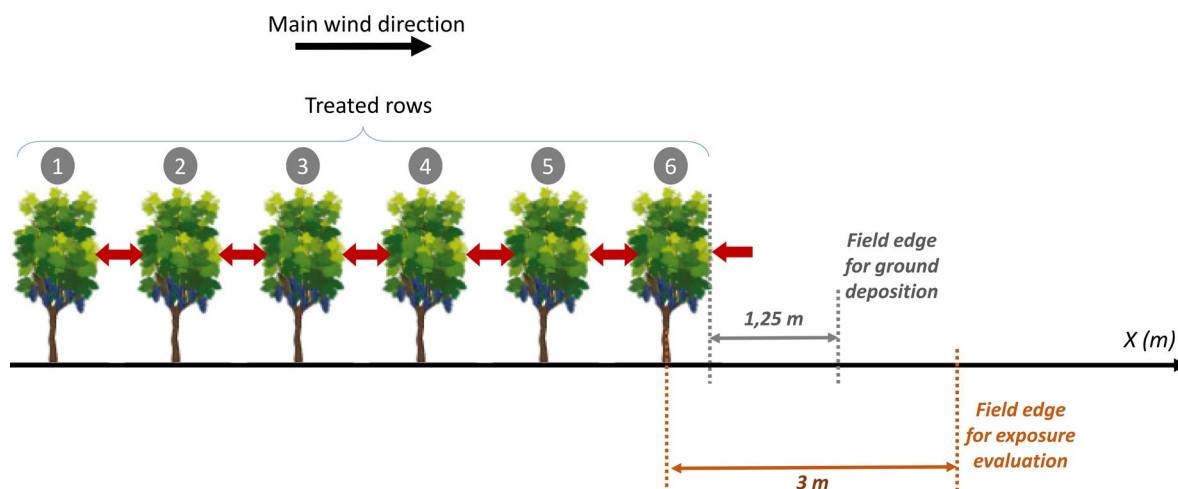
### **II.2.1. Scenarios building**

#### *II.2.1.1. Field configuration*

We choose a conventional treated field configuration in agreement with ISO 22866 (2005) recommendations for spray drift measurement in order to allow further comparison of the results obtained in this study with available experimental data. This virtual field is defined as a flat area of 20m width, composed of 6 rows with 2.5m inter-row distance, and a length of 40 m length. This field length correspond to edge area that is considered as to contribute directly to spray drift at field central line. Greater distances from the field boundary no longer contributed significantly to drift along this central line. The characteristics of the canopy (height, LAI, ...) are set to vary according to the chosen vine canopy growth stage. We consider that all rows sides are treated except the upwind side of the first row (Fig. IV.1). The spraying of the entire field is represented by a set of discrete and equidistant source points in the field, with 0.5 m between each source point along each treated row faces. Each source point encompasses various ejection points according to tested sprayers.

## Chapitre IV : Évaluation de la distribution des pesticides lors de l'application et de l'exposition des passants à la dérive sédimentaire et aérienne à l'aide d'une approche de modélisation

Figure IV.1. Representation of treated rows and the considered field edge for ground deposition and for exposure assessment. Trees are used to illustrate treated rows and the numbers over each of them indicate their respective position regarding field edge (going from inside to outside the treated field), red arrows represent treated row sides.



### II.2.1.2. Vineyard development stage

To investigate the effect of the development of vineyard canopy on sedimentary spray drift and the bystander exposure, two growth stages were selected, namely, an early and a developed growth stage corresponding to 19<sup>th</sup> and 57<sup>th</sup> phenological stages of BBCH scale, respectively. The characteristics of the canopy are summarised in Table IV.1.

Table IV. 1. Selected vineyard development stages and their corresponding characteristics

	March/April	May/June
<b>Growth stage * (BBCH growth scale)</b>	<b>Early stage (19)</b>	<b>Developed stage (57)</b>
<b>Canopy height (m)</b>	1.43	2.0
<b>Minimum leaves wall height (m)</b>	0.50	0.40
<b>LAI of the field</b>	1	3
<b>LAI of vine rows<sup>#</sup></b>	7,8	10,7
<b>Vine row width (m)</b>	0.32	0.70
<b>Leaf width (m)</b>	0.12	
<b>Leaf angle (°) with respect to the horizontal direction</b>	75	

\* According to BBCH scale of vine phenological stages, at the 1<sup>st</sup> growth stage (19) the vines have only few leaves (9 or more) while at the 5<sup>th</sup> growth stage their development reached its maximum (Inflorescences fully developed)

# The LAI of vineyard rows is calculated as a function of field LAI, rows spacing, width and height, and leaf wall starting.

II.2.1.3. *Source characteristics*

To illustrate the effect of sprayer type on spray drift deposition and bystanders' exposition, we tested the three main sprayers used in vineyards, namely, the pneumatic sprayer, the axial fan sprayer, and the side-by-side sprayer. For each sprayer type, ejection heights, angles and velocities, and the size distribution of generated droplet were defined according to the literature. Additionally, the effect of nozzle type was tested for the axial fan sprayer and for the side-by-side sprayer by choosing standard (S) and low drift nozzles (LD).

Accordingly, the mean diameters ( $\mu_{dp}$ ) and the standard deviation ( $\sigma_{dp}$ ) of droplet size distribution for each sprayer and for the two nozzle types were calculated (see Table IV.3) based on the  $D_{v0.1}$ ,  $D_{v0.5}$  and  $D_{v0.9}$  values (Table IV.2) given in the literature, that is Garella et al. (2020) for the pneumatic sprayer, Duga et al. (2014) for the axial fan sprayer and De Cock et al. (2016) for the side-by-side sprayer.

Table IV.2. Droplet diameter for which 10%, 50% and 90% of the applied volume consists of droplets with a diameter smaller than  $D_{v0.1}$ ,  $D_{v0.5}$  and  $D_{v0.9}$ , respectively.

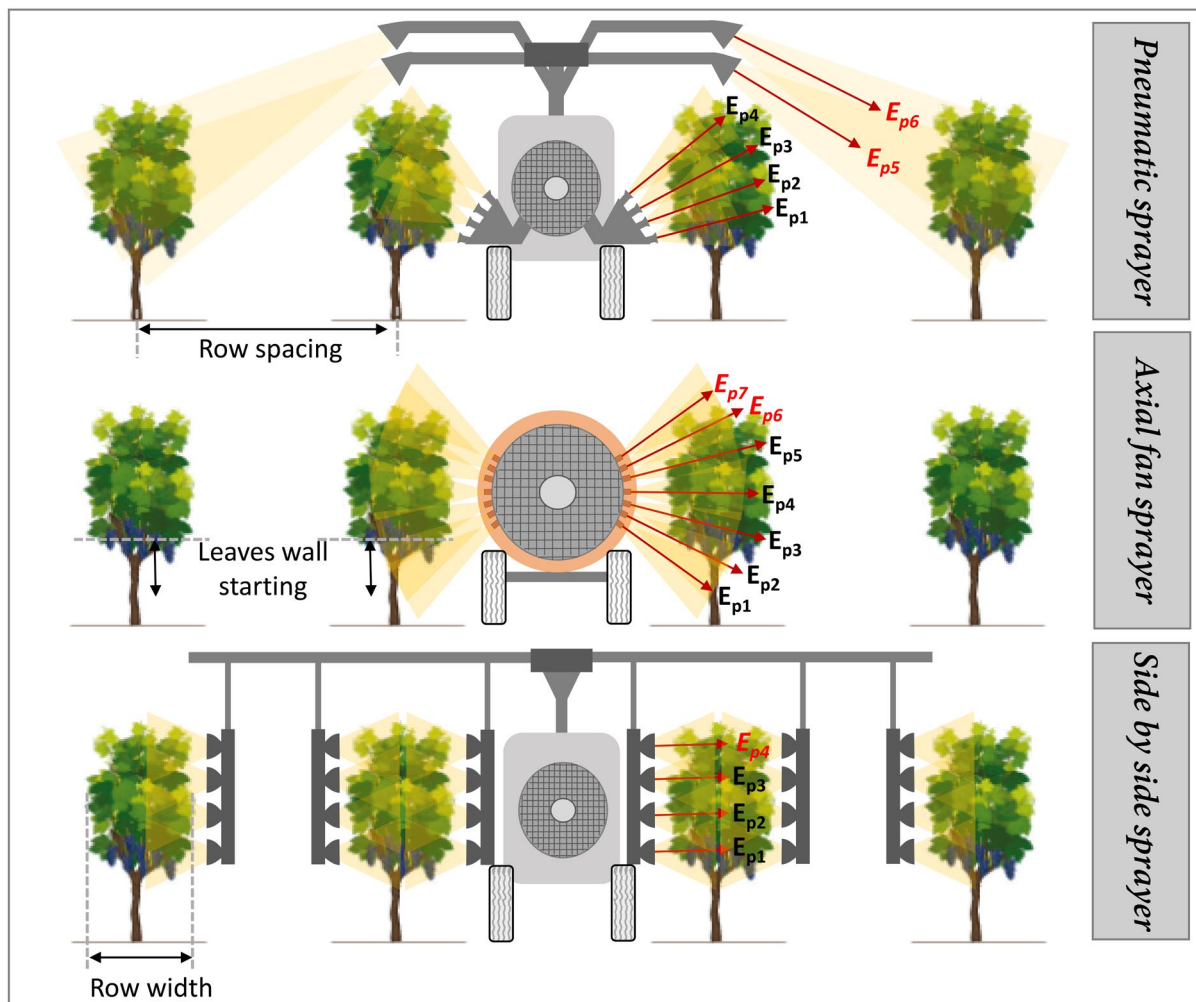
		Experimental data		
Nozzle type		$D_{v0.1}$	$D_{v0.5}$	$D_{v0.9}$
<b>Pneumatic</b>	No nozzle	50	80	250
<b>Axial fan sprayer</b>	Hollow cone nozzle – ATR Yellow – 10 bar	54	105	200
	Low-drift nozzle TVI 110 02 – 10 bar	180	550	800
<b>Side by side sprayer</b>	Standard nozzle AXI 110 02 – 2 bar	62	162	340
	Low-drift nozzle CVI 110 02 – 2 bar	150	450	750

As the active matter proportion within tank mixture affects the final diameter of droplets after evaporation (Djoughri et al., under review), we tested this effect by choosing three contrasted values representing the minimum, the mean and the maximum proportion that can be applied based on current practices for fungicide application and corresponding to 0.02%, 0.2 and 2% w/w of the applied rate, respectively.

Depending on the canopy growth stage and following good agricultural practices, the configuration of the sprayers may change in order to optimise the treatment. Especially, some nozzles/ejection points that may spray above or below the canopy are usually closed to reduce drift losses. Consequently, the configuration of the three sprayers was adapted to each canopy growth stages. The considered ejection points and their respective heights, angles and distance regarding treated row side used to generate droplet size distribution for each growth stage are represented in Figure IV.2 and Table IV.3.



Figure IV.2. Representation of the three used sprayers, considered ejection points and treated vineyard row sides. The 6 ejection points and their order are illustrated by the abbreviation ( $E_p$ ) followed by their respective order number. Ejection points that have been turned-off for simulations dealing with spraying during a vineyard early growth stage are represented in *italic red*.



In the case of developed growth stage of vineyard, six ejection points were set for the pneumatic sprayer where four ejection points are treating the adjacent rows sides and the two upper points are treating the left and right side of adjacent rows. For the axial fan sprayer, seven ejection points treating the row side are set, while for the side-by-side sprayer, we defined four ejection points spraying the rows sides. In the case of an early growth stage of vineyard, while canopy is lower, the upper ejection points of sprayers were turned-off. Consequently, the pneumatic sprayer and the axial fan sprayer were set so that 2 ejection points were removed  $E_{p5}$ ,  $E_{p6}$  and  $E_{p6}$ ,  $E_{p7}$ , respectively. While for the side-by-side sprayer, only one ejection point was turned-off ( $E_{p4}$ ) (see Table IV.3 and Fig IV.2).

Table IV.3. Characteristics of ejection points used for the three chosen sprayers. Ejection points that have been turned-off for simulations dealing with a spraying during a vineyard early growth stage are represented in italic red.

Used sprayer	Ejection points	Ejection height (m)	Ejection angle* (°)	Ejection velocity (m/s)	Droplet diameter (µm)		Distance regarding the treated row edge (m)	Treated row sides
					Mean	SD		
Pneumatic	Ep 1	0.50	90	60	42	29	0,39	4
	Ep 2	0.63	100				0,43	
	Ep 3	0.75	120				0,47	
	Ep 4	0.85	135				0,52	
	<i>Ep 5</i>	<i>2.35</i>	<i>65</i>				<i>2.25</i>	
	<i>Ep 6</i>	<i>2.50</i>	<i>65</i>				<i>2.25</i>	
Axial fan	Ep 1	0.88	45	55	$D_m^{(S1)}= 54$ $D_m^{(L1)}= 163$	$D_{sd}^{(S1)}= 30$ $D_{sd}^{(L1)}= 103$	0.65	2
	Ep 2	0.98	60				0.60	
	Ep 3	1.08	75				0.55	
	Ep 4	1.18	90				0.50	
	Ep 5	1.28	105				0.55	
	<i>Ep 6</i>	<i>1.38</i>	<i>120</i>				<i>0.60</i>	
	<i>Ep 7</i>	<i>1.48</i>	<i>135</i>				<i>0.65</i>	
Side by side	Ep 1	0.50	90	40	$D_m^{(S2)}= 48$ $D_m^{(L2)}= 125$	$D_{sd}^{(S2)}= 36$ $D_{sd}^{(L2)}= 87$	0.5	6
Ep 2	0.75							
Ep 3	1.00							
<i>Ep 4</i>	<i>1.25</i>							

\* Ejection angles are expressed regarding the vertical toward ground

$D_m$  and  $D_{sd}$  represent respectively the mean diameter and the standard deviation, while the  $(S)$  and  $(L)$  represent the used nozzle type, a standard and a low-drift nozzle, respectively. The used type of standard nozzle for the axial fan sprayer  $(S1)$  is Hollow cone nozzle – ATR Yellow – 10 bar and AXI 110 02 – 2 bar for the side-by-side sprayer  $(S2)$ . The used type of low-drift nozzle for the axial fan sprayer  $(L1)$  is TVI 110 02 – 10 bar and CVI 110 02 – 2 bar for the side-by-side sprayer  $(L2)$ .

#### II.2.1.4. Meteorological conditions

To illustrate the effect of meteorological conditions on the atmospheric dispersion of droplets, we set up two contrasted meteorological scenarios corresponding to an early morning treatment where the temperature and the wind speed are low and the relative humidity is high, and a midday treatment where the relative humidity is lower and the temperature and the wind speed are higher but remains within the limit of the spraying allowance for wind speed. Used parameters values while performing an early morning are  $1.5 \text{ m.s}^{-1}$  for wind speed at 3 m height, 80% for relative humidity and  $14^\circ\text{C}$  for the temperature. In the case of a midday treatment, the wind speed at 3m height, the relative humidity and the temperature were set equal to  $3.5 \text{ m.s}^{-1}$ , 40%,  $24^\circ\text{C}$ , respectively. The atmospheric stability condition was considered to be neutral. These values have been considered as representative of averaged meteorological conditions for fungicide applications in the South-East France. In both meteorological scenarios, the wind direction was set perpendicular to vine rows.

## II.2.2. Simulation design

### II.2.2.1. Simulation scenarios

Simulation scenarios were built based on previously presented factors, namely sprayers and nozzles types, growth stages, active matter proportions and the two combinations of three meteorological parameters and run for presented field configuration which results in sixty simulation scenarios (Table IV.4). Simulation duration was set equal to 15 min, which is the time scale at which generally spray drift occurs.

Table IV.4. Simulation scenarios combining four factors (the number between brackets refers to the number of levels considered for each factor)

Used sprayer and nozzle type (5)		Growth stage* (2)	Active matter proportion %** (3)	Meteorological parameters# (2)		
				Wind speed (m/s)##	Relative humidity (%)	Temperature (C°)
<b>Pneumatic</b>	No nozzle	<ul style="list-style-type: none"> <li>• Early (19)</li> <li>• Developed (57)</li> </ul>	<ul style="list-style-type: none"> <li>• 0.02</li> <li>• 0.2</li> <li>• 2</li> </ul>	<ul style="list-style-type: none"> <li>• 1.5 m.s<sup>-1</sup> – 80% – 14°C</li> <li>• 3.5 m.s<sup>-1</sup> – 40% – 24°C</li> </ul>		
<b>Axial fan</b>	Standard (ATR)					
	Low-drift (TVI)					
<b>Side-by-side</b>	Standard (AXI)					
	Low-drift (CVI)					

\* According to BBCH scale of vine phenological stages.

\*\* Percentage of active matter within tank mixture

# Combination of parameters

## at 3m height

### II.2.2.2. Output analysis

In order to evaluate the effect of each of the four factors on spray drift ground deposition and on bystander exposure, simulation results were averaged for all other factors and were compared regarding three different outputs: 1) distribution of pesticides at the time of application between what is deposited in field on the ground or intercepted by the canopy and what goes out of the field, 2) ground deposition downwind of the treated field, and 3) bystander exposition by inhalation and dermal deposition. All outputs were expressed in terms of percentage of application rate.

For spray drift ground deposition, results are presented at seven distances from the field edge along the central line  $y=0$ , namely at 3, 5, 7.5, 10, 15, 20 and 30 m. The exposure of bystanders was evaluated at seven different distances from the field edge (3, 5, 10, 20, 40, 80 and 160 m) also along the central line  $y=0$ . Note that the field edge corresponds to 1.25m from last row canopy edge for ground deposition and to 3m from last row trunk for exposure evaluation (Mercier, 2020; EFSA, 2022) (Fig. IV.1).

Exposure assessment was addressed in this study only for an adult that is assumed to be present at the defined distances during the whole spray drift process duration (15 min). Inhalation height and volume

and dermal deposition area were defined based on standard data commonly used in the literature (EFSA, 2022). Accordingly, inhalation height was set equal to 1.6 m and dermal deposition area was set to have 1.8m height, 0.6 m width and 0.2 m depth ( $V_{cubic-dermal}=0.216 m^3$ ). For inhalation, we considered that the individual could inhale particles passing through a volume of 0.5 m height and 1 m width and depth ( $V_{cubic-inhalation}=0.5 m^3$ ), centred on the inhalation height. For dermal exposure, we set an absorption coefficient equal to 10% in accordance with worst-case scenario (EFSA, 2014).

