Evaluation of human exposure to environmental pollutants through food

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Evaluation of human exposure to environmental pollutants through food

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List of Abbreviations

ANSES	Agence nationale de sécurité sanitaire de l'alimentation, de l'environnement et du travail
BMDL	BenchMark Dose Limit
CALIPSO	Consommations Alimentaires de poissons et produits de la mer et Imprégnation aux éléments traces, polluants et oméga 3
CVUA Sttutgart	Chemisches und Veterinäruntersuchungsamt Stuttgart (Chemical and Veterinary Investigation Office Stuttgart)
DGAL	Direction Général de l'ALimentation
EAT	Etude de l'Alimentation Totale
EDEN	Étude des Déterminants pré et postnatals précoces du développement et de la santé de l'ENfant
EFSA	European Food Safety Authority
EFSA ANS	EFSA Panel on Food Additives and Nutrient Sources added to food
ELFE	Étude Longitudinale Française depuis l'Enfance
ESTEBAN	Étude de Santé sur l'Environnement, la Biosurveillance, l'Activité Physique et la Nutrition
FAO	Food and Agriculture Organization
FI	Food Insecurity
FoodEx	Food Classification and Description System
FSA	Food Standards Agency
INCA	Enquete Individuelle et Nationale sur les Consommation Alimentaire
LB	Lower Bound hypothesis
MB	Middle Bound hypothesis
MOE	Margin Of Exposure
NOAEL	No Observed Adverse Effect Level
PEANUTS	Précarité Etudiant·es Alimentation Nutrition UniversiTé Santé
PNNS	Programme National Nutrition Santé
POP	Persistant Organic Pollutants
SPF	Santé Publique France
TDS	Total Diet Study
TRV	Toxicological Reference Value
UB	Upper Bound hyothesis
US DA	United States Department of Agriculture
US EPA	United States Environmental Protection Agency
WHO	World health Organization

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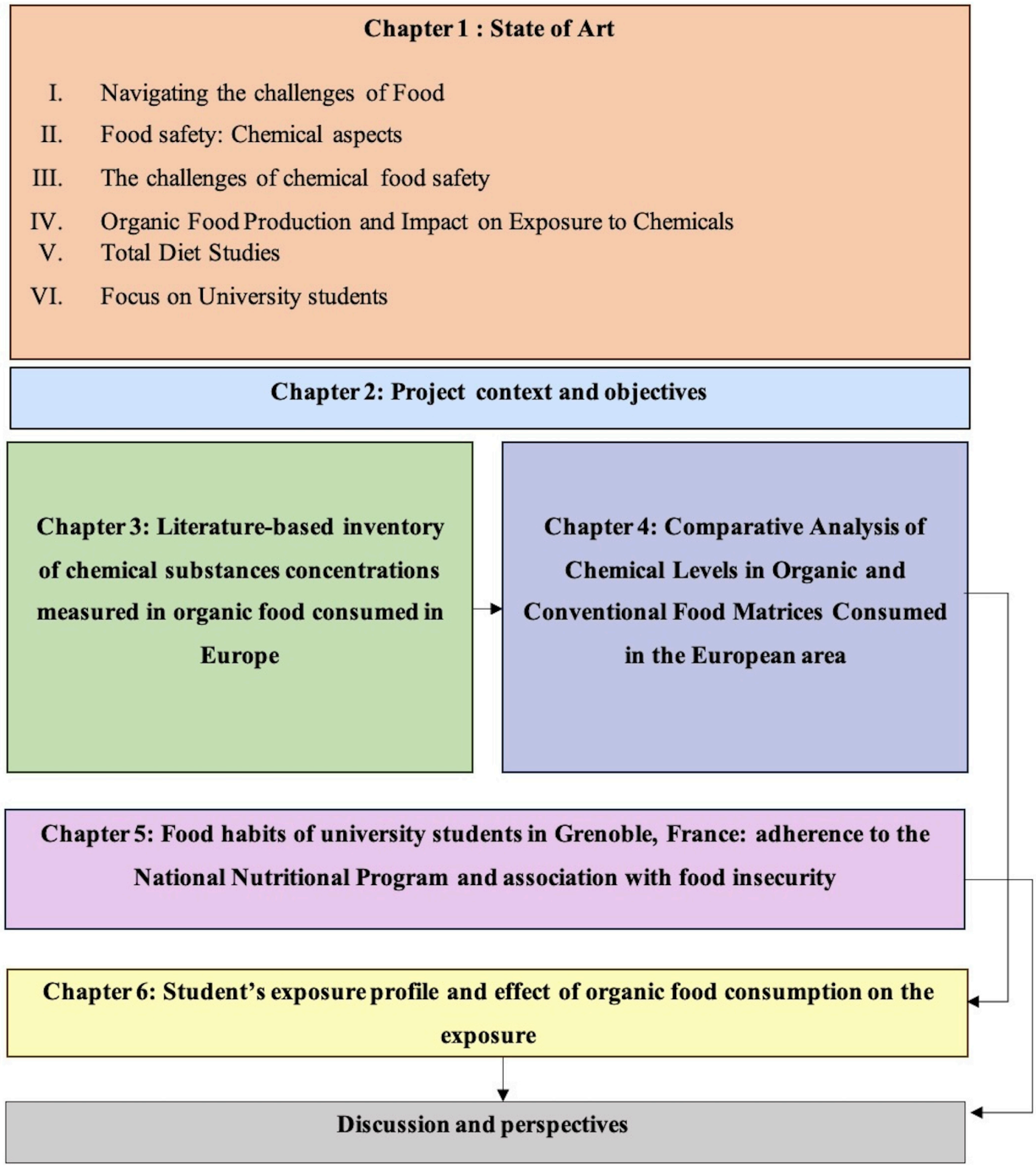