A sensitivity analysis regarding the needed number of particles to treat the designed plot was done prior to the simulations. This analysis revealed that 1,000,000 particles are necessary in order to limit the random variations of outputs between successive simulations trials. Accordingly, with 1,000,000 particles, the random variations were up to 2% at distances lower than 52 m from field edge and 5% for greater distances up to 250 m for both dermal and inhalation exposure estimation and from 10 to less than 30% for ground deposition at distances lower than 52 m.

#### II.2.2.3. Datasets used for output assessment

For assessing the simulated pesticide distribution within the treated field, the data reported by Hulin et al. (2021) for pneumatic and side-by-side (S) sprayers were used. In Hulin et al. (2021) the distribution between soil, vegetation, and air of a given a.i. (%) was estimated by experts by synthesizing the different existing experimental data within research organizations or professional institutes. Different distribution values are given depending on crops (distinguishing annual crops from low or high perennial ones), type of sprayers used according to the crop considered and the stage of development of the crop (see Table S.B.1, Annexe B). As Hulin et al. did not specify the meteorological conditions and the proportion of active ingredients, we considered for the simulations a range of conditions or values for these simulation inputs.

For assessing the simulated bystanders exposure, the experimental data of Mercier (2020) and the modelling data of Browse and German models as mentioned by Mercier (2020) were used (see Table S.B.2., Annexe B). Briefly, Mercier (2020) carried out an experiment to measure direct dermal and inhalation exposure of bystanders and residents during vine foliar application using a side-by-side sprayer with low drift nozzles. The a.i. concentration was 0.2%. For both German and BROWSE models estimations, Mercier (2020) compared his results to those of the two models at 5 m from field edge. For comparison with these data, we simulated the case of an axial fan sprayer and an a.i. percentage equal to 0.2%. In the absence of information concerning weather conditions or canopy growth stage, several simulations were performed considering a range of possible conditions and stages. The comparison with these model predictions was performed only at 5 m, bearing in mind that for BROWSE models data rather represent orchard conditions than vineyard conditions.

Finally, for the comparison with dermal exposure, we used the corresponding absorption percentage for the used a.i. in Mercier (cymoxanil) in order to get the appropriate exposure value for dermal deposition. Accordingly, the absorption percentage was set equal to 2.5% (Federal Register, 2003).

### **III. Results**

#### **III.1. Variation of within field pesticide distribution**

Figure IV.3, represents the simulated effects of sprayer and nozzle types, active ingredient percentage, canopy growth stage and meteorological conditions on the distribution of pesticides during application between 1) what is intercepted by the targeted canopy, 2) what is deposited on the field ground and 3) what goes beyond the treated field. All quantities are expressed as a percentage of the applied mass. Results obtained with all simulation runs are presented as boxplots.

##### **a. Sprayer and nozzle effect**

From the Fig. IV.3, we can see that the spray distribution in the different compartments within the treated field is significantly different from one sprayer to another and strongly varies for a given sprayer when using a low-drift nozzle instead of a standard one.

For *canopy interception*, the highest interception percentage of applied mass was observed for the side-by-side sprayer with standard (*S*) and low-drift (*LD*) nozzles, followed by the pneumatic sprayer then the axial fan *LD* sprayer and finally the lowest percentage was observed for the axial fan *S* sprayer.

This difference in interception percentages between sprayers can be firstly attributed to ejection angles and then to the velocity of the air assistance or the type of nozzle used. The ejection angle plays a principal role as it determines the trajectory of droplets for the first few meters especially at high ejection velocity. This behaviour was typically observed for the side-by-side sprayer where ejection points are spraying directly toward the canopy with an angle of 90° from the vertical. For the other sprayers, ejection angles are more distributed towards the canopy and the upper ejection points may generate some losses towards the atmosphere. However, this appeared to be corrected in part by the high speed air assistance that favours droplet penetration in the canopy and is used in the pneumatic sprayer, with in addition an air assistance directed towards the canopy in the case of the pneumatic sprayer which may explain its highest efficiency in canopy interception. The effect of using *LD* nozzles improved canopy interception of an axial fan sprayer but does not modify the ranking between sprayers in terms of canopy interception efficacy.

For *ground deposition*, deposits by all sprayers were rather moderate except for axial fan sprayer for which deposits were markedly the highest. Using *LD* nozzles on the axial and side-by-side sprayers increases slightly the ground deposits for both sprayers. As for canopy interception, these simulation results are firstly linked to ejection angles but also to the diameter of generated droplets that is different

for S and LD nozzles. As the axial fan sprayer has its lower nozzle spraying toward the ground, this favours the deposition of droplets to the ground, while for the pneumatic sprayer, even if the upper ejection points are spraying towards the ground, they are spraying over the canopy with smaller droplet size, which favours rather the dispersion of particles than their deposition to the ground. Similarly, LD nozzles generate bigger particles with bigger settling velocities, which increases their deposition to the ground.

For the *fraction leaving the field*, the pneumatic sprayer is the one contributing the most to outfield losses, followed by the axial fan sprayer and the side-by-side sprayer. This is due to the high speed of the air assistance of the pneumatic sprayer, the smaller droplet diameters and the position of the upper ejection points that spray the adjacent rows. The same issue concerning nozzle orientation is implicated for the axial fan sprayer that also shows rather high outfield losses. As could be expected, the side-by-side sprayer, given its ejection angles directed towards the canopy, presents the lowest outfield losses. Finally, it is worth to observe the very positive influence of the LD nozzles that reduce outfield drift by almost 50% for both the axial fan and the side-by-side sprayers, which is in concordance with experimental data (EFSA, 2022). Indeed, as these nozzles are generating bigger droplets, they rapidly deposit and, thus, prevent outfield losses.

These distribution results for the pneumatic sprayer and the side-by-side sprayer (S) are in general agreement with the range of values provided by Hulin et al. (2021) with similar ranking and order of magnitude of canopy interception, infield ground deposition and outfield dispersion between the pneumatic and side-by-side sprayers. Indeed, regarding the *canopy interception*, our simulation results ranged between 36 to 67% and 59 to 78% for the pneumatic and the side-by-side sprayers respectively, against 20 to 60% and 40 to 80% in Hulin et al. (2021) for an early and a normal fungicide and insecticide application in low perennial crops. For *infield ground deposition*, modelled results range between 1 - 18% and 2 -24% for the pneumatic and the side-by-side sprayers respectively, versus 10 - 30% and 10 - 20% respectively in Hulin et al. (2021). Finally, for the *fraction leaving the treated field*, results are ranging from 26 - 58% and 18 - 34% for the pneumatic and the side-by-side sprayers respectively, against 30 - 50% and 10 - 40% respectively in Hulin et al. (2021).

#### **b. [Active matter percentage effect](#)**

The percentage of a.i. within the tank mixture slightly affected the distribution of pesticides (Fig. IV.3). Indeed, when the a.i.% decreased, the simulated values of *canopy interception* and *infield ground deposition* tended to decrease while the fraction leaving the treated field tended to increase. This effect was more marked for this latter fraction with an increase of the percentage of outfield losses by almost the double between the highest and the lowest a.i.%, namely 2 and 0.02%. This is related to the role of a.i.% on droplet minimum diameter after water evaporation, which is assumed to be proportional to the



a.i.% in the tank mixture. So, lower is the a.i.%, lower is the droplet diameter after water evaporation, and in turn larger are droplet driftability and losses outside the treated field.

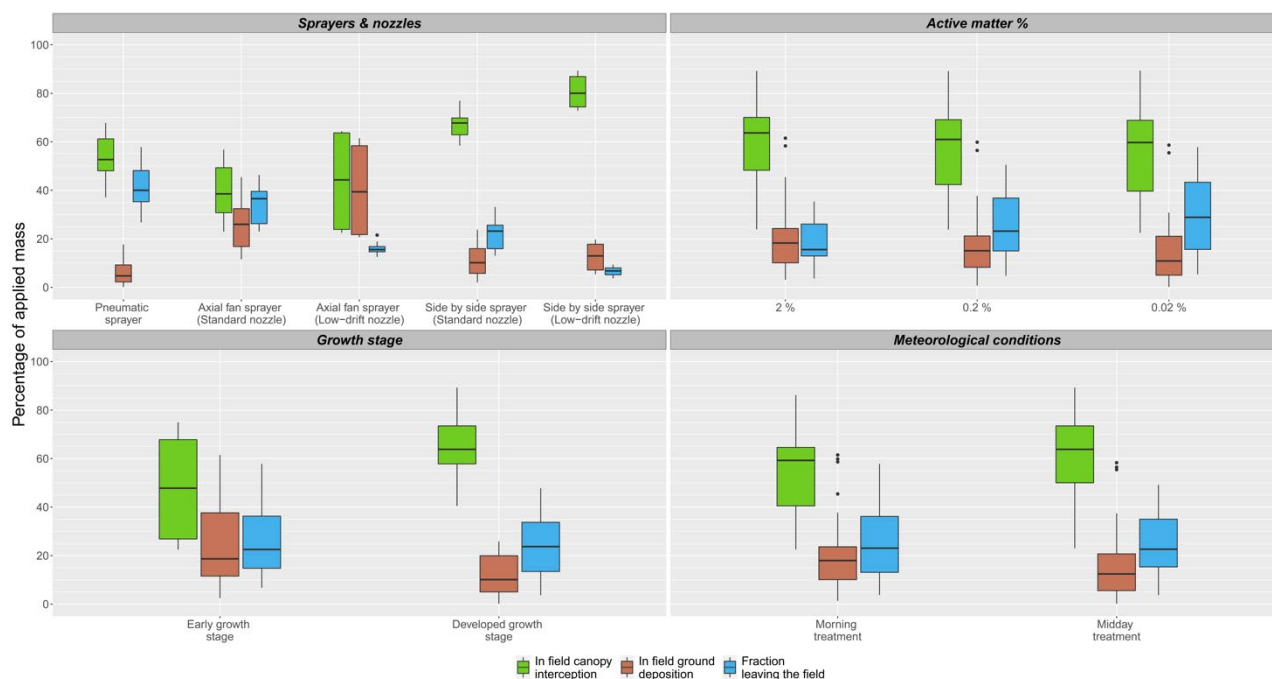
c. Growth stage

Fig. IV.3 shows that growth stage affected mainly canopy *interception* and infield *ground deposition* but not significantly the fraction leaving the field (Fig. IV.3): interception was higher in the developed than the early growth stage while the *infield ground deposition* is lower. The suppression of some nozzles for the early growth stage certainly explains why drift remained almost similar between the two growth stages. Additionally, for the developed growth stage, the row height, width and density are indeed higher than for the early growth stage, which increases particle interception probability. It is striking to see in Fig. IV.3 that, between the two growth stages, the increase in canopy interception is approximately counterbalanced by the decrease in within-field ground deposition.

d. Meteorological condition

Between a morning and midday treatment, it appears that meteorological conditions had only a slight effect on the distribution of pesticides (Fig. IV.3). Interception slightly increased by about 5% in a midday application as compared to a morning one and in contrast infield deposition decreased with almost the same amount. No significant effect was observed for the fraction leaving the field.

Figure IV.3. Influence of sprayer and nozzle types, active substance percentage, canopy growth stage and meteorological conditions on pesticide distribution during application expressed as a percentage of application mass. The infield canopy interception and ground deposition and outfield losses variation are represented by boxplot of different colours, namely, green, brown and blue, respectively.



### III.2. Sedimentary spray drift

Figure IV.4 represents the effect of sprayer and nozzle types, active ingredient percentage, canopy growth stage and meteorological conditions on sedimentary spray drift downwind to the treated field. Results are represented by the average deposits obtained for all runs at each distance and expressed as the percentage of applied mass.

#### a. Sprayers and nozzles effect

As the pneumatic sprayer and the axial fan sprayers with a standard nozzle showed a high contribution to outfield losses (Fig. IV.3), a high outfield deposition percentage via sedimentary drift was also observed for these sprayers (Fig. IV.4). At all distances, these two sprayers showed higher deposition percentage than the others except at 3 m where the side-by-side *S* sprayer showed the highest value (6.9%) followed by the pneumatic (6.5%) and the S-axial fan (5.7%). The use of *LD* nozzle for the side-by-side sprayer decreased significantly sedimentary drift by almost one third at 3m and by 5 at 5m. The same reduction effect was observed for the axial fan equipped with *LD* nozzle, however, the reduction was greater at 3m than at 5m, almost by 3 and 2 times respectively. For all sprayers and conditions, sedimentary deposits decreased with the distance to the treated field, except for the axial fan equipped with *LD* nozzle, for which an increase in deposition was observed until 15 m followed by a decrease further away. This specific behaviour is induced by the ejection angles of upper nozzles of the axial fan that are directed towards the atmosphere. As droplets are bigger than when using an *S* nozzle, their ejection towards the atmosphere propels them out of the field within a few seconds at larger distances than it does for smaller droplets. This will consequently give these particles a bigger inertia, making them depositing at greater distances, which can explain the low deposition at 3 m and its increase with distance. Over 15 m, it is rather smaller droplets that are depositing, at these distance droplets have already lost their inertia, thus their deposition is mainly driven by atmospheric turbulence rather than ejection initial conditions (sprayer and nozzle type). This behaviour has also been observed in experimental data (Polveche et al., 2011).

#### b. Active matter percentage effect

From Fig. IV.4, we can see that when a.i. percentage increased the outfield deposition percentage increased. This is correlated to the fact that, as already discussed above, the a.i. % influences particle diameter after evaporation. After evaporation, high a.i. % results in bigger droplets, thus higher outfield deposition percentages at short distances while lower a.i. % results in smaller droplets and lower outfield deposition at the same distances. For 2% a.i. %, the decrease of outfield sedimentary spray drift with distance is significantly marked, a reduction of up to almost 4 times is observed between 2 and 30 m, while it is significantly lower for the other a.i. %: 3 and 1.5 times for 0.2 and 0.02% of a.i. % respectively. This is induced by the fact that bigger particles deposit rapidly within few meters, while smaller ones tend to disperse at longer distance under the influence of atmospheric turbulence.



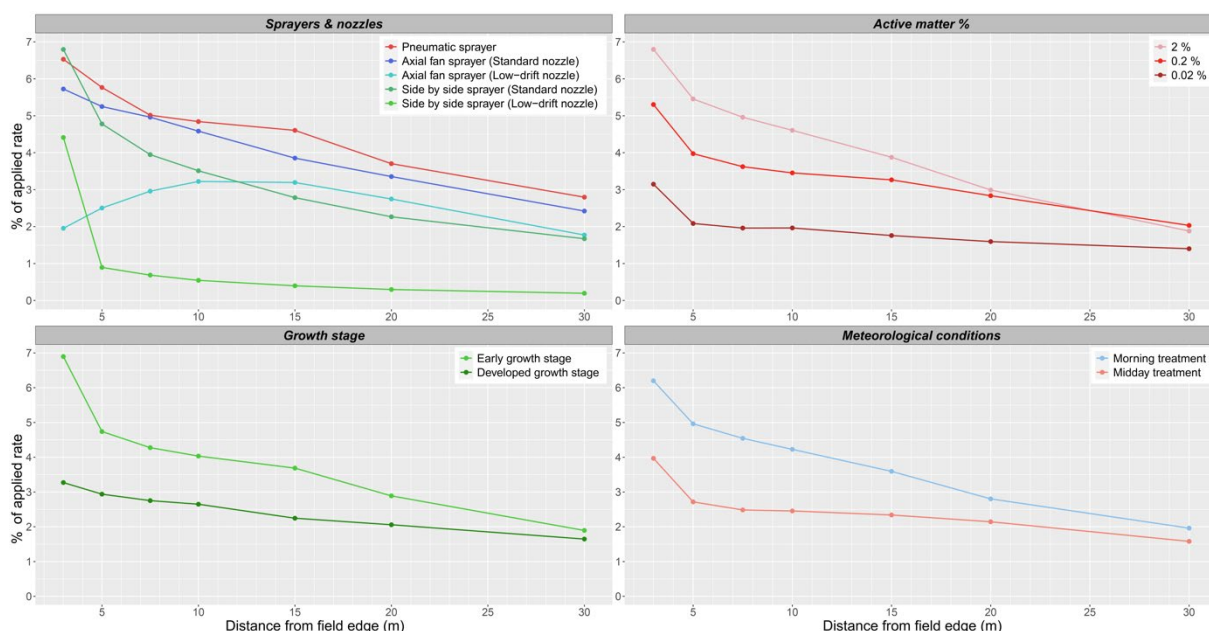
c. Growth stage

Growth stage affects outfield sedimentary spray drift: for early growth stage, deposition next to the field up to 30 m was larger than for developed growth stage by almost the double and the decrease with distance is much marked for the first one. This behaviour may be attributed to the fact that, for the developed growth stage, it is almost smaller particles that pass through the canopy and leave the field.

d. Meteorological conditions

Morning application led to higher outfield deposition than midday one, especially at shorter distance (< 20 m) (Fig. IV.4). Then, for further distances, the effect became less significant, the deposition difference was small at 30m as compared to almost 1/3 less at 3 m. Weather conditions drive droplet evaporation rate: at noon the temperature is high and the relative humidity is low which increases evaporation, thus results in smaller particles that will less deposit at short distance (< 20 m) and are more easily transported by wind. Deposition percentages started to be close at distances greater than 20 m since only small particles, that had already evaporated, reach this distance under the effect of wind flow (Gil et al., 2008).

Figure IV.4. Influence of sprayer and nozzle types, active substance percentage, canopy growth stage and meteorological condition on outfield sedimentary spray drift, expressed by the mean deposition of percentage of application mass.