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Chapter 1- State of Art

This chapter aims to offer a comprehensive overview of human exposure to chemical substances through food, shedding a light on the diverse challenges of food and food safety risk assessment along with what was done in the literature to address these concerns. The chemicals studied in this thesis are not detailed in this chapter, yet information on the included substances is provided in the Appendix (A1).

I. Navigating the Challenges of Food

Food, a basic necessity for life, is linked to a complex set of challenges that requires collective and individual actions to ensure a sustainable, balanced food system that guarantees nutritional needs for all (Willett et al., 2019). These challenges include disparities in access to food, concerns about nutritional adequacy, environmental impacts on human health and ecosystems, and food safety concerns, each of which poses unique constraints to ensuring a safe and healthy diet-based future.

1. Ensuring Nutritional Adequacy

Nutrition is the basis of human health and well-being. It provides the essential building blocks for our bodies to grow, develop, and function properly (Lean, 2019). However, for various reasons (e.g., lack of financial resources, poor eating habits), many people do not have access to a diet that is optimal for their health. Achieving nutritional adequacy requires addressing both burdens of malnutrition, which include undernutrition and overweight/obesity (Tzioumis & Adair, 2014).

Undernutrition, characterized by inadequate intake of calories and essential nutrients, affects an estimated 462 million people worldwide, particularly in low- and middle-income countries. It can lead to stunted growth, impaired cognitive development and increased susceptibility to disease (WHO, 2023a).

On the other hand, overweight and obesity have become a global epidemic, affecting 1.9 billion adults (WHO, 2023a). In Europe, obesity is a growing challenge: one in six European citizens is considered obese and more than half of EU adults are overweight (Eurostat, 2016). This is mainly due to unhealthy diets high in processed foods, sugary drinks, and unhealthy fats, combined with limited physical activity (Dey & Kashyap, 2020). Overweight and obesity contribute to a number of chronic diseases, including heart disease, stroke, type 2 diabetes, and certain cancers (Dey & Kashyap, 2020). It is worth mentioning that obesity can be caused by other factors other than unhealthy habits such as endocrine disruptors from environmental pollution. These disruptors interfere with the hormonal regulation of metabolism, leading to inappropriate fat deposition and disrupting the balance of energy use, thus contributing to the development of obesity (Darbre, 2017).

Therefore, the challenge of ensuring nutritional adequacy is critical in the prevention of the double burden of under- and over-nutrition, yet this challenge is not only a matter of food quantity, but also a matter of the maintenance of high food quality. Striking a delicate balance is essential to mitigate the adverse effects of an unhealthy diet and promote optimal health outcomes in diverse populations. National nutrition programs play a critical role in promoting public health by addressing nutritional challenges, educating the public, and implementing strategies for a balanced diet. Examples of national nutrition programs include PNNS (Programme National Nutrition Santé) in France (Manger Bouger, 2022) and Dietary Guidelines for Americans in the United States (US DA, 2020). These initiatives contribute to the overall health of the population by promoting and educating the public about healthy eating habits.

2. Environmental Impact of Food Production

Developments in agricultural science and technology (e.g. mechanization in agriculture) have played a key role in the increase in food production since the mid-20th century. This revolution was necessary to meet the demands of a growing global population and evolving dietary preferences (Chávez-Dulanto et al., 2021), e.g . preferences for a more varied and year-round supply of fruits, vegetables, and grains.

The key challenge for the food production system is to meet this need of increased production while addressing the environmental impacts of the current food system, from production to consumption. Intensified agricultural practices, fueled by escalating demand, have contributed to deforestation, soil degradation, water pollution and greenhouse gas emissions (Fróna et al., 2019). Moreover, excessive use of chemical fertilizers and pesticides aggravates the problem, threatening biodiversity and water quality (Riah et al., 2014). As a result, many agricultural production methods have been developed and coexist today, including organic, biodynamic or sustainable farming. (Reganold & Wachter, 2016; Turinek et al., 2009; Velten et al., 2015).