### III.3. Bystander exposure through inhalation and dermal deposition

#### III.3.1. Exposure through inhalation

Figure IV.5 represents the effect of sprayer and nozzle types, active ingredient percentage, canopy growth stage and meteorological conditions on the variation of bystander exposure to spray drift through

inhalation pathway following the distance to the field edge. Results are represented by the average exposure at each distance expressed by the percentage of applied mass that is inhaled.

a. [Sprayers and nozzles effect](#)

From Fig. IV.5, we can see that the sprayer that led to the largest exposure of bystanders by inhalation was the pneumatic sprayer with 0.35% and 0.12% of applied mass at 3 and 160 m from field edge respectively, followed by the axial fan (*S*) sprayer with 0.29 and 0.08% at the same distances respectively. For the latter sprayer, we can see that the use of *LD* nozzles decreased exposure through inhalation by more than a half with an exposure equal to 0.075% of applied mass at 3 and 5 m. This reduction by almost 50% was also observed in trials used as reference for estimating bystanders exposure when using *LD* nozzles (EFSA, 2022; Mercier, 2020). The use of the side-by-side sprayer with and without *LD* nozzles showed a significant reduction of exposure by inhalation compared to other sprayers (with an exposure of 0.003% and  $5 \times 10^{-5}\%$  for *S* and *LD* nozzles respectively). The exposure by inhalation produced by a side-by-side sprayer is about 2 to 4 orders of magnitude less than that produced by a pneumatic sprayer, depending on whether *S* or *LD* nozzles, are used.

This important difference in exposure, which is in agreement with the results shown in Fig. IV.1 regarding the effect of sprayer types on the fraction of spray leaving the field, may firstly be due to the difference in ejection angles between the three sprayers. Compared to the side-by-side sprayer, the upper ejection points of axial fan and the pneumatic sprayers may generate more losses outside the treated field and thus a greater exposure by inhalation due to the fact that they are slightly directed toward the atmosphere or are spraying at 2.5m high over the canopy, respectively. The second influencing factor is the droplet initial diameter that is much bigger when using (*LD*) nozzles, which will result in more very short-range deposition than an aerial exposure via inhalation.

b. [Active matter percentage effect](#)

For the a.i. %, in the contrary to sedimentary deposition, the lower the a.i. % is, the greater the exposure of bystanders by inhalation is Fig. IV.5. This is related to the influence of a.i.% on the minimum droplet size after evaporation. As particles with higher a.i.% (e.g. 2%) have a bigger diameter after evaporation than those containing a small amount of a.i. (e.g. 0.02%), these particles are more subject to deposition at very short distances rather than to remain in the air for longer period.

c. [Growth stage](#)

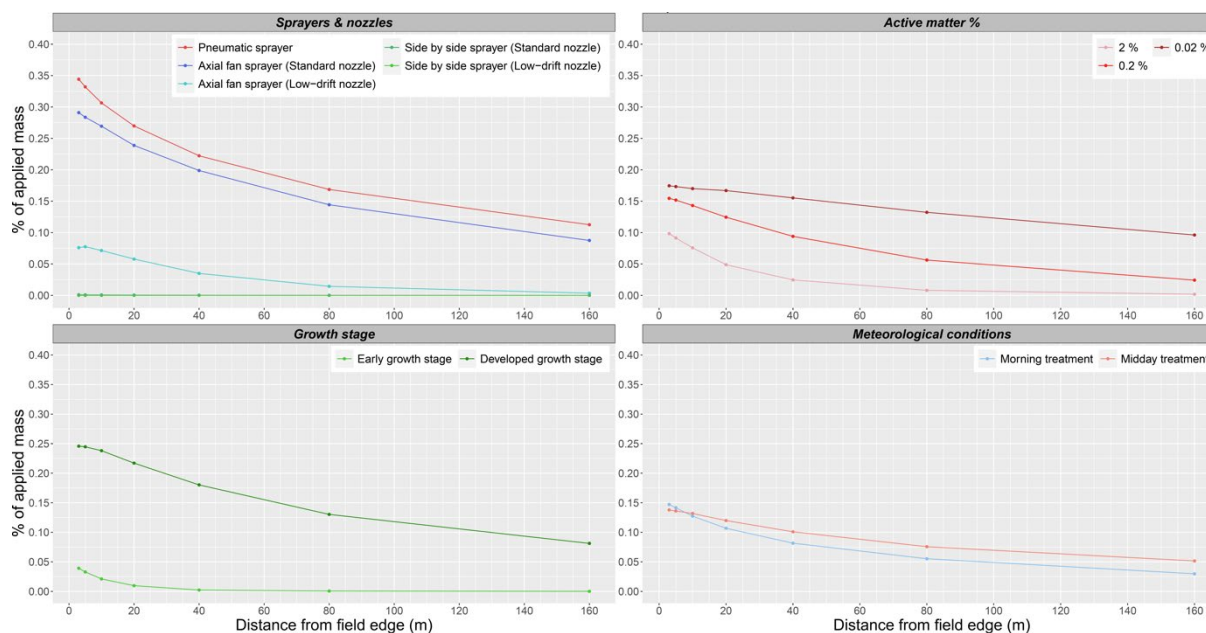
The effect of canopy growth stage on the exposure of bystanders by inhalation was inverted as compared to its effect on the sedimentary drift. Indeed, exposure of bystanders was larger at developed than at early growth stages (Fig. IV.5). As seen in Fig. IV.3, the mass of pesticide leaving the field was shown to be slightly larger for early than for developed growth stages. So only differences in size and/or vertical

distribution of the droplets leaving the field can explain the large difference in bystander exposure by inhalation between the two growth stages. Indeed, in case of developed canopies, large diameter particles are more at risk to be intercepted and thus the proportion of particles with small diameters that leave the canopy is larger, which favours transfer in the air and exposure by inhalation. In case of small canopies, the mass leaving the field also includes a marked proportion of large particles that are prone to deposit at low distance and do not contribute to exposure by inhalation. Besides, for developed canopies, most of the particles leaving the field pass at high heights whereas for less developed canopies they can pass at lower heights. Since the inhalation height at 1.6 m is rather high as compared to the early stage canopy, it is likely to collect more particles in case of the developed canopy. To these explanations, it can be added the effect of the canopy characteristics on the atmospheric turbulence and wind profile in and above the canopy. Developed canopies are denser and taller (higher displacement length), which induces a bigger acceleration of wind profile over the canopy and thereby favours the dispersion at longer distances especially as we are considering the turbulence over the field to be homogenous.

d. Meteorological conditions

The effect of meteorological conditions on the exposure of bystanders by inhalation is reversed and less significant compared to outfield sedimentary drift. In general, bystanders are more exposed when applying pesticides at noon with approximately 1.3 times comparing to a morning application at all distances except at 3 and 5 m Fig. IV.5. At 3 and 5 m, exposure percentage by inhalation is quite similar at both treating periods, with a slightly higher exposure at morning treatment than at midday. This is linked to the fact that at noon evaporation is greater than in the morning, thus under the effect of evaporation, particles will have smaller diameter at noon, inducing so a greater dispersion at longer distances and bigger lifetime in the atmosphere, which increases the exposure by inhalation. At 3 and 5m and under morning conditions particles evaporate less, keep so larger diameters, deposit faster and contribute in turn less to long-range exposure.

Figure IV.5. Influence of sprayer and nozzle types, active ingredient percentage, canopy growth stage and meteorological conditions on the variation of bystander exposure to spray drift through inhalation with down field distance. Expressed by the average exposure at each distance as the percentage of applied mass



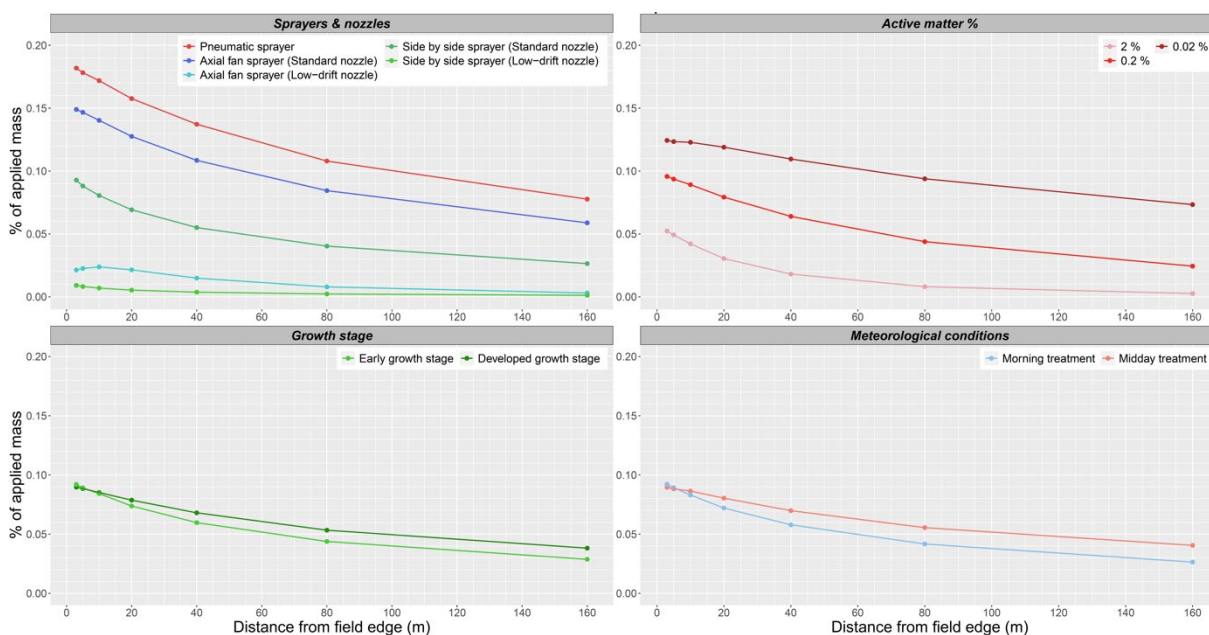
### III.3.2. Exposure through dermal deposition

Figure IV.6 presents the effect of sprayer and nozzle types, active ingredient percentage, canopy growth stage and meteorological conditions on the variation of bystander exposure to spray drift through dermal deposition pathway with down field distance. Results are represented by the average exposure at each distance and expressed in the percentage of applied mass.

From this figure, we can see that globally the simulated effect of the four factors on the exposure of bystanders via dermal deposition was quite similar to those of exposure by inhalation except for one sprayer and a slight difference for growth stage effect. Unlike for the exposure via inhalation, the side-by-side *S* sprayer induced 4 times more exposure by dermal deposition than axial fan *LD* sprayer. This can be due to the fact that unlike inhalation, dermal exposure occurs over a larger volume where particles can deposit from ground up to 1.8 m (adult mean height), which means that for dermal deposition, the vertical gradients of pesticide concentration have less importance than for inhalation which apply on a more punctual height.

Even if canopy growth stages had globally the same effect on bystanders exposure by dermal deposition than by inhalation, the difference induced by the two growth stages is much less marked for dermal exposure and at very short distances (3 and 5m) it is nearly null. This also can be related to the difference in the dimensions of deposition/inhalation volumes and the importance of vertical gradients for inhalation and not for dermal deposition.

Figure IV.6. Influence of sprayer and nozzle types, active ingredient percentage, canopy growth stage and meteorological conditions on the variation of bystander exposure to spray drift through dermal deposition with down field distance. Expressed by the average exposure at each distance in percentage of applied mass.



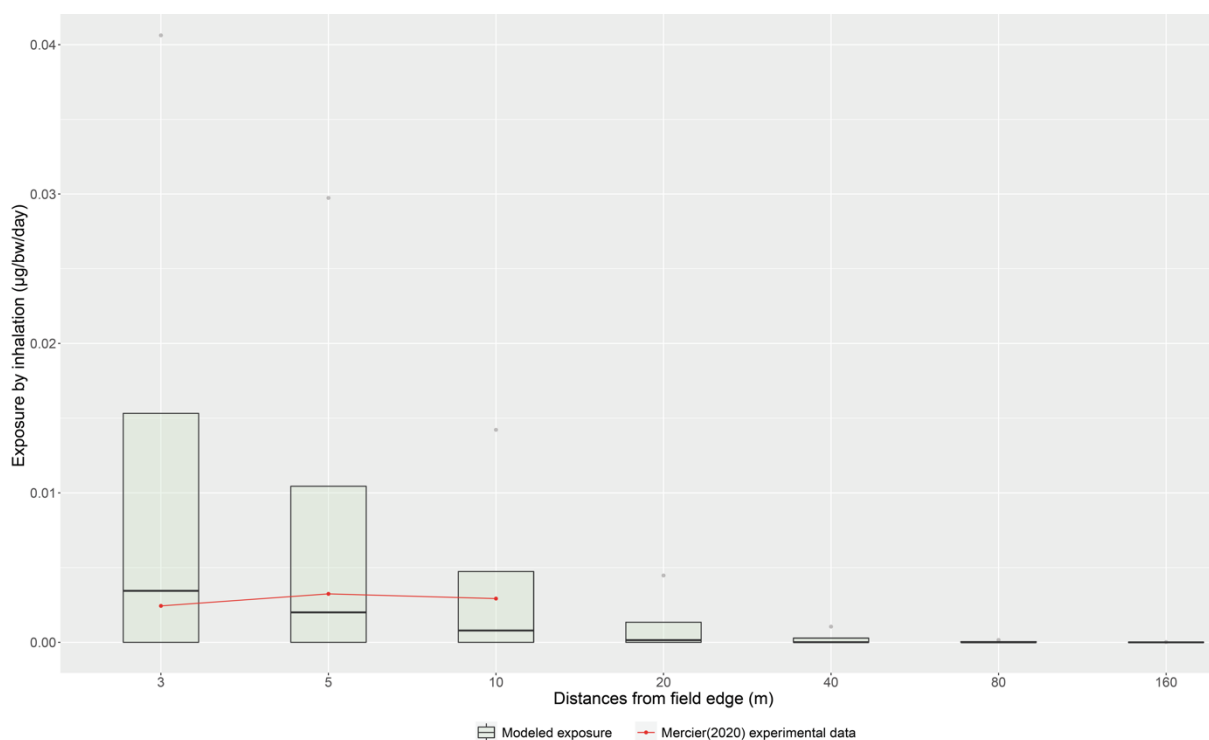
### III.4. Confrontation with available data

#### III.4.1. Confrontation to Mercier experimental study

Figures IV.7 and IV.8 present the comparison of modelling results of the exposure of bystanders through inhalation and dermal deposition respectively, for the side-by-side (*LD*) sprayer and 0.2% a.i. percentage with the experimental data of Mercier (2020). Modelled results are represented with the green boxplots while experimental data are represented with red dots and line. Results are expressed in  $\mu\text{g}$  of body weight (bw) per day.

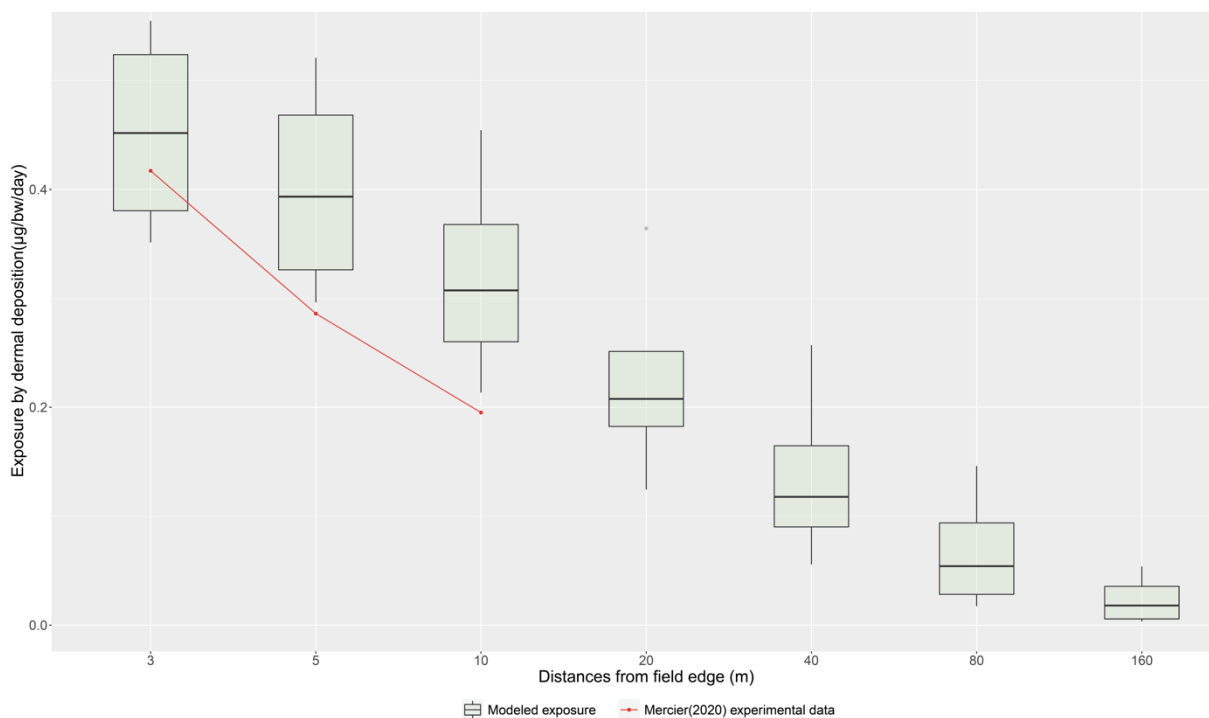
From Fig. IV.7, we can see that at the 3 distances (3, 5 and 10 m) experimental data for bystanders exposure via inhalation were within the 25<sup>th</sup> and 75<sup>th</sup> quantiles of modelled results for closer simulation scenarios. This means that the model represents in a fairly satisfactory way the exposure through inhalation at these distances.

Figure IV.7. Confrontation of modelling results of the exposure of bystanders through inhalation to the experimental data of Mercier (2020). Comparison of modelling results is for scenarios of the side-by-side (LD) sprayer and 0.2% a.i. percentage. Results are expressed in  $\mu\text{g}$  of body weight (bw) per day.



For the exposure through dermal deposition at the same distances, modelled results had a slight tendency to overestimate exposure at 5 and 10 m while at 3m the prediction is within the 25<sup>th</sup> and 75<sup>th</sup> quantiles (Fig. IV.8). This can be related to the fact that in our model we are maximising deposition over the body by considering that all particles that are passing through the defined volume are deposited. In reality, there is a deviation of the air around the human body proportional to its width, which leads to the deviation of particles having not enough inertia to impact the body from their initial trajectory (Aylor, 1982; Gil, 2007). This deviation is more pronounced for small droplets, which contributes to the overall concentration increasing with distances. That behaviour explains why the discrepancies between modelling results and data increase with distances.