Not to forget the alarmingly high level of global food waste, estimated at 1.3 billion tons per year, or nearly 40% of all food produced, with the EU, producing nearly 57 million tons of food waste (Dere, 2023). This wasted food decomposes, emitting methane - a potent greenhouse gas (Krause et al., 2023).

Thus, a sustainable food production system, that expands the food supply while simultaneously incorporates social, economic, and environmental considerations, reversing and preventing further resource degradation and environmental impacts is necessary, and achieving it is seen as a challenge for the current food system.

3. Economic Disparities and Food Insecurity

Food insecurity is defined by the Food and Agriculture Organization (FAO) as a state in which people do not have guaranteed access to sufficient, safe, and nutritious food for normal growth, development, and maintenance of an active and healthy lifestyle (FAO, 2023a). This insecurity results from multiple factors such as the unavailability of food, limited purchasing power, inappropriate distribution, or inadequate utilization of food at the household level. One way to measure food insecurity is through the Food Insecurity Experience Scale (FIES), which classifies individuals into three levels based on different degrees of severity: (a) food secure/low food insecurity, (b) moderate food insecurity, and (c) severe food insecurity (FAO, 2023a).

Vulnerable populations most at risk of developing food insecurity include low-income households due to economic disparities, children and the elderly due to their specific nutritional needs and dependence on others for food access (Flores & Amiri, 2019) and university students due to lack of income (Abbey et al., 2022; Davitt et al., 2021; DeBate et al., 2021; Nikolaus et al., 2019).

In addition, rising food prices, exacerbated by inflation, have further exacerbated food insecurity (SPF, 2023). Government policies typically play a key role in influencing food security policies. In France, where inflation has risen to around 6.2% as of February 2023, according to The French National Institute of Statistics and Economic Studies (Insee), many citizens are cutting back on their food spending. In response to this pressing situation, the French Minister of Solidarity unveiled a program called "Mieux manger pour tous" - a comprehensive plan to provide food assistance to the most vulnerable and ensure access to healthy, sustainable, and quality food (SPF, 2023).

4. Ensuring Food Safety: A Multi-pronged Approach

Ensuring food safety stands out as a major challenge within the food system, playing a critical role in protecting public health and preventing undesirable health effects, often caused

by infectious or toxic agents. These illnesses, which can range from mild to severe and can be life-threatening, underscore the need for a robust framework for food safety (WHO, 2022a).

Food safety hazards come in different forms: biological (bacteria, viruses, parasites), chemical (pesticides, heavy metals, industrial chemicals), physical (rocks, insects, packaging residues), and allergens (naturally occurring proteins in foods such as milk, eggs, peanuts (Food Allergy Research and Education, 2024)) (Jackson, 2009). The range of hazards requires a multifaceted approach to food safety. Key components include:

- **Proper food handling:** Educating consumers about good food handling practices, including hand washing, thorough cooking, and safe storage, significantly reduces the risk of foodborne illness (WHO, 2001).
- **Effective food inspection:** Routine inspections of food production and processing facilities by government agencies serve to identify and eliminate potential sources of contamination (Cunha et al., 2022).
- **Traceability systems:** Implementation of traceability systems that track food from origin to consumption facilitates rapid identification and recall of contaminated products (Lin et al., 2022).
- **Enhancing food safety research:** Investing in research on foodborne pathogens, their behavior, and prevention strategies is proving critical to developing effective mitigation measures (USDA, 2023).
- **Promoting sustainable agriculture:** The use of sustainable agricultural practices, including organic farming , not only reduces reliance on chemicals, thereby reducing pollution, but also minimizes the potential for chemical exposure through food consumption (Thomson & Vijan, 2016).

Despite joint efforts and the implementation of these strategies, ensuring food safety remains a complex and ongoing challenge in the food sector. The multifaceted nature of the challenge underscores the ongoing need for innovative approaches and continued attention to protect consumers and maintain the integrity of the food supply chain.

II. Food Safety: Chemical Aspects

The chemicals found in food can be either naturally occurring, such as minerals and phytoestrogens, or linked to environmental pollution caused by human industrial or agricultural activities, such as persistent organic pollutants (POP), plant protection products, inorganic contaminants and others (ANSES, 2011b). These chemicals can also arise from their use or formation during the stages of producing, processing, or preserving raw materials or foods, such as additives or newly formed substances (PAHs). The possibility of contamination can occur at any point in the food chain, illustrating the complexity of implementing contaminant management and monitoring programs (Scanlan, 2007). Details on the chemicals in food evaluated in this thesis, including sources, toxicological effects, and toxicological reference values, are provided in Appendix A1.

In France, ANSES (the French Agency for Food, Environmental and Occupational Health & Safety) primarily ensures health safety in the environmental, labor and food sectors by assessing the health risks they may pose. Further, it provides recommendations for monitoring and controlling chemical contamination of foodstuffs at various stages of the food chain (ANSES, 2019b). It also provides a multi-criteria decision methodology that assists the collective expertise in prioritizing management actions by ranking food hazards or hazard-food pairs based on public health risks and potentially other domains (e.g., socioeconomic, nutritional, environmental) (ANSES, 2020).

1. Contaminants in Food

A. Definition

By definition, a contaminant is a substance that results from the unintentional presence of a substance of natural or synthetic origin in food. It may be a substance used in the production and processing of food, or a substance naturally present in the environment or resulting from environmental pollution (Abraham et al., 2018). The most significant food contaminants include mycotoxins; polychlorinated biphenyls (PCBs), dioxins, persistent unauthorized pesticides, brominated flame retardants, metals such as arsenic, cadmium, lead, and mercury; and process contaminants such as acrylamide and furan (EFSA, 2024a).

B. Evaluation of Human Risks to Contaminants

A toxicological reference value (TRV) is a toxicological index used to qualify or quantify a risk to human health. It establishes the link between exposure to a toxic agent and the occurrence or probability of occurrence of an undesirable health effect. By definition, a TRV is set for the most sensitive adverse effect and thus protects against all toxic effects in the available studies. TRVs are specific to a substance, route of exposure (oral, respiratory, dermal, etc.) and duration (per day, per week, etc.). TRVs are established by international bodies such as the World Health Organization (WHO) or European bodies such as EFSA, and also at national level (e.g. ANSES in France) (ANSES, 2017c).

TRVs are usually classified into two categories:

- **Threshold dose TRVs** are used for substances causing health damage beyond a certain dose, and the severity of their effects increases with the increasing dose (ANSES, 2017c). These TRVs can be expressed as Acceptable Daily Intake (ADI) values or Tolerable Daily Intake (TDI) values. In theory, individuals exposed to a level below these thresholds do not develop an undesirable effect due to these exposures. The difference between ADIs and TDIs lies in the nature of the substance to which we are exposed. ADIs were originally established for food additives and pesticide residues in food, hence the term "acceptable" as they are expected to be food while TDIs are established for substances whose presence in food is unexpected or inevitable, such as environmental contaminants (ANSES, 2012).

- **Non- threshold dose TRVs** concern substances for which a health effect can occur regardless of the dose, and the probability of damage increases with the increasing dose (ANSES, 2017c). These are generally assigned for direct genotoxic carcinogenic effects. The TRV is then expressed as a Unit Risk Estimate (URE), which represents the increase in the probability that an individual exposed to a unit dose of the substance over a lifetime will develop a pathology compared to an unexposed individual (ANSES, 2017c).

Endocrine Disruptors

Some substances do not fall into either of these categories to define a TRV, such as endocrine disruptors (ED), which are substances that interferes with the hormonal system,

causing harmful effects. These include phthalates, bisphenol A, certain pesticides (DDT) etc. As stated, before TRVs are constructed from observed dose-response relationships and is specific to a route, duration, and type of effect, with or without a threshold dose, yet endocrine disruptors have complex mechanisms of action, challenging traditional toxicological analysis paradigms (Demeneix & Slama, 2019).

With regard to EDs, the challenges are:

- **Effects at very low doses:** Endocrine disruptors are suspected to have effects without a threshold, similar to carcinogenic substances, except that the effects are non-monotonic (Vandenberg et al., 2012).
- **Non-monotonous effects:** The dose/response relationship for some endocrine disruptors may not be monotonic; E.g., for bisphenol A, stronger effects were shown at lower doses, even opposite effects to those observed at high doses (U shaped curve) (Vandenberg et al., 2012).
- **Varying susceptibility:** Sensitivity to endocrine disruptors may vary during life periods. The critical "exposure window" mainly corresponds to the gestation period but also includes early childhood and puberty (Vandenberg et al., 2012).
- **Progeny effects:** The consequences of endocrine disruptors go beyond the exposed parents, manifesting in subsequent generations (Ruaux, 2012).

Benchmark Dose Limit

Another method for evaluating the risk to contaminants is through **the Benchmark Dose Limit ($BMDL_x$)** which is the dose level at which the observed change in response is expected to be less than x%, with the term "likely" determined by the statistically credible level, typically set at the 95% confidence level (EFSA, More, et al., 2022). The response increase “x%” is set to either 1%, 5% or 10% of the change in the adverse impact response rate compared to the control group's reaction rate (U.S.EPA & Hogan, 2012).