Figure IV.8. Confrontation of modelling results of the exposure of bystanders through dermal deposition to the experimental data of Mercier (2020). Comparison of modelling results is for scenarios of the side-by-side (*LD*) sprayer and 0.2% a.i. percentage. Results are expressed in  $\mu\text{g}$  of body weight (*bw*) per day



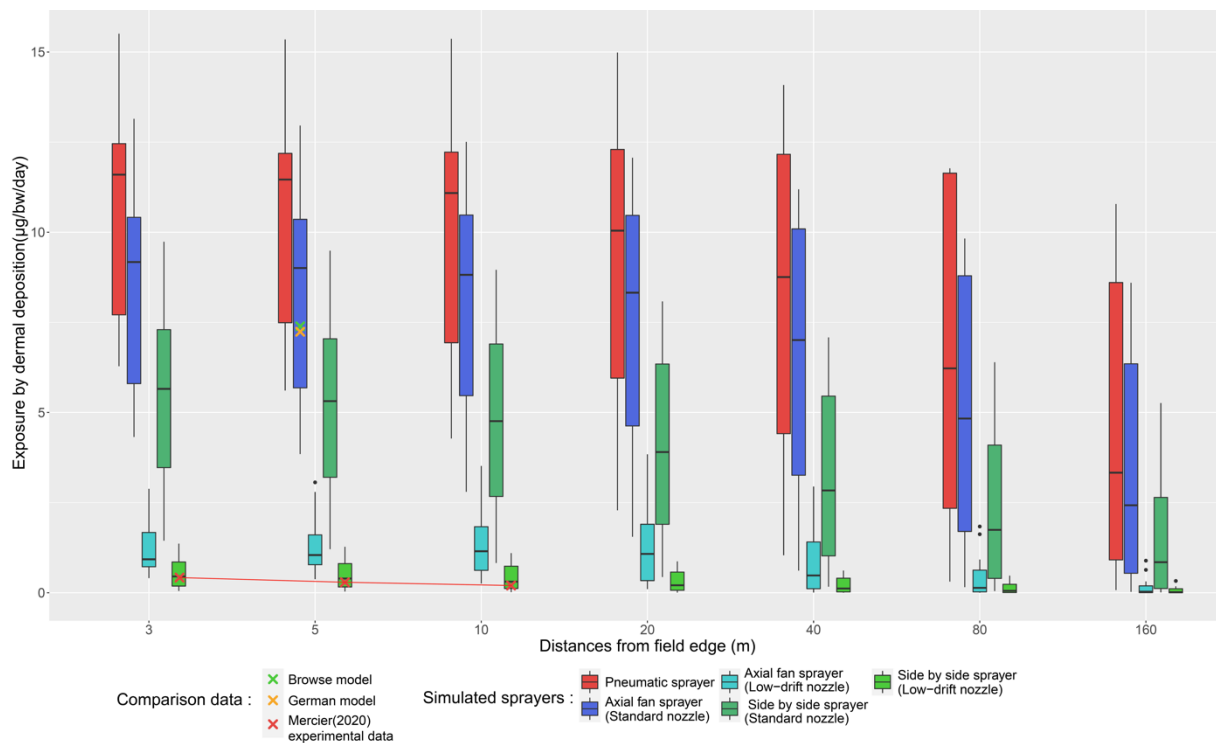
### III.4.2. Global confrontation to Mercier experimental study, to the BROWSE and to the German models

Figures IV.9 and IV.10 present a global comparison of modelling 1) results of exposure through dermal deposition and inhalation respectively, for all sprayers with (*S*) and (*LD*) nozzles and all simulation conditions with 2) the experimental data of Mercier (2020) and 3) simulated data by the BROWSE and German models. Modelled results are represented in boxplots with different colours, where each colour corresponds to a specific sprayer, while the data considered for comparison are represented by different coloured crosses. Results are expressed in  $\mu\text{g}$  of body weight (*bw*) per day.

For the exposure via dermal deposition, the model is able to represent the exposure related to the appropriate sprayer, where Mercier results are well positioned at the side-by-side (*LD*) sprayer and the modelled data of the German model and the BROWSE model at the axial fan (*S*) sprayer Fig. IV.9. These results suggest that the model is able to simulate the effect of sprayers and used nozzles on the exposure of bystanders through dermal deposition.



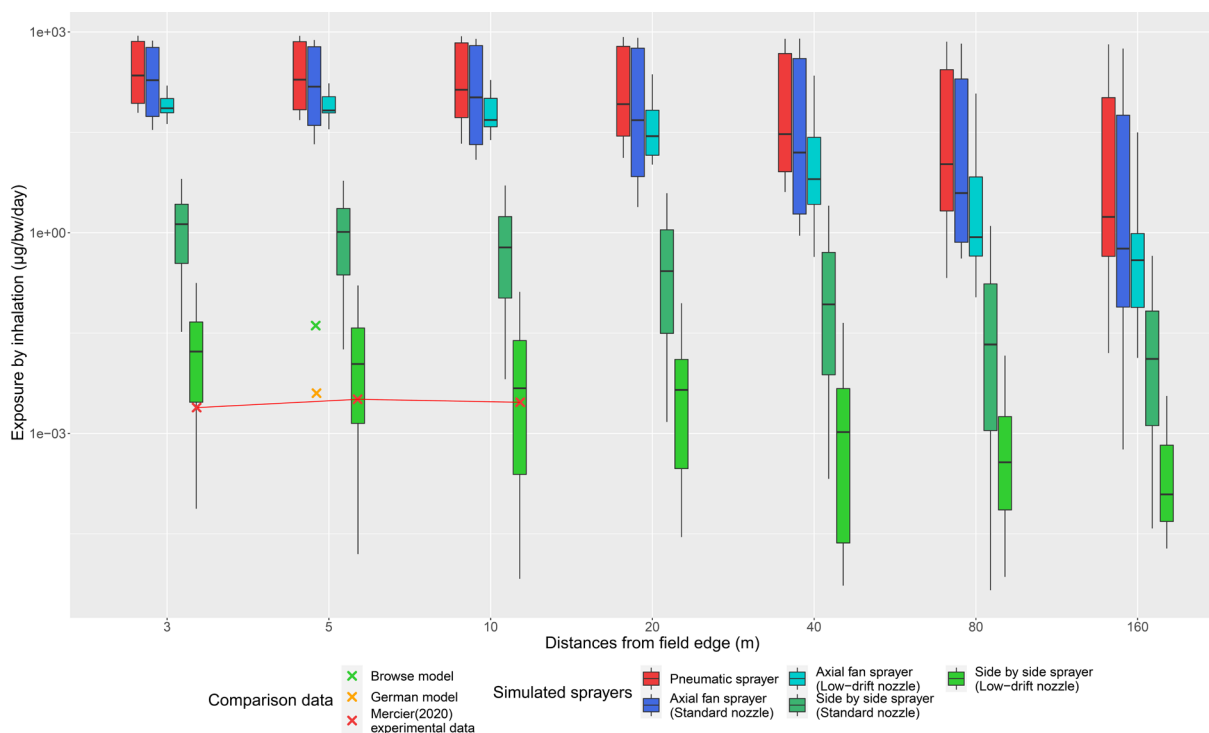
Figure IV. 9. Confrontation of modelling results of the exposure of bystanders through dermal deposition to the experimental data of Mercier (2020) and modelling data of BROWSE and German models. Comparison of modelling results is made with all simulation scenarios. Results are expressed in  $\mu\text{g}$  of body weight (bw) per day.



From Fig. IV.10, as previously presented, we can see that the model is able to represent the exposure via inhalation for the experimental data of Mercier while using the side-by-side sprayer equipped with (*LD*) nozzle. The discrepancy between ADDI-SprayDrift model and BROWSE and German models (where an axial fan sprayer with (*S*) nozzle is used ) for exposure of bystanders through inhalation can be related to the possible difference in spraying conditions (weather conditions, canopy growth stage and the a.i. percentage within the tank mixture) as exposure through inhalation is more sensitive to these conditions and to the vertical gradient of concentration of pesticides in the air compared to dermal deposition.



Figure IV.10. Confrontation of modelling results of the exposure of bystanders through inhalation to the experimental data of Mercier (2020) and modeling data of BROWSE and German models. Comparison of modelling results is made with all simulation scenarios. Results are expressed in  $\mu\text{g}$  of body weight (bw) per day.



## IV. Discussion and conclusion

The new multi-source version of ADDI-spray Drift model gives the possibility to assess the effect of sprayer and nozzle types, active substance concentration within the tank mixture, canopy growth stage and meteorological conditions on pesticide distribution within the treated field, on sedimentary drift and on the exposure of bystanders via inhalation and dermal deposition of spray droplets while treating the entire field at once.

Model's predictions for pesticide initial repartition and bystander exposure via inhalation and dermal deposition show a good agreement with experimental data for the considered sprayers with however a slight overestimation of exposure of bystanders through dermal deposition, associated to the maximisation of deposition over the body.

Through the analysis of the effect of the four tested factors on pesticide distribution within the treated field, outfield sedimentary spray drift and bystander exposure of spray drift droplets, some main effects were identified:

- Among the tested sprayers, the side-by-side sprayer seems to confer a good equilibrium between application efficiency in terms of interception by the foliage and losses reduction in and out of the

field, compared to the pneumatic and the axial fan sprayers. It also has a significant reduction effect on exposure of bystanders through inhalation. However, its effect on sedimentary spray drift and exposure via dermal deposition is less marked which may be linked to the interaction with the other factors and to the fact that dermal deposition is less influenced by pesticide vertical gradient of concentration as inhalation does. Then, even if the pneumatic sprayer seems more efficient than the axial fan sprayer as it allows a better interception by the foliage and less infield loss (ground deposition), it seems to generate more outfield losses. However, outfield sedimentary drift and bystander' exposure are very close for both sprayers. If the use of low-drift nozzles seems to reduce the outfield losses for the axial fan and side-by-side sprayers, it increases infield losses to the soil, while their effect on crop efficiency is secondary comparing to sprayer design.

- Active substance percentage within the tank mixture effect seems to influence more the proportion of outfield losses rather than the distribution infield, where the increase of a.i. percentage decreases the exposure via both dermal deposition and inhalation. However, it increases the sedimentary drift at short distances (up to 30m).
- In opposition to the percentage of a.i., canopy growth stage influences more pesticide infield repartition between canopy interception and ground deposition than outfield losses, given the reduction in the number of nozzles for the early growth stage. In spite of this, due to the increase of the dispersion of small particles caused by the acceleration of wind profile induces by the presence of denser and higher vegetation (canopy developed growth stage) and as inhalation is more sensitive to the droplet concentration vertical gradient, canopy growth stage influences nevertheless the exposure of bystanders by inhalation.
- The meteorological conditions, as here tested to represent morning or midday applications, seem to have a little effect on infield repartition of pesticides and a more significant effect on sedimentary spray drift and its evolution as function of distance.

Even if the influence of an appropriate adaptation of spraying device to treated canopy characteristics (ejection angle and height) is evident for increasing application efficiency here linked to the crop interception and reducing infield and outfield losses, establishing a hierarchy of the relative contribution of each of the tested factors to sedimentary drift and bystanders' exposure is more difficult. This difficulty is linked to the variability of the effect of tested factors, which in turn is related to the tide of interactions existing between them. Further analysis of the results is required as well as additional tests to establish such a hierarchy of effects

In the last report of the European Food Safety Authority (EFSA, 2022), a lack of experimental or modelling data have been highlighted regarding the effect of application conditions on the exposure of bystanders and residents through droplet spray drift. It also emphasizes the impossibility of comparing the few existing data because of the difference in application conditions in terms of meteorological

conditions, sprayer types and also crop types. The novelty of this approach is the ability of ADDI-SprayDrift model to reproduce the effect of different factors on pesticide distribution infield, sedimentary drift and the exposure of bystanders via inhalation and dermal deposition of spray droplets. ADDI-SprayDrift model is also applicable to assess child or resident exposure by considering appropriate parameters. As long as sources and canopy characteristics are correctly provided and after appropriate evaluation, the model could be used for other crop types.

Despite the good behaviour and predictions of the model, some improvements are necessary in particular concerning the dermal exposure of bystanders where the efficiency of impaction of droplets over the human body must be integrated by including the effect the latter on the surrounding turbulence.

The evaluation of the effect of complex spraying device like the recycling panels sprayer is not possible in the current version of the model. Further improvement for the source characterisation and on turbulence are necessary in order to consider the specific turbulence induced by the air assistance between panels and the effect of sprayer forward velocity on it.

Additional validation of the model for exposure assessment and for pesticide outfield vertical profile at different distances is necessary using more exhaustive experimental data.

Finally, model results depend on the number of released particles. Accordingly, it is important to use the appropriate particles number in order to reduce the variability related to the stochastic character of the model and to get a good balance between precision and simulation time.

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## *Conclusion générale*

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## Conclusion générale

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Face aux enjeux concernant la réduction des pertes de pesticides dans l'environnement et leurs impacts sanitaires, écototoxicologiques et sur la baisse de la biodiversité, une bonne compréhension des processus majeurs impliqués dans leur transfert dans les différents compartiments environnementaux et dans l'exposition des différents êtres vivants présents dans ces derniers est nécessaire. Dès l'application, la distribution des pesticides entre la fraction atteignant la cible, celle perdue au sol dans la parcelle ou celle transportée par le vent en dehors de cette dernière joue un rôle prépondérant sur le devenir des pesticides dans l'environnement. La détermination de cette distribution est un premier pas vers une compréhension globale des possibilités de transfert ultérieurs, dans le sol, vers les eaux mais aussi dans l'atmosphère. Cette thèse s'inscrit dans la perspective d'étudier la contribution des transferts atmosphériques au moment de l'application par pulvérisation au devenir global des pesticides dans l'environnement en recherchant à identifier les différents processus impliqués dans leur distribution lors de leur application et leur transfert par dérive des gouttelettes en aval de la parcelle traitée. Nous avons visé deux objectifs :

- 1) Développer un modèle mécaniste décrivant la distribution de pesticides lors de l'application, la dérive des gouttelettes en dehors du champ traité et l'exposition des passants par dépôt cutané et inhalation en fonction de la distance à la parcelle traitée, en essayant de considérer l'ensemble des facteurs gouvernant les processus impliqués, tout en étant applicable pour des approches de spatialisation à des échelles territoriales.
- 2) Évaluer dans le cas de pratiques en viticulture, après une phase de confrontation des prédictions du modèle à des données expérimentales, l'influence sur ces processus, de facteurs tels que le matériel de pulvérisation utilisé (pulvérisateur, buse), le stade de développement du couvert traité et les conditions météorologiques.

### **I. Développement du modèle d'interception par le couvert et de dérive**

Le développement d'un nouveau modèle s'est avéré utile car l'étude bibliographique menée sur les principaux modèles mécanistes utilisés dans la communauté scientifique a révélé une absence d'outil de modélisation générique incluant les principaux processus impliqués dans la prédiction à la fois de la distribution des pesticides lors de leur application, la dérive sédimentaire (dépôt de gouttelettes à la

surface en aval de la parcelle traitée) mais également la dérive aérienne (gouttelettes restant dans l'atmosphère à une distance donnée en aval du champ traité).

Nous avons alors développé **un modèle mécaniste, conceptuellement générique** et parcimonieux en termes de mécanismes représentés qui répond aux lacunes soulignées dans la littérature. *ADDI-SprayDrift* est un modèle lagrangien de type marche aléatoire considérant les principaux processus impliqués dans la dérive et la distribution des pesticides lors de l'application à savoir l'émission des gouttelettes, leur transport atmosphérique, évaporation et dépôts sur les surfaces, que cela soit foliaire lors de leur interception par les cultures traitées ou au sol, dans la parcelle traitée et en aval de la parcelle traitée. Son caractère lagrangien permet d'avoir une information précise sur la position individuelle des particules, ici les gouttelettes de pulvérisation. Il permet ainsi une représentation détaillée des phénomènes étudiés à différentes échelles spatiales, depuis la parcelle traitée jusqu'à des centaines de mètres en aval. Son application nécessite une vigilance par rapport au nombre de particules émises en fonction de la surface traitée et de la distance de modélisation visée : du fait du caractère stochastique de l'approche choisie, ce nombre doit être suffisant pour réduire la variabilité liée au caractère stochastique du modèle tout en maintenant un bon équilibre entre la précision et le temps de simulation.

Une attention particulière a porté sur la description de **la source de gouttelettes lors de la pulvérisation**, en s'attachant à pouvoir considérer des traitements par rang, par face de rang de vigne ou bien de toute la parcelle à la fois, ainsi que des caractéristiques d'émission des gouttelettes comme les hauteurs, angles, directions et vitesse d'éjection des gouttelettes à chaque point d'émission (buse) ainsi que la distribution en diamètre des gouttelettes en fonction du dispositif de pulvérisation utilisé. La littérature avait permis d'identifier ce besoin de précision dans la caractérisation de la source, ce qui a été confirmé par l'analyse de sensibilité réalisée avec *ADDI-SprayDrift*. Afin d'analyser la contribution de différents rangs traités à la dérive selon leur localisation dans la parcelle, le test de sensibilité a été mené sur deux situations : une pulvérisation sur un rang au sein de la vigne et une pulvérisation sur le rang en bordure. Les caractéristiques de la source ont un impact sur la distribution des pesticides dans le champ lors de l'application mais aussi sur les pertes en dehors du champ. Cet effet est certainement d'autant plus marqué pour les cultures pérennes au vu de l'orientation des buses et de la hauteur du couvert. En effet, à travers l'analyse de sensibilité, l'angle d'éjection des gouttelettes a été identifié comme étant le paramètre le plus influent sur leur interception par le couvert et sur les pertes au sol dans et en dehors de la parcelle traitée, et cela dans toutes les conditions, que le rang traité soit au sein de la parcelle ou en bordure de la parcelle, indépendamment des autres paramètres. Les autres paramètres à savoir : la proportion de matière active dans la bouillie, la vitesse d'éjection, la hauteur du couvert/hauteur d'éjection et la vitesse du vent, ont un effet variable selon la position du rang traité (dans ou en bordure du champ) avec un potentiel effet d'interaction entre eux. Ces résultats soulignent l'importance d'acquérir des données sur les paramètres liés à la source qui restent encore insuffisamment renseignés.

En ce qui concerne **la dispersion atmosphérique**, un module de turbulence simplifié, qui considère la turbulence comme étant homogène sur tout le domaine spatial, permet de décrire la dispersion des gouttelettes dans des conditions atmosphériques stables, instables ou neutres. Le profil de vent dans et au-dessus du couvert est décrit en fonction de la hauteur et de la densité du couvert. Toutefois, les structures turbulentes qui se forment au sommet de la végétation sont considérées dans le modèle comme étant similaires à celle d'un couvert horizontalement homogène, ce qui, dans le contexte viticole, n'est vrai que quand la direction du vent est perpendiculaire ou à 45° par rapport aux rangs de vigne. Des améliorations seront donc nécessaires pour considérer le cas où le vent est parallèle aux rangs de vigne, où les pertes en dehors de la parcelle peuvent être assez conséquentes à cause de l'effet de canalisation du vent dans l'inter-rang.

Enfin, en plus de simuler le **dépôt** au sol par sédimentation, le modèle permet également de simuler l'interception des gouttelettes par la végétation par le biais d'une approche lagrangienne qui calcule la probabilité d'interception par sédimentation sur la face horizontale des feuilles et par impaction inertielle sur les faces horizontale et verticale des feuilles. La représentation du mécanisme d'interception augmente la généralité du modèle et son applicabilité à la simulation de différents scénarios d'application de pesticides, l'approche choisie permettant de calculer l'interception en fonction de la géométrie du couvert, de la densité et de la taille de(s) élément(s) sur le(s) quel(s) se fait l'interception, ce qui est original par rapport à la littérature. En effet, dans les modèles mécanistes identifiés dans la synthèse bibliographique, les cultures considérées concernaient essentiellement des couverts bas ou du sol nu (sauf pour ARPS et le modèle de Hong et al. (2018 b,c)). Ainsi, bien que la capacité de filtration des particules par le couvert puisse avoir un effet sur la dérive des gouttelettes en aval du champ ainsi que relevé par certains auteurs, l'interception est souvent soit négligée soit représentée de manière simplifiée ou estimée par le biais d'équations empiriques spécifiques à un couvert particulier.

## **II. Evaluation de la distribution des pesticides lors de l'application et de la dérive de gouttelettes en dehors du champ et de sa contribution à l'exposition des passants:**

En premier lieu, une analyse de sensibilité a permis de tester la cohérence des réponses du modèle en termes de distribution des pesticides entre la fraction déposée dans la parcelle au sol ou sur la végétation, celle sortant de la parcelle et la distance maximale à laquelle 95% de la masse appliquée est dispersée. Ensuite, les performances de prédiction du modèle ont été évaluées par rapport à des données de références et/ou comparées à celles des modèles identifiés dans la synthèse bibliographique.

### *1. Représentation de la dérive sédimentaire :*

La capacité du modèle à représenter la dérive sédimentaire en aval de la parcelle traitée a été évaluée *via* la confrontation des prédictions du modèle aux données expérimentales en vigne de Ganzelmeier et al. (1995), qui présentent l'avantage de compiler un grand nombre de jeux de données par type de



cultures. Cette confrontation a démontré, après un calage de deux paramètres non renseignés dans le jeu de données et identifié lors de l'analyse de sensibilité comme étant les plus influents, à savoir l'angle et la vitesse d'éjection des gouttelettes, un accord très satisfaisant entre la dérive sédimentaire modélisée et mesurée en fonction de la distance par rapport au bord du champ. En effet, les prédictions du modèle se trouvent dans les intervalles d'incertitude des observations pour toutes les distances supérieures à 5 m ; ce n'est qu'au bord du champ (3 et 5 m) que les prédictions sous-estiment les valeurs moyennes observées par un facteur de 2.7 et 1,8 respectivement. Cette sous-estimation a été identifiée comme étant possiblement liée aux incertitudes sur les caractéristiques de la source, notamment la taille et la distribution initiales des particules, les effets de turbulence spécifique en bordure de parcelle et à la non prise en compte par le modèle de l'effet du jet d'air du pulvérisateur sur la turbulence locale. En raison de leur plus grande inertie et de leur plus grande vitesse de sédimentation, les grosses gouttelettes contribuent davantage à la dérive sédimentaire de la pulvérisation au bord du champ que les gouttelettes plus fines. Par conséquent, on s'attend à ce que le dépôt à cette distance dépende largement de la vitesse et des angles d'éjection des gouttelettes, qui ne sont pas connus pour l'ensemble de données de Ganzelmeier, ce qui entraîne une possible imprécision du modèle. Une calibration plus poussée concernant une gamme plus fine de variation des angles d'éjection pourrait améliorer l'efficacité de nos prédictions à 3 et 5 m. Ces imprécisions concernant les prédictions du modèle à 3 et 5 m peuvent également être liées à la simplification de la turbulence, considérée dans notre approche comme homogène sur tout le domaine spatial, ce qui représente une hypothèse assez forte qui ne permet pas de décrire les effets de transition entre parcelle viticole et sol nu et la potentielle augmentation du dépôt en bordure du champ, liée à la zone de turbulence accrue induite par le changement de hauteur de la canopée vers le sol nu. Par ailleurs, l'effet du jet d'air généré par le pulvérisateur sur la turbulence locale n'est pas pris en compte dans le modèle, ce qui peut contribuer à son tour à la réduction de la précision des prédictions du modèle près du pulvérisateur à ces distances proches de la parcelle traitée. La non représentation de l'effet du jet d'air induit en effet une sous-estimation de la vitesse des particules les plus grosses en début de leur trajectoire, particules qui contribuent au dépôt à ces distances. Toutefois, en plus de ce phénomène s'ajoute un processus complémentaire qui n'est décrit dans aucun modèle de dérive et qui correspond à une sédimentation accélérée par un balayage préférentiel lié à l'inertie des particules « *preferential sweeping and clustering of particles* ». Les particules ayant un nombre de Stokes  $\sim 1$  (l'échelle de temps inertielle est alors égale à l'échelle de temps du mélange turbulent), quittent les plus petits tourbillons sur le côté descendant des tourbillons, ce qui augmente la vitesse de dépôt. Des sur ou sous-estimations en proximité du champ traité ont aussi été relevées dans plusieurs des modèles analysés lors de la synthèse bibliographique.

Ainsi, malgré les simplifications de la turbulence, les performances du modèle développé (*ADDI-SprayDrift*) telles qu'évaluées par rapport au jeu de données de Ganzelmeier et al. (1995) apparaissent tout à fait satisfaisantes sur une large gamme de distances allant de 7 à 30 m. Une évaluation plus

approfondie face à des données complémentaires et exhaustives sur d'autres cultures et autres sorties du modèle tels que le profil vertical de concentration et l'exposition des passants par inhalation et dépôt cutané est cependant nécessaire pour renforcer la confiance dans les prédictions du modèle.

2. *Représentation de la distribution des pesticides lors de l'application et de l'exposition des passants par dépôts cutanés ou d'inhalation des gouttelettes de pesticides*

L'évaluation de l'exposition des passants repose sur le calcul de la concentration en gouttelettes en masse ou en nombre dans un volume de dimension défini à hauteur d'inhalation et du dépôt sur les surfaces d'un cube représentant le corps humain, en faisant l'hypothèse que toutes les particules traversant ce volume sont interceptées, ce qui peut conduire à une maximisation de l'exposition par cette voie. En effet, la présence de l'individu modifie la turbulence à petite échelle, créant ainsi une déviation de l'air de part et d'autre de ce dernier, ce qui peut induire une plus faible interception. Il est possible de paramétrer les dimensions  $x$ ,  $y$  et  $z$  des cubes considérés pour l'inhalation et pour le dépôt cutané selon la catégorie d'exposition individuelle étudiée (e.g. adultes, enfants). Ainsi, le modèle *ADDI-SprayDrift* est également applicable pour évaluer l'exposition des enfants ou des résidents en considérant les paramètres appropriés.

Les prédictions du modèle en termes de distribution des pesticides lors de l'application et de l'exposition des passants par dépôt cutané ou inhalation des gouttelettes de pesticides ont été confrontées à des données de références. Des simulations ont été réalisées 1) pour un ensemble de 60 scénarios construits en considérant différents types de pulvérisateurs et buses parmi les plus utilisés en viticulture, de stades de croissance de la vigne (précoce et tardif), de conditions météorologiques (typiques de traitement en matinée ou à midi) ainsi que de concentrations en matière active dans la bouillie et 2) pour un scénario construit afin de s'approcher des conditions expérimentales du jeu de données sélectionné. Les résultats obtenus se sont montrés tout à fait satisfaisants. En effet, de bons accords ont été obtenus avec les deux données de référence de distribution des pesticides lors de l'application et de l'exposition des passants par dépôt cutané ou inhalation pour les mêmes pulvérisateurs, respectivement, avec cependant une légère surestimation de l'exposition des passants par dépôt cutané. Malgré la légère surestimation associée à la maximisation du dépôt sur le corps, les résultats renforcent la confiance dans les prédictions du modèle en termes de répartition initiale du pesticide et d'estimation de l'exposition des passants à la dérive de pulvérisation. Parmi les principaux résultats obtenus, on peut retenir que parmi les pulvérisateurs testés, le pulvérisateur face par face semble conférer un bon équilibre entre l'efficacité de l'application (que l'on associe ici à la capacité d'interception du couvert) et la réduction des pertes dans et hors du champ, par rapport aux pulvérisateurs pneumatiques et à ventilateur axial. Il a également un effet de réduction significative de l'exposition des personnes présentes par inhalation. Cependant, son effet sur la dérive sédimentaire de la pulvérisation et l'exposition par dépôt cutané est moins marqué, ce qui peut être lié à l'interaction avec les autres facteurs et au fait que le dépôt cutané est moins influencé par le gradient vertical de concentration des pesticides que l'inhalation qui s'applique à une hauteur

donnée. Ensuite, même si le pulvérisateur pneumatique semble générer plus de pertes hors champ, il semble plus efficace que le pulvérisateur à ventilateur axial car il permet une meilleure interception et moins de pertes au sol à l'intérieur du champ traité. De plus, la dérive sédimentaire hors champ et l'exposition des passants sont très proches pour les deux pulvérisateurs. Pour l'aéroconvecteur et le face par face, l'utilisation de buses anti-dérive semble réduire les pertes à l'extérieur du champ et, en revanche, augmenter les dépôts à l'intérieur du champ, alors que pour l'interception, leur effet est secondaire par rapport au type de pulvérisateur. La concentration en substance active (s.a.) dans la bouillie semble influencer davantage les pertes hors champ que la distribution du pesticide à l'intérieur du champ traitée, avec une augmentation de la concentration entraînant une diminution de l'exposition par dépôt cutané et par inhalation, mais une augmentation de la dérive sédimentaire à courte distance (jusqu'à 30m), effet que nous pouvons relier au lien entre concentration en substance active et taille des gouttes après évaporation. Quant au stade de croissance de la vigne au moment du traitement, celui-ci influence davantage la distribution des pesticides dans le champ traité entre l'interception de la canopée et le dépôt au sol que les pertes hors champ, excepté l'exposition des personnes présentes par inhalation, qui dépend du gradient vertical de concentration. Or ce dernier est impacté par l'augmentation de la dispersion des petites particules causée par l'accélération du profil du vent induite par la présence d'une végétation plus dense et plus haute au stade de croissance développé.

Enfin, les conditions météorologiques testées ici pour représenter des applications le matin ou en cours de journée semblent avoir peu d'effet sur la distribution des pesticides dans la parcelle traitée et un effet plus significatif sur la dérive sédimentaire et son évolution en fonction de la distance.

Ainsi, l'ensemble de ces résultats montre que *ADDI-SprayDrift* est un outil tout à fait pertinent pour analyser les effets de plusieurs grandeurs clefs gouvernant la distribution de la pulvérisation au sein de la parcelle traitée, et la dispersion atmosphérique en aval de celle-ci, source d'exposition des populations et d'écosystèmes non cibles. Il pourrait être pertinent d'en dégager une hiérarchisation. Cependant, même si une adaptation appropriée du dispositif de pulvérisation aux caractéristiques du couvert végétal traité (angle et hauteur d'éjection) est évidente pour augmenter l'interception de l'application et réduire les pertes en champ et hors champ, il est plus difficile d'établir une hiérarchie de la contribution relative de chacun des facteurs testés à la dérive sédimentaire et à l'exposition des passants, en lien avec la variabilité de l'effet des facteurs testés selon la sortie considérée, qui à son tour est liée aux interactions existant entre eux. Des tests supplémentaires sont donc nécessaires pour établir une telle hiérarchie des effets.

Une validation supplémentaire du modèle pour l'évaluation de l'exposition et pour le profil vertical du champ de concentrations en pesticides à différentes distances est nécessaire en utilisant des données expérimentales plus exhaustives. Toutefois, de tels jeux de données restent rares ainsi que relevé dans le dernier rapport de l'Autorité européenne de sécurité des aliments qui souligne également

l'impossibilité de comparer les quelques données existantes en raison de la différence des conditions d'application en termes de conditions météorologiques, de types de pulvérisateurs et également de types de cultures.

En perspective, trois points peuvent être abordés pour améliorer et/ou compléter l'approche entreprise.

- 1) Le premier consiste en une amélioration de la description de certains processus pour permettre une meilleure précision et/ou une plus grande généralité du modèle si nécessaire.

Cette amélioration concerne principalement le module de turbulence atmosphérique pour lequel deux hypothèses ont été faites. La première hypothèse concerne l'homogénéité de la turbulence sur tout le domaine de calcul. Cette hypothèse ne permet pas de considérer les trois phénomènes suivants: 1) la zone de turbulence accrue liée à transition entre le couvert traité et un sol nu ou une végétation basse en aval, générant une incertitude sur la prédiction de la dérive en bordure de la parcelle ( $\leq 5$  m), 2) la présence d'un obstacle comme une barrière physique et son effet sur la turbulence et in fine sur la dérive des gouttelettes et l'exposition des riverains et passants et 3) la dispersion et l'effet de canalisation du vent au niveau des inter-rangs quand le vent est parallèle au rangs de vigne (cas des couverts en rangs seulement), situation pour laquelle les pertes en dehors du champ peuvent être importantes. La deuxième hypothèse concerne le fait de négliger l'ensemble des effets du jet d'air du pulvérisateur sur la dispersion des particules. En effet, bien que l'effet de jet d'air sur la vitesse initiale des particules à la sortie des buses soit pris en compte, l'absence de prise en compte de l'effet du jet d'air sur la turbulence à proximité des buses induit une sous-estimation de la vitesse des particules à proximité du pulvérisateur qui peut contribuer à une incertitude sur la prédiction de la dérive en bordure de la parcelle. Cette hypothèse peut également induire une sous-estimation de la dispersion latérale des gouttelettes d'un diamètre inférieur à 100  $\mu\text{m}$  qui à son tour peut contribuer à la réduction de la précision des prédictions du modèle à des distances supérieures à  $\sim 50\text{m}$ . Selon les objectifs de la modélisation envisagée, de potentielles solutions peuvent être proposées.

- Dans l'objectif de simuler l'effet de la variation du vent par rapport aux rangs de vigne, de l'effet du jet d'air et la considération de l'effet de certaines structures écologiques (e.g. les haies) sur la réduction de la dérive en aval de la parcelle traitée, un couplage du modèle avec des modèles de turbulence en conditions hétérogènes (k-epsilon) est nécessaire.
- Dans l'objectif de l'amélioration des prédictions du modèle concernant la dérive sédimentaire en bordure de la parcelle ( $\leq 5$  m), la mobilisation de modèles complexes de turbulence ne semble pas la meilleure solution en termes d'efficacité d'amélioration et de temps de calcul, étant donné que la sous-estimation du dépôt à ces distances n'est pas uniquement liée à cette hypothèse d'hétérogénéité de surface, mais aussi à la caractérisation de la source et au phénomène de balayage et d'agrégation préférentiel « *preferential sweeping and clustering* » des particules, cet effet ayant été identifié pour expliquer la sous-estimation aussi observées avec les modèles k-

epsilon. Il serait plus bénéfique d'intégrer le phénomène en question au lieu de complexifier la turbulence et augmenter le temps de calcul. Cet effet pourrait être intégré en modifiant la paramétrisation de la vitesse de sédimentation, à l'aide de facteurs correctifs dépendant du nombre de Stokes.

Ensuite, une analyse de sensibilité plus détaillée par rapport au nombre de particules minimal nécessaire en fonction de la taille de la parcelle traitée et de la distance de modélisation en aval de la parcelle traitée est indispensable avant l'utilisation du modèle, dans des cas d'application très différents de ceux présentés, pour permettre un bon équilibre entre la diminution de la variabilité des sorties et l'optimisation du temps de calcul. A titre d'exemple, lors des simulations établies, 20,000 particules étaient suffisantes pour simuler une source ponctuelle et la dérive à 30 m de la parcelle, alors que pour une parcelle de 540 m<sup>2</sup> et pour des distances allant jusqu'à 160m de la parcelle, un minimum de 1,000,000 de particules est nécessaire.

- 2) La deuxième amélioration consiste en la réalisation de validations supplémentaires en termes de dérive sédimentaire et aérienne et d'exposition des riverains et des passants et cela face à des données plus exhaustives ou bien des données issues de la même expérimentation et qui regrouperaient les trois grandeurs. Cela permettrait une meilleure connaissance des performances du modèle ou d'apports supplémentaires pouvant améliorer l'évaluation des pertes pour dans des cas exceptionnels d'utilisation de pulvérisateur plus complexe tel que le pulvérisateur à panneaux récupérateurs.

En effet, des tests supplémentaires avec plus de scénarios détaillés sont nécessaires pour améliorer notre connaissance de l'effet des types de pulvérisateurs, de buses, de proportion en matière active dans la bouillie, du stade de développement de la végétation et des conditions météorologiques sur la distribution des pesticides lors de l'application, la dérive sédimentaire et aérienne et l'exposition des riverains et passants à la dérive des gouttelettes en fonction de la distance, mais aussi pour pouvoir hiérarchiser leur effets.

Une description du terme source dans le cas de système de pulvérisation complexe tel qu'un pulvérisateur à panneaux récupérateurs est envisageable. Il est dans ce cas nécessaire de simuler la turbulence entre les deux panneaux, induite par la forte assistance d'air, mais aussi l'effet de l'avancement du tracteur qui dans le cas de ce pulvérisateur, et vu le faible diamètre des gouttelettes générées, risque d'avoir un effet significatif sur la dispersion des très fines gouttelettes.