This approach is applicable to all food chemicals, **regardless their category or origin**, and is particularly useful in uncertain NOAEL identification, **providing a reference point to genotoxic and carcinogenic effects**. EFSA Scientific panels and units are encouraged to adopt this approach (EFSA, 2009b) .

As for the risk characterization, this approach requires the calculation of a Margin of Exposure (MOE) which is the BMDL value divided by the level of exposure to the substance. Based on the $BMDL_{10}$, EFSA suggested a method to evaluate health risks through the MOE: "an MOE of 10,000 or higher is considered of low concern from a public health perspective and may be deemed a low priority for risk management actions and vice versa" (EFSA, 2005b). However, EFSA's guidelines lack clarity regarding the appropriate MOE when $BMDL_{01}$ or $BMDL_{05}$ values from animal studies or BMDLs from human studies are available.

In this thesis, lead and arsenic were evaluated using the BMDL approach. Yet only $BMDL_{01}$ values were determined for these substances. Consequently, for the evaluation, the same methods applied in the EFSA reports for lead and arsenic in the scientific opinion reports (EFSA Journal, 2010, 2021), were applied in chapter 6. These were also the methods applied in EAT2 (ANSES, 2011b). For lead, an MOE greater than 10 is sufficient to ensure that there is no risk of prevalence of cardiovascular or nephrotic effects (EFSA Journal, 2010). As for arsenic, no specific reference point was identified for MOEs; instead, exposure levels were compared to the $BMDL_{01}$ range (0.3-8 µg/kg bw/day). If exposure values fell within this range, indicating a small to no MOE, this implied that the possibility of excluding the risk was unlikely (EFSA Journal, 2009, 2021).

2. Chemical residues in Food

A. Definition

Chemical residues in the context of food refer to substances that have been intentionally added during the production process with specific purposes in food production. These include plant protection products like authorized pesticides, and veterinary drugs, serving as safeguards against harmful organisms or pathogens. Moreover, residues include food additives, which are added to either preserve quality or extend shelf life (e.g., sulfites, nitrites, and nitrates), enhance nutritional value (e.g., iron), or improve the texture and appearance (e.g., rocou) as well as the taste (e.g., tartaric acid) of the food product (Abraham et al., 2018). Although deliberately added to food for specific functions, the residues of these substances can still potentially pose a health risk to consumers if used without controls. Residues assessed in this thesis include authorized pesticides (deltamethrin, pyrethrin, λ-cyhalothrin, pirimiphos- methyl), copper, and sulfites. Toxicological information about these substances are found in the appendix (A1).

B. Regulatory Standards for Residue Management in Food Production

Residue levels are evaluated through a Maximum Residue Limit (MRL), which refers to the maximum concentration of a substance's residue that is allowed to be found in or on food products. These MRLs are established in accordance with European regulations by domain and are defined for specific pairs of matrix (fruit, vegetable, or cereal) and substance residue (ANSES, 2017b). The MRLs are set well below toxicological thresholds, ensuring that the amounts of residues an individual might encounter daily in their diet are never toxic, neither in the short term nor the long term (French Ministry of Agriculture and Food, 2013).

In France, food safety and quality, as well as animal and plant health and protection, are ensured by the General Directorate for Food (DGAL). This regulatory agency works with various stakeholders, including agricultural professionals, veterinarians, nonprofit associations, consumers, and sanitary agencies such as ANSES, to monitor the entire food chain. The DGAL formulates regulations that define its primary missions (monitoring), and ensures their effective implementation by relying on decentralized services in different departments and regions (French Ministry of Agriculture and Food, 2023).

On a European level, EFSA assesses risks associated with these residues and provides scientific opinions to the European Commission. The Commission then establishes the MRLs for various substances in food products. For pesticides, Regulation (EC) No 396/2005, commonly referred to as the MRL Regulation defines these MRLs for various commodities treated with pesticides. Furthermore, for additives, Directive 95/2/EC establishes a maximum permitted level of these substances to ensure the health safety of consumers.

III. The challenges of chemical food safety

1. Exposome and Cocktail Effects

The environmental factors have a major impact on human health: the World Health Organization (WHO) estimates that these factors are responsible for 12.6 million deaths each year. In Europe, the organization estimates that they are responsible for at least 15% of mortality, or about 1.4 million deaths per year (WHO, 2016).

Consequently, the study of exposure to environmental influences stands as a crucial aspect of public health. The term "exposome" was first used in 2005 by Christopher Wild, a researcher at the University of Leeds in the United Kingdom, to define the history of our environmental exposures from conception to death. At the time, this definition met with little response from the scientific and medical communities. Subsequently, it has been expanded (G. W. Miller & Jones, 2014; Rappaport, 2011; Vermeulen et al., 2020), and the exposome is now defined as the set of quantifiable and cumulative environmental exposures and associated biological responses that occur throughout life and that may influence our health status. In 2023, in its opinion on the integration of the exposome into its activities, ANSES defined the exposome as the totality of both harmful and beneficial exposures to chemical, biological and physical agents, in interaction with the physiological status, the living environment and the psychosocial context experienced by a living organism from the conception to death, in order to explain its state of health (ANSES, 2023a).