- 3) Pour finir, globalement pour compléter cette approche de modélisation et pour un passage à une estimation des pertes à une échelle spatiale et temporelle plus large, *ADDI-SprayDrift* pourrait être couplé à 1) un modèle de volatilisation en post application des pesticides depuis la parcelle traitée en post application pour acquérir une vision plus globale de la contamination et de l'exposition liées à la voie atmosphérique et d'en évaluer les contributions relatives ou 2) à un modèle de devenir des pesticides dans l'environnement étant donné qu'*ADDI-SprayDrift* fournit la distribution des pesticides lors de l'application dans et en dehors de la parcelle traitée.

Le premier couplage permettra une évaluation des pertes par voie atmosphérique, de la contamination des zones non ciblées par les retombées gazeuses et particulaires et l'exposition des différents êtres vivants y compris les humains par ces derniers. Des propositions d'optimisation des techniques de pulvérisation pourraient ainsi être identifiées.

Le deuxième couplage permettra une évaluation plus globale à l'échelle du paysage ou même du territoire de l'effet des conditions initiales de pulvérisation sur le devenir des pesticides dans l'environnement. Ainsi l'efficacité de certaines mesures de réduction de la contamination sera évaluée et certains leviers de réduction du transfert et de la contamination pourront être identifiés. L'intégration d'*ADDI-SprayDrift* dans un tel outil fait partie des développements en cours dans le cadre du projet Ecophyto RIPP-Viti, où l'outil MIPP (Modélisation Intégrée du devenir des Pesticides dans le Paysage) est déployé pour prédire le devenir des pesticides dans le paysage et identifier les leviers d'action à cette échelle.



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# *Annexes*

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## Annexe A

*Modélisation de la dérive de gouttelettes et de la distribution des pesticides lors de l'application*

## ADDI-SprayDrift: A COMPREHENSIVE MODEL OF PESTICIDE SPRAY DRIFT WITH AN ASSESSMENT IN VINEYARDS

### 1. Supplementary model equations

#### 1.1. Evaporation parameters

The evaporation model follows the differential equation for the droplet diameter  $D$ :

$$\frac{\partial D}{\partial t} = - \frac{D}{2 \tau_e \left(1 - \frac{t}{\tau_e}\right)} \quad (\text{A1})$$

where  $D$  is the droplet diameter (m),  $t$  is the time (s) and  $\tau_e$  is the droplet evaporation time scale (s).

In the evaporation model and equation 3 of the manuscript, the evaporation rate  $\lambda$  ( $\mu\text{m}^2 \text{s}^{-1}$ ) is:

$$\lambda = \frac{8 \kappa}{\rho L} \quad (\text{A2})$$

where  $\kappa$  is the water thermal conductivity equal to  $0.024 \text{ W m}^{-1} \text{ K}^{-1}$  at  $20^\circ\text{C}$ ,  $\rho$  the water density equal to  $1 \text{ g cm}^{-3}$  and  $L$  the water latent heat of evaporation equal to  $2257 \text{ kJ kg}^{-1}$  at  $20^\circ\text{C}$ .

The wet-bulb temperature depression  $\Delta\theta$  in  $^\circ\text{C}$  results from the difference between  $\theta_d$  the dry-bulb temperature ( $^\circ\text{C}$ ) and  $\theta_w$  the wet-bulb temperature ( $^\circ\text{C}$ ), computed as:

$$\Delta\theta = \theta_d - \theta_w \quad (\text{A3})$$

Wet-bulb temperature ( $^\circ\text{C}$ ) can be calculated using the empirical equation of (Stull, 2011):

$$\begin{aligned} \theta_w = T \text{ atan}[0.151977 (RH\% + 8.313659)^{1/2}] \\ + \text{ atan}(T + RH\%) \text{ atan}(RH\% - 1.676331) \\ + 0.00391838 (RH\%)^{3/2} \text{ atan}(0.023101 RH\%) - 4.686035 \end{aligned} \quad (\text{A4})$$

Where  $T$  is the air temperature in ( $^\circ\text{C}$ ) and  $RH\%$  is the air relative humidity in (%).

The droplet relative velocity  $U$  ( $\text{m s}^{-1}$ ) is given by the following equation:

$$U = \sqrt{(u_p)^2 + (v_p)^2 + (w_p - v_s)^2} \quad (\text{A5})$$

The kinematic viscosity of the air ( $\text{m}^2 \text{s}^{-1}$ ) calculated as function air temperature ( $T$ ) in ( $^{\circ}\text{C}$ ) (Haynes, 2017):

$$\nu(T) = (0.0009331 \times T + 0.1316) \times 0.0001 \quad (\text{A6})$$

## 1.2. Dispersion parameters

The Stokes time is equal to:

$$T_s = v_s/g \quad (\text{A7})$$

where  $g$  is the acceleration of the earth's gravity ( $9.81 \text{ m s}^{-2}$ ).

The settling velocity can be calculated according to the following equation:

$$v_s^2 = \frac{4}{3} \frac{g D \rho_p}{C_D \rho} \quad (\text{A8})$$

$$C_D = \frac{24}{Re_p} \left(1 + 0.158 Re_p^{2/3}\right) \quad (\text{A9})$$

where  $\rho$  is the air density in  $\text{kg m}^{-3}$ ,  $D$  is the droplet diameter (m),  $\rho_p$  is the particle density set equal to  $1000 \text{ kg m}^{-3}$  and  $C_D$  is a dimensionless number representing the drag coefficient. It can be calculated by Eq. (14) given by (Loubet et al., 2007) and taken from (Aylor, 2002), this equation can be used to predict the drag coefficient for a wide range of particle Reynold's numbers (from 0.001 to 200). Note that when  $Re_p$  tends to 0, Eq. (A9) tends to the Stokes law equation. Additionally, for this equation, the Reynolds number  $Re_p$  ( $Re = \frac{D_0 U}{\nu}$ ) is computed using  $v_s$  as the particle velocity and  $D$  as the characteristic length scale.

## 1.3. Wind speed and turbulence above the canopy

The mean wind speed profile at a height ( $z$ ) is represented by the following logarithmic equation (Stull 1988; Loubet 2000; Loubet et al., 2006; Kasbadji Merzouk et al., 2007):

$$\bar{U}(z) = \frac{u_*}{\kappa} \ln\left(\frac{z-d}{z_0}\right) - \Psi_M\left(\frac{z-d}{L}\right) + \Psi_M\left(\frac{z_0}{L}\right) \quad (\text{A10})$$

Where  $\kappa$  is the Von Karman constant ( $\kappa = 0.41$ ),  $z$  (m) is the height above the ground,  $z_0$  (m) is the roughness length (set equal to 1/10 of the canopy height (Hashem & Parkin, 1991)),  $L$  is the Monin–Obukhov length (m) (see Eq. A17),  $u_*$  is the friction velocity ( $\text{m s}^{-1}$ ) (Eq. A18), and  $\Psi_M$  is the stability correction function (Eq. A11):

$$\Psi_M(\xi) = \begin{cases} 2 \ln\left(\frac{1+x}{2}\right) + \ln\left(\frac{1+x^2}{2}\right) - 2 \text{Arc tan}(x) + \frac{\pi}{2} & \text{for } -2 \leq \xi \leq 0 \\ -5(\xi) & \text{for } 0 \leq \xi \leq 1 \end{cases} \quad (\text{A11})$$

where  $\xi = \frac{z-d}{L}$  and  $x = (1 - 16 \xi)^{1/4}$ . Notably, this formulation is valid only for moderately stable, neutral and moderately unstable conditions ( $-2 \leq \xi \leq 1$ ). The Lagrangian time scale is described as follows above the canopy:

$$\tau = c \left[ \frac{z-d}{u_*} \right] \quad (\text{A12})$$

where  $z$  is the height above the ground (m),  $u_*$  is the friction velocity ( $\text{m s}^{-1}$ ),  $d$  is the displacement height (m) and  $c$  is a constant ( $d = 0.63 \times \text{crop height}$  and  $c = 0.4$  (Hashem and Parkin, 1991; Loubet et al., 2006)). Following Loubet et al. (2006), we further take the following:

$$\sigma_u = \sigma_v = u_* (12 - 0.5 z/L)^{1/3} \quad (\text{A13})$$

with  $\sigma_u = \sigma_v \approx 3 u_*$  (for typical values of  $z/L$  during daytime) and

$$\sigma_w = 1.3 u_* (1 - 3 (z-d)/L)^{1/3} \quad (\text{A14})$$

where  $L$  is the Monin–Obukhov length (m).

#### 1.4. Wind speed and turbulence in the canopy

Through the drag effect, the vegetation adsorbs part of the movement, which induces a deceleration of the airflow in the canopy. The resulting velocity profile within a homogenous canopy has an exponential form (Eq. A15) (Legg & Raupach, 1982), and it depends on the height ( $z$ ) above the ground.

$$\bar{U}(z) = \bar{U}(h) \times \exp\left(\gamma \left(\frac{z}{h} - 1\right)\right) \quad (\text{A15})$$

where  $h$  is the canopy height (m) and  $\gamma$  is the extinction coefficient within the crop with  $\gamma = 3.0$  (Legg & Raupach, 1982). The Lagrangian time scale in the canopy is computed as:

$$\tau = c \left[ \frac{h-d}{u_*} \right] \quad (\text{A16})$$

In the canopy, the stability parameter is assumed to be constant and equal to  $\xi = \frac{h-d}{L}$ . Some corrections are necessary for the vertical standard deviation of wind speed ( $\sigma_w$ ) and the Lagrangian time scale using the Monin–Obukhov length correction function (Loubet, 2000). The correction equations are presented in Eqs. (A20) to (A26).

Equation of Monin-Obukov length (Stull, 1988):

$$L = \frac{\rho C_p u_*^3 T_a}{\kappa g H} \quad (\text{A17})$$

Where  $\rho$  is the air density in ( $\text{kg m}^{-3}$ ),  $C_p$  the heat capacity of air at constant pressure ( $\text{J kg}^{-1} \text{K}^{-1}$ ),  $u_*$  the friction velocity ( $\text{m s}^{-1}$ ),  $T_a$  air temperature (K),  $\kappa$  the von Karman constant (a dimensionless number equal to 0.4),  $g$  the acceleration of the earth's gravity ( $9.81 \text{ m s}^{-2}$ ) and  $H$  the sensible heat flux ground-air ( $\text{W m}^{-2}$ ).

The variation of the atmospheric turbulence at the boundary layer is described by the friction velocity which depends on the wind speed, the area covered, its height and its length (Thompson & Ley, 1983).

$$u_* = \sqrt{\tau_s / \rho} = \sqrt{v \times \left. \frac{\Delta \bar{u}}{\Delta y} \right|_{y=0}} \quad (\text{A18})$$

Where  $\tau_s$  is shear stress,  $v$  is kinematic viscosity of air,  $\Delta \bar{u}$  is fluid velocity at a height  $y$  and  $\Delta y$  is the coordinate space coordinate representing the position of the fluid.

$$\tau_s = \rho v \left. \frac{\Delta \bar{u}}{\Delta y} \right|_{y=0} \quad (\text{A19})$$

Monin-Obukhov length correction function for the vertical standard deviation of wind speed ( $\sigma_w$ ) and the Lagrangian time scale (Loubet, 2000).

$$\varphi_w = \begin{cases} 1.25 (1 + 3 \xi)^{1/3} & -2 \leq \xi \leq 0 \\ 1.25 (1 + 0.2 \xi) & 0 \leq \xi \leq 1 \end{cases} \quad (\text{A20})$$

$$\varphi_h = \begin{cases} (1 + 16 \xi)^{-1/2} & -2 \leq \xi \leq 0 \\ 1 + 5 \xi & 0 \leq \xi \leq 1 \end{cases} \quad (\text{A21})$$

Above the canopy  $\xi = \frac{z-d}{L}$  and in the canopy  $\xi = \frac{h-d}{L}$ .

Before correction,  $\sigma_w$  and  $\tau$  are expressed as following:

Above the canopy:

$$\tau = \frac{0.3 \times (z - d) \times h}{u_* \times (h - d)} \quad (\text{A22})$$

In the canopy:

$$\sigma_w = u_* \times \exp \left[ 0.6 \times LAI \times \left( \frac{z}{h} - 1 \right) \right] \quad (\text{A23})$$

$$\tau = \frac{0.3 \times h}{u_*} \quad (\text{A24})$$

By integrating, the correction functions  $\varphi_w$  and  $\varphi_h$  the Lagrangian time scale and the vertical standard deviation of wind speed are calculated as following:

$$\sigma_w = \sigma_w \times \frac{\varphi_w}{1.25} \quad (\text{A25})$$

$$\tau = \tau \times \frac{(1.25)^2}{(\varphi_w)^2 \times \varphi_h} \quad (\text{A26})$$

In thermal neutrality conditions, these correction functions tend to 1.

The turbulent diffusivities are computed as follows:

$$\begin{cases} K_x = k_p \times \sigma_u^2 \times \tau \\ K_y = k_p \times \sigma_v^2 \times \tau \\ K_z = k_p \times \sigma_w^2 \times \tau \end{cases} \quad (\text{A27})$$

where  $\tau$  is the turbulent Lagrangian time scale and  $\sigma_u, \sigma_v, \sigma_w$  are the horizontal, crosswind and vertical standard deviations of the wind speed ( $\text{m s}^{-1}$ ), respectively (Loubet et al., 2006; Thompson & Ley, 1983). These are detailed in the next section.

The particle diffusivity is reduced compared to the gas diffusivity by a factor  $k_p$  taken from Bouvet and Wilson (2006):

$$k_p = \frac{1}{\sqrt{1 + (1.6 * v_s/u_*)^2}} \quad (\text{A28})$$

### 1.5. Deposition parameters

The impaction efficiency  $Ei$  is given by (Aylor, 1982; Aylor and Flesch, 2001) as:

$$Ei = \frac{0.86}{1 + 0.442 St^{-1.967}} \quad (\text{A29})$$

where  $St$  is the Stokes number ( $St = v_s \sqrt{\langle u + u_p \rangle^2 + \langle v + v_p \rangle^2 + \langle w + w_p \rangle^2} (gl)^{-1}$ ), which depends on settling velocity ( $v_s$ ), acceleration of gravity ( $g$ ), relative wind speed influencing the droplets  $\sqrt{\langle u + u_p \rangle^2 + \langle v + v_p \rangle^2 + \langle w + w_p \rangle^2}$  and vegetation element characteristic size ( $l$ ). In our model, interception is considered to occur only on crop leaves, so the width (m) of leaves is taken as the characteristic size of the canopy element.

### 1.6. Model evaluation

The efficiency factor (EF), the root mean square error (RMSE), and the relative root mean square error (RRMSE) were estimated as follows:

$$EF = 1 - \frac{\sum_{i=1}^n (y_{sim,i} - y_{exp,i})^2}{\sum_{i=1}^n (y_{exp,i} - \bar{y}_{exp})^2} \quad (\text{A30})$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^n (y_{sim,i} - y_{exp,i})^2}{n}} \quad (\text{A31})$$

$$RRMSE = \sqrt{\sum_{i=1}^n \left( \frac{y_{sim,i} - y_{exp,i}}{\sigma_{exp}} \right)^2} \quad (\text{A32})$$

where  $y_{sim,i}$  represents the simulated values,  $y_{exp,i}$  the experimental values,  $\bar{y}_{exp}$  the mean of the experimental values and  $\sigma_{exp}$  the standard deviation of the experimental values, and  $i=1:N$  is the index over the number of points.

## 2. Evaluation of evaporation model

Figure S.A.1.a. Simulated percentages of diameter loss due to evaporation of drops from 10 to 100 $\mu\text{m}$  by ADDI-Spraydrift. Curves colours refer to the temperature ( $^{\circ}\text{C}$ ) and the relative humidity (%) used for the simulation whereas the symbols refer to the droplets initial diameter.