The dynamic nature of an individual's exposome poses many challenges in terms of characterization especially that it is based on a time component (from conception to death), with some life stages having higher vulnerabilities such as pregnancy, childhood, and puberty. Additionally, its **multifactorial nature**, incorporating factors of different types (socio-economic, geographical, demographic factors, as well as chemical, physical, microbiological, pharmaceutical, lifestyle, dietary, and infectious environmental factors), also complicates the study of the exposome (Jégou, 2020). Moreover, the ability to measure all past exposures is limited. Thus, today, there is a challenge to develop methodologies to characterize the exposome to link it with health data (Maitre et al., 2022).

Within the exposome**, food** plays a special role both as a vital source of essential nutrients and as a vector for contaminants and residues (Hennig et al., 2012). Multiple contaminants can be present in the same food, presenting a challenge for risk assessment and management.

Currently, most risk assessments in the literature focus on individual substances or families of substances, overlooking potential combined effects, known as the "cocktail effect".

The mixture effect of "cocktail effect" is therefore a major challenge in risk assessment studies. Depending on the case, the effect of being exposed to a mixture of substances may be:

1. **Antagonistic** (the effect of the combination is less than the effect of one of the substances),

2. **Additive** (the combined effect is the sum of the two isolated effects),

3. **Synergistic** (the effect of the combination is greater than the sum of the effects of the substances in isolation).

Currently, due to a lack of data on interaction evidence in mixtures, the additive effect is recommended as the default assumption for assessing the risks of mixtures (EFSA et al., 2019). Methodologies exist, especially for active substances with either the same toxic action mechanism or a similar mechanism or for molecules from the same chemical class (EFSA, 2021a). Nevertheless, these methodologies fall short of fulfilling all expectations, prompting ongoing efforts on this subject.

An individual's exposure, outside the occupational context, occurs through **chronic exposure** to a multitude of substances, meaning daily exposure to low doses throughout life. The long-standing toxicological literature on exposure to environmental contaminants is abundant, but often focuses on high doses, unrealistic mixtures, and often irrelevant routes of administration. Today, researchers are increasingly developing realistic exposure models (low doses, complex mixtures) to assess and understand the impact of environmental contaminants on human health, especially via food, which is a major source of exposure.

2. Vulnerable Populations

Chemical exposures pose significant risks to human health, and certain segments of the population are particularly vulnerable to their adverse effects. Vulnerable populations, notably pregnant women, children under 3 years of age (ANSES, 2011b), and elderly individuals (Risher et al., 2010), are disproportionately impacted by the adverse effects of chemical exposures due to unique physiological vulnerabilities at different life stages. Recognizing and addressing the heightened susceptibility of these groups is imperative for formulating targeted interventions and safeguarding the health of those most at risk.

In France, several studies have investigated the dietary exposure and impregnation to environmental pollutants in pregnant women and infants, as summarized in Table 1.

Table 1. Characteristics and results of studies on pregnant women and infants

Study	Year	Population	Results	Ref.
EDEN	2003/2006	2002 pregnant women and their children up to age 5	*Risk not to be excluded for acrylamide inorganic arsenic, lead, and BDE-99 *Significant exceedance of TRVs for NDL-PCBs, T-2 and HT-2 toxins, and deoxynivalenol before pregnancy, but not in the third trimester	(Chan-Hon-Tong et al., 2013)
ELFE	2011	18000 French pregnant women and their children followed from birth to age 20	*TRVs exceeded for Lindane (with 2.4% under LB hypothesis), and Heptachlor, Dimethoate, Dieldrin, Carbofuran and Diazinon (only under UB hypothesis) *Significant cumulative risk for neurological effects in children associated with high exposure during pregnancy to chlorpyrifos, pyrimiphos-methyl and dimethoate (fruits and cereals)	(de Gavelle et al., 2016)
SEPAGE	2014/2017	484 pregnant women and their children	*Phenols and phthalates frequently detected in urine samples *Evidence of an association between exposure to phenols and phthalates during pregnancy and an increase in scores indicating greater risk of behavioral problems in two-year-old girls	(Guilbert et al., 2021)

EDEN : Étude des déterminants pré et postnatals du développement de la santé de l'enfant

ELFE : Étude Longitudinale Française depuis l'Enfance

SEPAGES : Suivi de l'Exposition à la Pollution Atmosphérique durant la Grossesse et Effets sur la Santé

BDE99 : 2,2′,4,4′,5-PentaBromoDiphenyl Ether

T-2 and HT-2 toxins: Fusarium mycotoxins

Moreover, of the three total diet studies conducted in France, only one, the French Total Diet Study for Infants (TDSi), focused specifically on vulnerable populations such as infants and children (ANSES, 2016), underscoring the importance of expanding research efforts to address the unique vulnerabilities of these groups. These studies provide insightful results that underscore the need for additional research on a wider range of substances, also including populations that have not been included in exposure studies, such as the elderly. In addition to vulnerable populations, it could be interesting to study specific populations with particular diets for different reasons (food insecurity, particular conviction...), which may lead to overconsumption of foods that could be the cause of overexposure compared to the general population.

3. Benefit-Risk Balance

While assessing the exposure profile of chemicals from food is important, it's equally important to consider the inherent benefits of food in order to balance the benefits and risks. Despite the relevance of this perspective, comprehensive risk assessment studies addressing both aspects are notably lacking in the literature.

One notable study that has addressed both dimensions is the **CALIPSO** study (étude des Consommations ALimentaires de produits de la mer et Imprégnation aux éléments traces, PolluantS et Oméga 3), which was conducted between 2003 and 2006 on populations from four coastal sites in France. These sites were chosen because they represent significant consumers of fish and seafood products. The study aimed to assess the dietary intake of fish and seafood products, as well as levels of biological impregnation of individuals with trace elements, pollutants, and omega-3 fatty acids.

As a result, CALIPSO showed that the contamination levels in seafood were generally below the risk thresholds set by regulations. Oily fish, such as salmon, mackerel and sardines, were identified as major contributors of persistent organic pollutants (POPs), while providing high levels of beneficial omega-3 fatty acids.

This study makes a significant contribution to better characterizing both the benefits and risks associated with seafood consumption. Key findings from CALIPSO include: for the general population, the consumption of fish should be at least twice a week, including oily fish. For pregnant or lactating women, the consumption of predatory fish should be limited to once a week. In addition to these general recommendations, this study highlights the importance of

diversifying the species of fish and seafood products consumed, both in terms of proportions and origin, to ensure a well-considered balance between the benefit and risk components that is compatible with nutritional recommendations and toxicological considerations (Leblanc et al., 2006).

4. Exposure from Different Food Production Methods

Since the end of World War II, conventional agriculture has been the dominant agricultural system worldwide. The widespread adoption of this mode of production has had negative environmental and economic consequences (Fouilleux, 2015). Environmental degradation is manifested through widespread pollution, including the depletion of soil fertility in agricultural regions due to the extensive use of pesticides, heavy reliance on fossil fuels, and a significant contribution to global warming. In addition, the conventional agricultural system exposes both farmers and consumers to potentially harmful chemicals. Farmers are chronically exposed to moderate or high but repeated doses of pesticides during their agricultural practices (Damalas & Koutroubas, 2016), while consumers are exposed to residues of these pesticides through the food supply, with potential health implications (WHO, 2022b).

In response to these issues, various alternative forms of production have emerged that are perceived to be more sustainable and safer. Among these alternative, the most developed at present are organic, biodynamic, and integrated farming (Muhie, 2022). However, despite the growing interest in these alternative farming systems, comprehensive studies on the impact of these practices on human chemical exposures remain scarce, as food risk assessment studies have mostly focused on exposures from conventional foods.

Within the existing literature, studies of biodynamic and integrated farming (sustainable farming) practices have emphasized their positive environmental outcomes and sustainability attributes (Brock et al., 2019; Walia & Kaur, 2013). The focus of these studies tends to be on the pro environmental impacts of these farming methods, with limited attention paid to a comprehensive assessment of their impact on human exposure.

Furthermore, few studies on biodynamic farming showed a notable focus on the increased nutritional quality of products derived from this agricultural approach, as observed in a study where the consumption of dairy products of biodynamic origin is correlated with higher nutritional quality of breast milk in lactating women (Simões-Wüst et al., 2011), and another

showing higher phenolic acid and antioxidants in biodynamic strawberries instead of conventional ones (Tarozzi et al., 2010).

On the other hand, organic farming, which is the most common alternative to conventional production, occupying 10.7% of the agricultural land in France (Agence BIO, 2022), has received more scientific attention regarding its effects on exposure. The prevalence of research on organic farming compared to other types of sustainable farming underlines its importance within the discourse on sustainable agriculture. The following part (IV. Organic Food Production and Impact on Exposure to Chemicals) will address organic food farming practices and their impact on human exposure, in line with existing literature.

Today, there is a gap in the literature in understanding the impact of different alternative methods to conventional practices on human exposure to chemicals, requiring extensive research to ensure a balanced assessment of different methods and to provide evidence-based policies for sustainable agriculture.