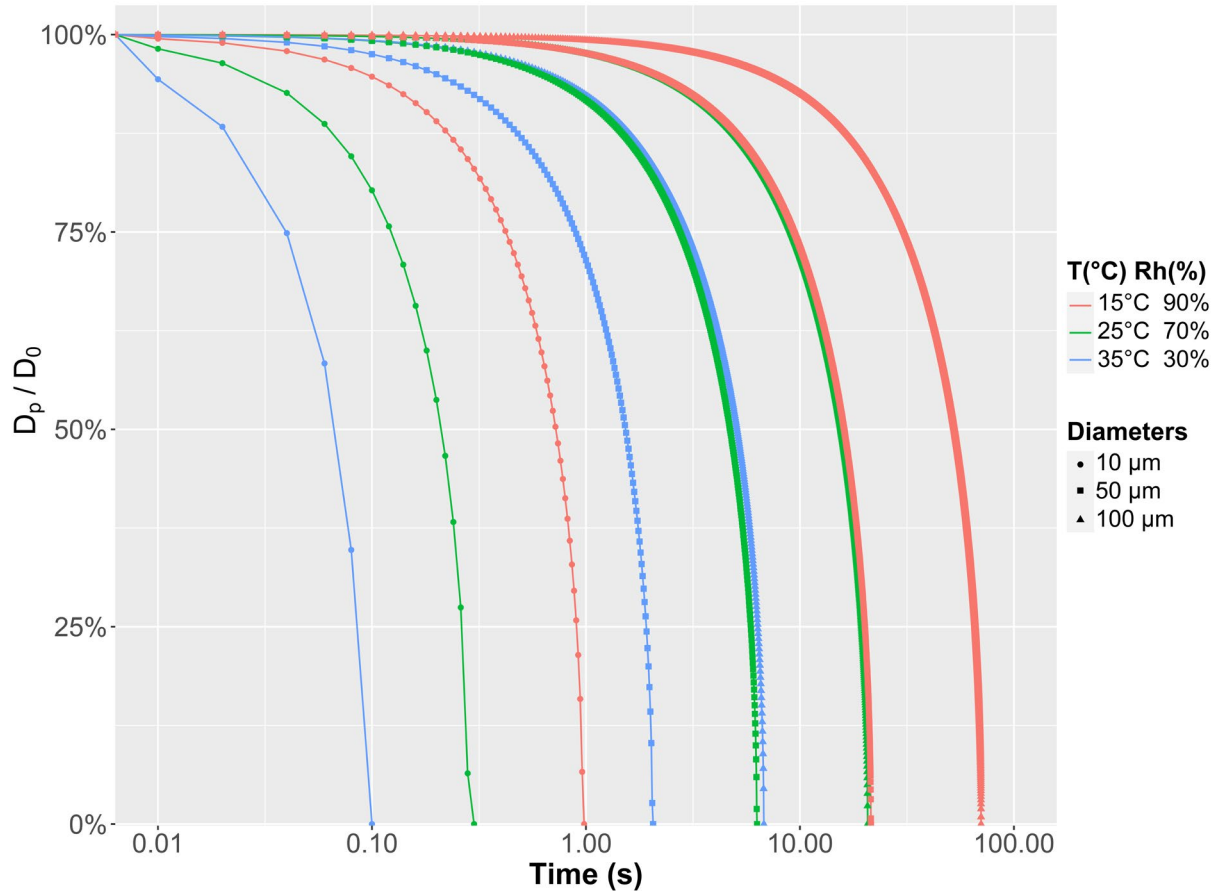
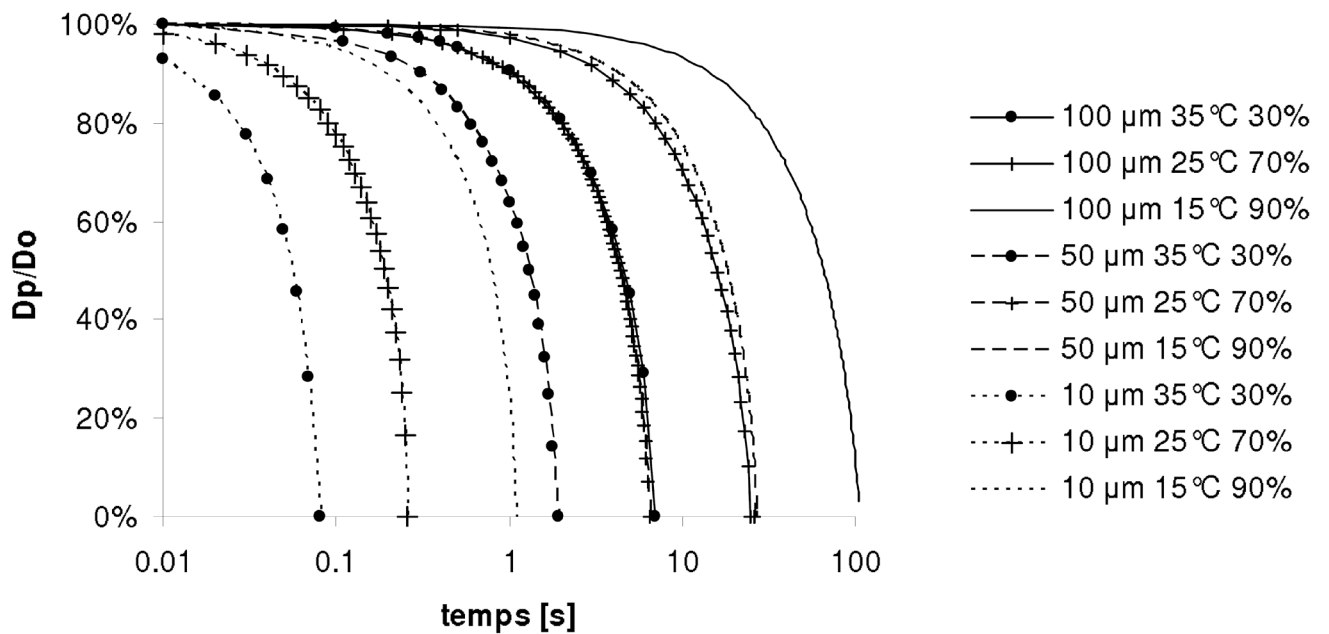


Figure S.A. 1.b. Simulated percentages of diameter loss due to evaporation of drops from 10 to 100 $\mu\text{m}$  by Holterman's model following initial drop size, temperature ( $^{\circ}\text{C}$ ) and relative humidity (%).



### 3. Model implementation

Table S.A.1. Model inputs and outputs

<b>Model inputs</b>	<b>Simulation parameters</b>	<ul style="list-style-type: none"> <li>▪ Simulation duration in s</li> <li>▪ Time step (t) in s</li> <li>▪ Number of released particles</li> </ul>
	<b>Meteorological and turbulence parameters</b>	<ul style="list-style-type: none"> <li>▪ Wind speed (u) in m/s</li> <li>▪ Friction velocity (u*) in m/s</li> <li>▪ Monin-Obukow length (LMO) in m</li> <li>▪ Averaged height of the domain at which turbulence parameters are calculated / measurement height (zmean) in m</li> <li>▪ Air temperature (Ta) in °C</li> <li>▪ Air relative humidity (Rha) in %</li> </ul>
	<b>Application parameters (Source and canopy characteristics)</b>	<ul style="list-style-type: none"> <li>▪ Particles mean diameter (mean_dp) in <math>\mu\text{m}</math></li> <li>▪ Particle diameter standard deviation (SD_dp) in <math>\mu\text{m}</math></li> <li>▪ Droplet density (density_part) in <math>\text{kg m}^{-3}</math></li> <li>▪ Active mater proportion within tank mixture (active_m_proportion) in %</li> <li>▪ Particles resuspension rate (Res) dimensionless</li> <li>▪ Source height (zsource) in m</li> <li>▪ Source distance regarding the canopy/treated surface (source_distance) in m</li> <li>▪ Ejection angle (AngleInjection) in °</li> <li>▪ Ejection velocity (VelocityInjection) in <math>\text{m s}^{-1}</math></li> <li>▪ Canopy height (hc) in m</li> <li>▪ Leaf Area index (LAI) dimensionless</li> <li>▪ Leaves typical size – width- (l) in m</li> <li>▪ Leaves inclination angle (leaf_angle) in °</li> </ul>
	<b>Specific canopy characteristics parameters for vine application</b>	<ul style="list-style-type: none"> <li>▪ Vine rows width (row) in m</li> <li>▪ Rows spacing width (rows_spacing) in m</li> <li>▪ First row starting position (row_1_start) in m</li> <li>▪ Number of rows</li> <li>▪ Size of vine/tree trunk (leaves_start) in m</li> <li>▪ Spraying direction (spraying_direction) : “same”/”opposite” to win direction</li> <li>▪ Treated row (treated_row) : number of the treated row from field start</li> </ul>
<b>Model outputs*</b>	<ul style="list-style-type: none"> <li>▪ Quantity of pesticides deposited on soil in the field</li> <li>▪ Quantity of pesticides deposited on soil out of the field</li> <li>▪ Quantity of pesticides intercepted by the canopy</li> <li>▪ Pesticide concentration in the air at various heights, distances and time</li> </ul>	

(\*) Model output consist on one file containing all droplets trajectories at each time step. The presented output variable can be deduced from this single file.



## 4. Case study and model evaluation

Figure S.A.2. Model setup for the Ganzelmeier et al. (1995) case study: spraying material, treated rows and spatial dimension representation. The red arrows represent source positions and spraying directions in/opposite to wind direction. Numbers represented in the rectangles represents rows positions regarding the field edge.

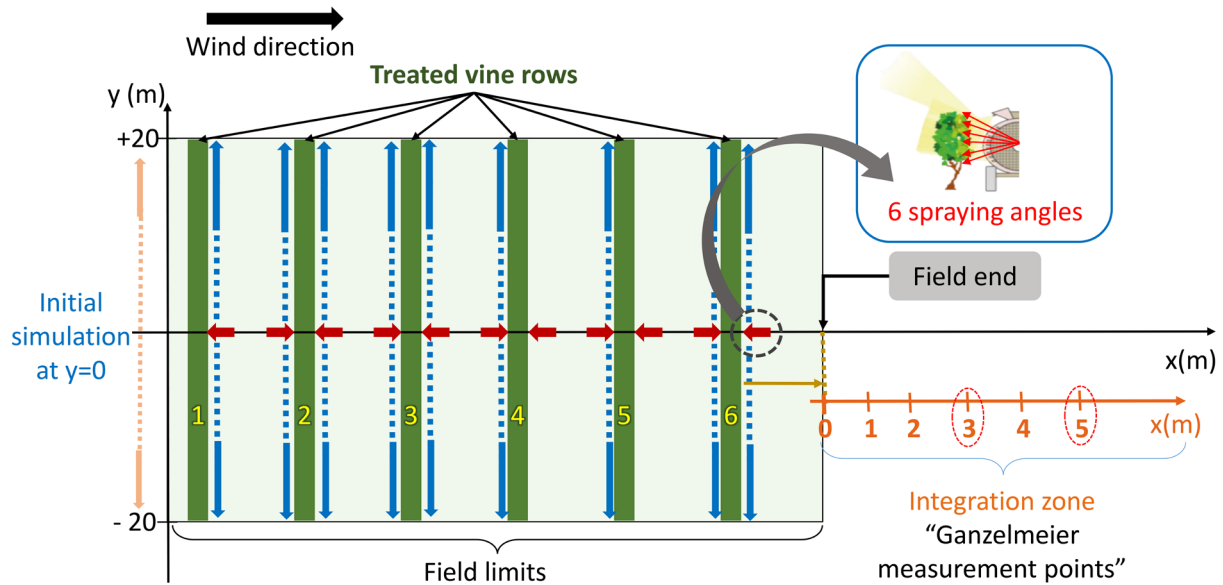


Figure S.A.3. Droplet size distribution (A) and the corresponding settling velocity (B) and Stokes time (C).

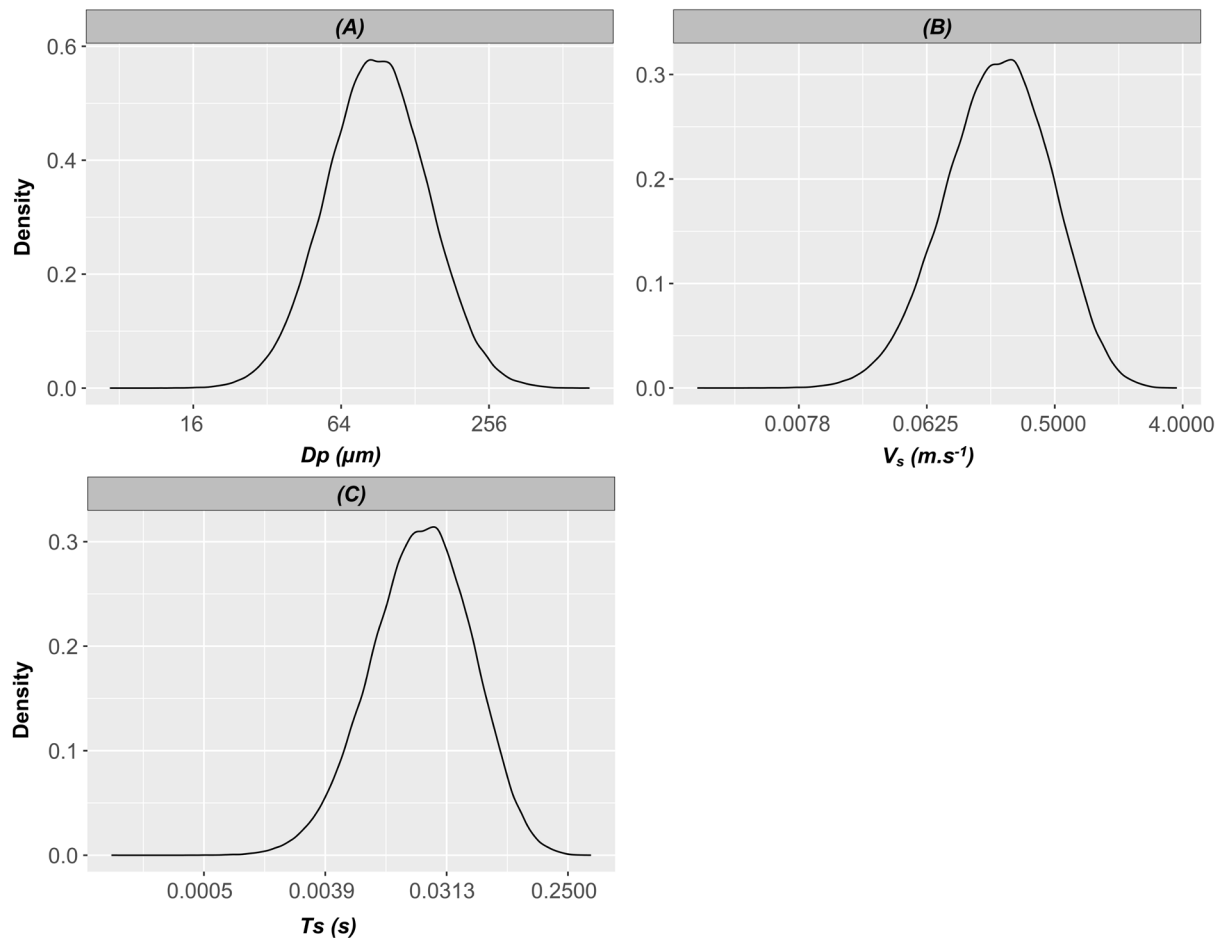


Figure S.A.4. 2D representation of the changes in particles mean diameter ( $\mu\text{m}$ ) per  $0.4\text{m}^3$  in the simulated pesticide plume for the first 20 seconds following spraying of 2nd and 6th vine rows (ROW- 2 and ROW-6).

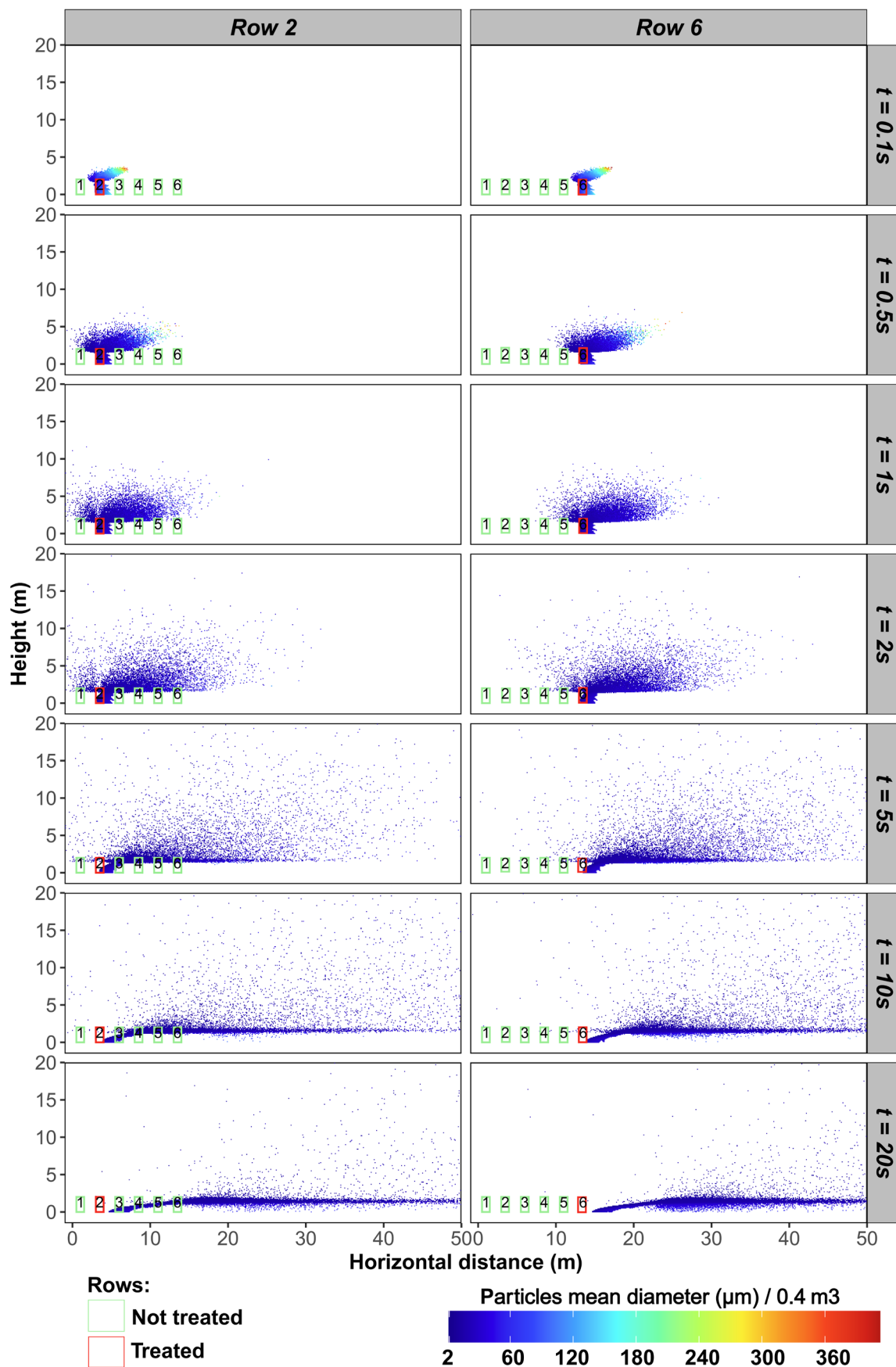


Figure S.A.5. Variation of four model outputs following the values of five main influential model parameters when treating the second and sixth rows of vines based on four output variables against

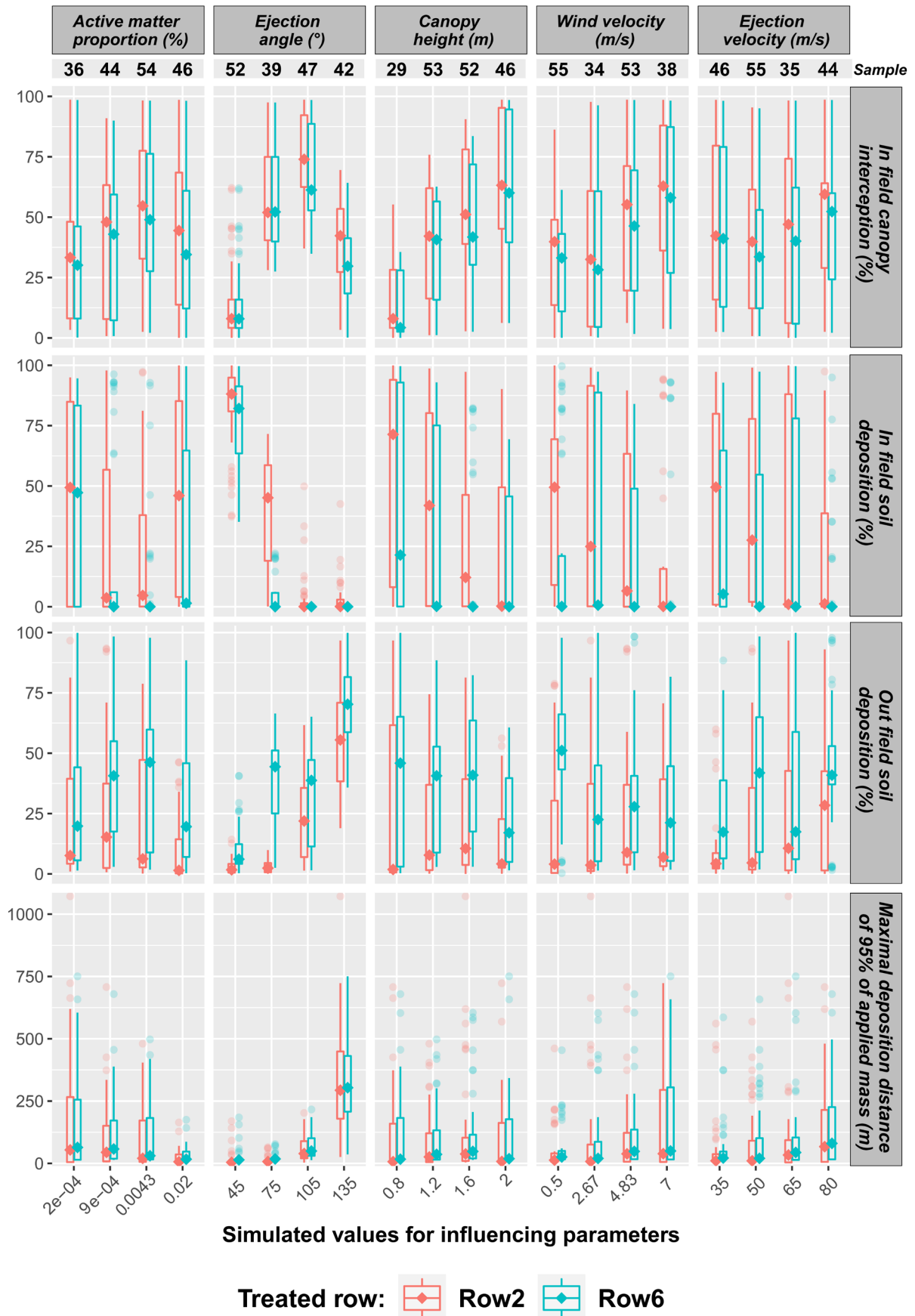


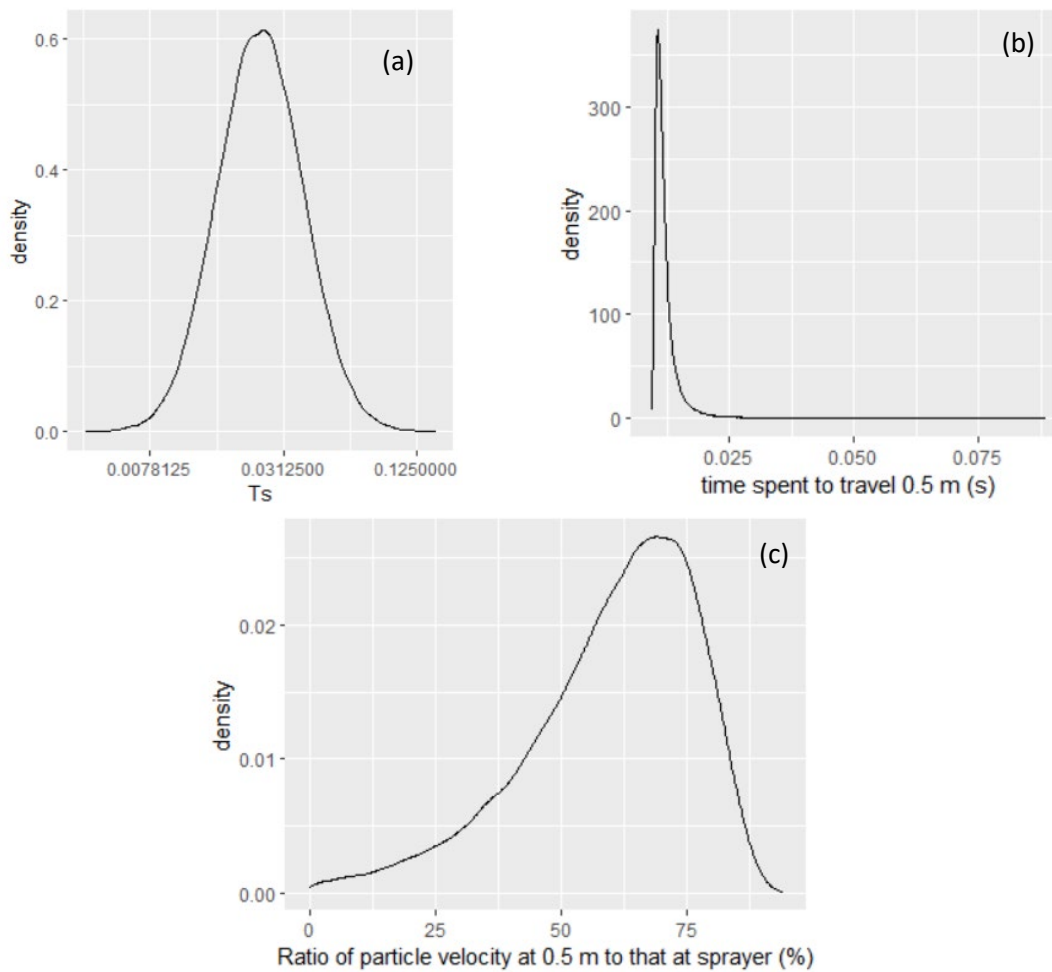
Table S.A.2. Comparison between Ganzelmeier et al. (1995) experimental measurement and model results of pesticides spray drift deposition materiel in % relative to application rate along the horizontal distance from the source

x	Ganzelmeier Overall mean deposition	SD mean deposition	Model
3	4,99	1,42	1,86
5	2,68	0,92	1,50
7,5	1,48	0,44	1,16
10	0,91	0,26	0,88
15	0,43	0,14	0,42
20	0,24	0,09	0,23
30	0,11	0,04	0,11

## 5. Analysis of the potential impacts of neglecting effect of the air jet on the local turbulence.

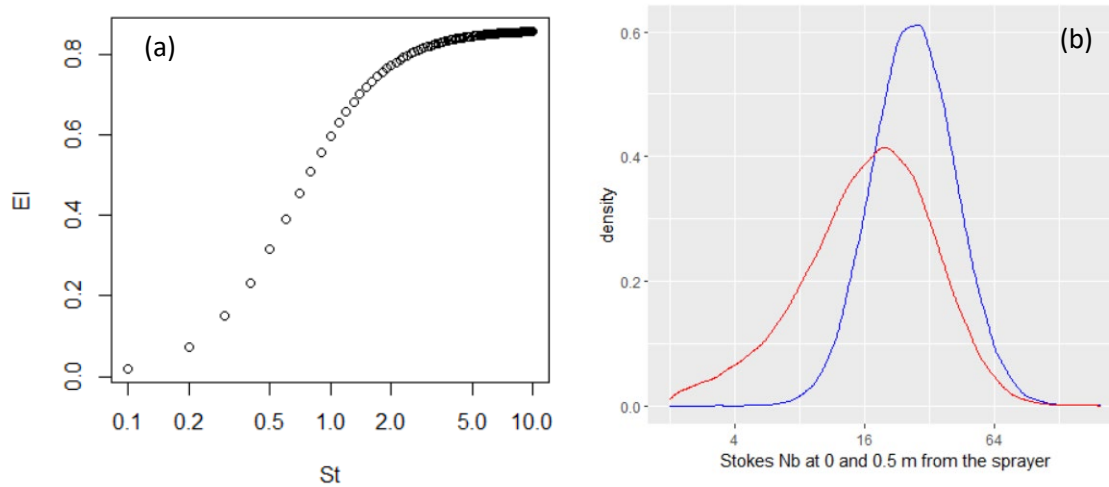
The first effect of the air jet is to produce a flow that has the same speed and direction as the droplets at the release point. This will hence reduce the air drag on droplets (virtually zero drag) compared to our model, and therefore will increase the particle velocity at further distances from the sprayer. Indeed, for air assisted sprayers, measurements from Gu, Zhu, & Ding (2012) and modelling from Hong, Zhao, & Zhu, (2018a) indicate that air velocity at 0.5 m from the sprayer is ~ 30% of the initial air velocity. The droplets travel time over that distance is smaller than the Stokes time for our droplet diameter distribution (Fig. S.A.6.a and b). This means that the droplets will have a velocity close to their initial velocity at that distance. If we now compare with the results obtained with our modelling approach which does not explicitly describe the effect of air jet on droplets (inducing thus a large drag force), the droplet velocity at 0.5 m is simulated to be around 60% of the initial velocity in average (see Fig. S.A.6.c below). We therefore can say that our model would underestimate the particle velocity by around 40% on average at 0.5 m from the sprayer due to air jet misrepresentation. In principle, this may subsequently bias droplet interception by canopy and droplet evaporation.

Figure S.A.6. (a) Inertial time scale (or Stokes time) distribution of the droplets released. (b) Distribution of particle velocity at 0.5 m downwind from the sprayer normalised by initial velocity. (c) Distribution of time spent to travel 0.5 m for log-normally distributed droplets with mean 100  $\mu\text{m}$  and sd 50  $\mu\text{m}$  and initial velocity 55 m s<sup>-1</sup>



Regarding droplet interception, the effect of a change of 40% in the particle velocity will not reduce significantly the impaction efficiency ( $E_i$ ) because the Stokes number remains larger than 4 (Fig.S.A.7.b), a value for which  $E_i$  remains at its maximum in Aylor (1982) and Aylor and Flesch (2001) law (see Fig. S.A.7.(b) and (a) below, respectively). We can hence stipulate that not accounting for the air blast effect on the drag would only affect particle deposition to canopy in the smallest range of sprayer droplet sizes, meaning we will produce a rather conservative estimate of particle settling drift.

Figure S.A.7. (a) Impaction efficiency as a function of the Stokes number from (Aylor, 1982; Aylor & Flesch, 2001). (b) Density distribution of Stokes numbers among droplets at the sprayer outlet (blue) and 0.5 m downwind (red) with no air jet.



Regarding droplet evaporation, the effect of misrepresenting the air jet flow in the model and the subsequent underestimation of particle velocity also needs to be evaluated as the evaporation rate depends on the particle to air flow velocity difference. The evaporation time until full water evaporation has been calculated for a range of particle initial velocity (Table S.A.3). Results indicate that evaporation time does not vary largely according to ejection velocity, whatever the particle size. An increase in droplet initial velocity of 8 times from  $10 \text{ m}\cdot\text{s}^{-1}$  to  $80 \text{ m}\cdot\text{s}^{-1}$  will induce a maximum decrease of  $\sim 37\%$  of evaporation time. Moreover, particles evaporation time is for most particle size much larger than the transport time over the 0.5 m (see Fig. S.A.6.b). We can therefore expect that misrepresenting evaporation may not affect significantly the initial spread and deposition of droplets in the field. However, we must keep in mind that any uncertainties in evaporation prediction may tend to bias the predicted droplet diameter change, therefore leading to uncertainties in deposition and dispersion at further distances.

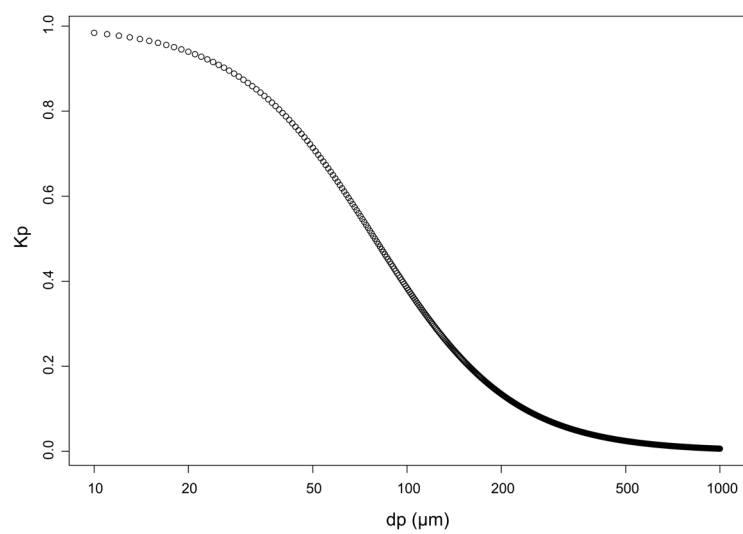
Table S.A.3. Variation of evaporation time (s) until the total water evaporation as a function of droplet initial ejection velocity for eight droplet diameters.

	$D_{p_{ini}} = 300 \mu\text{m}$	$D_{p_{ini}} = 250 \mu\text{m}$	$D_{p_{ini}} = 200 \mu\text{m}$	$D_{p_{ini}} = 150 \mu\text{m}$	$D_{p_{ini}} = 100 \mu\text{m}$	$D_{p_{ini}} = 50 \mu\text{m}$	$D_{p_{ini}} = 30 \mu\text{m}$	$D_{p_{ini}} = 10 \mu\text{m}$
$E_v$ (m/s)	<i>Evaporation time (s) - time to reach the min diameter according to active matter proportion</i>							
<b>10</b>	1.8	1.54	1.28	1.01	0.72	0.4	0.26	0.1
<b>20</b>	1.54	1.33	1.11	0.88	0.63	0.36	0.23	0.09
<b>30</b>	1.41	1.21	1.02	0.81	0.58	0.33	0.22	0.08
<b>40</b>	1.32	1.14	0.95	0.76	0.55	0.31	0.21	0.08
<b>50</b>	1.25	1.08	0.91	0.72	0.52	0.3	0.2	0.08
<b>60</b>	1.2	1.04	0.87	0.7	0.5	0.29	0.19	0.07
<b>70</b>	1.16	1	0.84	0.67	0.49	0.28	0.18	0.07
<b>80</b>	1.12	0.97	0.82	0.65	0.47	0.27	0.18	0.07

A second major effect of the additional turbulence generated by the air jet is to lead to an increased dispersion locally and dilute the plume slightly more than what is represented in the model. We evaluate that this effect is not important for particles larger than  $100 \mu\text{m}$  for which inertia attenuate the dispersion, as measurable by the attenuation factor of the turbulent diffusion from Bouvet and Wilson (2006) use in our model,  $k_p = \frac{1}{\sqrt{1+(1.6 g T_s/u_*)^2}}$ . We see from the Figure S.A.8 that the diffusivity is attenuated by more than 90% for particles larger than  $200 \mu\text{m}$ . For the particle distribution we used in this study, the median attenuation would be around 50%.

A third effect of neglecting the turbulence generated by the air jet is the induced movement of the leaves by the air sprayer. This effect would be very difficult to evaluate and to our knowledge, it is also neglected in all models we have surveyed.

Figure S.A.8. Diffusivity attenuation factor as a function of droplet diameter.







**Annexe B***Évaluation de la distribution des pesticides lors de l'application et de l'exposition des passants à la dérive sédimentaire et aérienne à l'aide d'une approche de modélisation***Assessment of pesticide distribution at application and bystander exposure to pesticide spray drift using a mechanistic modelling approach**

Hulin et al. (2021) reported data concerning the distribution of the spray between the soil, vegetation, and air issued from experts estimations by synthesising the different existing experimental data within research organizations or professional institutes. These data give the distribution of the spray in active substance (%) depending on the type of crop treated (low or high crops), the type of device used (standard or optimized), the vegetative stage (early or developed) and the adjustment performed (average or optimal).

Table S.B. 1. Distribution of sprayed products by spray process, crop type, substance type and crop stage at the time of treatment. (\*)Annual crops including coated seeds, (\*\*) mainly vines and (\*\*\*) Such as arboriculture and some high annual crops such as hops. HA, HB, FIA and FIB correspond to Pre-emerging herbicide treatments (stage A), post-emergence herbicide treatments (stage B), early fungicide and insecticide treatments (stage A) and treatments Normal fungicides and insecticides (stage B), respectively.

		% parc	Annual Crops and Open Field Vegetables(*)												Low Perennial Crops(**)												High Perennial Crops(***)											
			ACOFV												LPC												HPC											
			HA			HB			FIA			FIB			HA			HB			FIA			FIB			HA			HB			FIA			FIB		
			Soil	Target	Air	Soil	Target	Air	Soil	Target	Air	Soil	Target	Air	Soil	Target	Air	Soil	Target	Air	Soil	Target	Air	Soil	Target	Air	Soil	Target	Air	Soil	Target	Air	Soil	Target	Air			
Reference	Spray boom with conventional nozzles (standard)	80	80	0	20	60	20	20	50	30	20	20	60	20	80	0	20	80	0	20							80	0	20	80	0	20						
Optimized	Buffer zone configuration with AI-nozzles (optimum)	20	90	0	10	80	10	20	50	40	10	5	90	5	90	0	10	90	0	10							90	0	10	90	0	10						
Reference	Pneumatic arch multi-row (standard)	80													30	20	50	10	60	30																		
Optimized	Multi-row face to face with AI nozzles (optimized)	19													20	40	40	10	80	10																		
Optimum	Multi-row with recycling panels (optimum)	1													5	90	5	5	90	5																		
Reference	Hollow cone nozzles – axial fan sprayer (standard)	90																									10	20	70	10	50	40						
Optimized	Axial fan sprayer with AI nozzle (optimized)	7																									10	40	50	10	70	20						
Optimum	Crossflow sprayer with AI nozzle (optimum)	3																									10	50	40	10	80	10						
			82	0	18	64	18	18	50	32	18	17	66	17	82	0	18	82	0	18	28	25	48	10	64	26	82	0	18	82	0	18	10	22	68	10	52	38
			100			100			100			100			100			100			100			100			100			100			100					

Source (Hulin et al., 2021)

Table S.B.2. Bystanders dermal and inhalation exposure measured by Mercier (2020) and modelled by the German and Browse models

	Experimental study (µg/kg bw/day)	Browse model (µg/kg bw/day)	German model (µg/kg bw/day)
Spray drift--dermal			
95 <sup>th</sup> percentiles	0.286	7.39	7.24
Spray drift--inhalation			
95 <sup>th</sup> percentiles	0.00327	0.0409	0.00400

Source (Mercier, 2020)

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