5. Emerging Contaminants

Emerging contaminants constitute a group of recently identified or new concerning substances that are characterized by their toxicity, cumulative nature, and persistence in the environment. In addition, their production and use are not effectively managed, their environmental fate is incompletely understood, and their environmental toxicology and health risks are not comprehensively studied (Gavrilescu et al., 2015; U.S.E.P.A. Federal, 2008). Globally recognized emerging contaminants include persistent organic pollutants such as organochlorine pesticides and polybrominated diphenyl ethers, endocrine disruptors such as perfluorinated compounds, personal care products, engineered nanomaterials (ENMs), antibiotics, and resistance genes (Li et al., 2022).

The concentrations of emerging contaminants in the environment are typically too low to detect, making it challenging to accurately assess their environmental health risks and implement effective management strategies (Gavrilescu et al., 2015). Additionally, once a new contaminant enters the environment, a series of biogeochemical behaviors can occur, including transfer, transformation, and transmission or accumulation along the food chain, resulting in property changes that increase the difficulty of its detection and modify its environmental health effects.

One example is **perfluorooctanoic acids (PFOAs)**, which can be absorbed by soil or dust particles, inhibiting their migration and bioavailability and reducing their environmental risks (Lyu et al., 2018). Another example is the environmental aging of **microplastics**, which leads to the co-migration of coexisting contaminants and the release of inherent components, such as plastic nanomaterials, resulting in more complex environmental risks associated to microplastics (Romera-Castillo et al., 2018; Song et al., 2020; Town & van Leeuwen, 2020).

While many studies in the laboratory have demonstrated the predominantly adverse health effects of emerging contaminants, including nervous system disorders, cytotoxicity, and genotoxicity (Nel et al., 2006; Pereira et al., 2015; Rocco et al., 2015), it's important to note that these results are often derived from exposure experiments using single organisms, elevated contaminant concentrations, and controlled environmental conditions. These controlled settings may not reflect the true environmental risks posed by emerging contaminants in natural, uncontrolled settings, hence a big effort to study the toxicology of these substances is yet to be done.

6. Evolution of dietary habits

As dietary habits evolve, new methods must be adapted to assess the risks associated with new food trends. One example is the inclusion of insects. With the growing demand for nutritious, environmentally friendly foods, edible insects are gradually finding their way onto Western plates. FAO considers insects to be a serious alternative to animal protein, as natural resources become limited, and the world's population is growing (FAO, 2023a).

In France, for several years now, pioneering companies such as Jimini's and Micronutris have been marketing edible insects: roasted for aperitifs, ready to cook, or powdered into protein bars, cookies, and flour. Forecasts by Meticulous Research suggest that European sales of edible insects could exceed $2.98 Billion by 2030 (Meticulous Research, 2023).

However, there is currently a significant knowledge gap regarding the potential chemical substances associated with the use of insects in food and feed due to the lack of systematically collected data on animal and human consumption of insects (EFSA, 2015). The risk assessment through insect consumption is challenging for researchers, mainly due to the quality and availability of data, which are still scarce, and the great diversity among insect species (EFSA, 2021b). Consequently, there is a need to develop appropriate analytical methods and conduct more studies to generate data on this issue.

Another dietary trend is the increased consumption of organic foods. To date, from what is found in the literature, the impact of organic food consumption is not yet conclusive, especially with the limited and heterogeneity of available data and the limited evaluation of exposure studies from organic food. This will be discussed further in the following chapter.

IV. Organic Food Production and Impact on Exposure to Chemicals

1. Organic Food Production and Benefits

Organic agriculture is a worldwide agricultural production system that combines the most effective environmental practices, upholds biodiversity, conserves natural resources, and maintains high standards of animal welfare (Remongin, 2023).

In Europe, organic agriculture has seen a progressive increase from 14.7 million hectares in 2020 to 15.9 million hectares in 2021, **i.e.,** 9.9% of the total agricultural area, with France having the largest organic production area in the EU with 2.8 million hectares in 2021 (Eurostat, 2023).

Organic agriculture has been expanding mainly because it has been shown to have many positive impacts on the environment, making it a sustainable development option (Muhie, 2022). It benefits biodiversity and the environment in several ways. According to the Food and Agriculture Organization (FAO), organic agriculture promotes interactions within the agro-ecosystem, resulting in benefits such as soil formation and conditioning, waste recycling, carbon sequestration, and reduced groundwater pollution (FAO, 2023b). In addition, organic agriculture reduces the use of non-renewable energy, contributes to the mitigation of the greenhouse effect, and reduces the need for agrochemicals, thereby reducing the risk of soil and water pollution (Taghikhah et al., 2021).

Furthermore, in addition to its positive impact on the environment, many studies have shown that organic agriculture produces food with higher nutritional quality, such as: higher antioxidant content content (Barański et al., 2014; Das et al., 2020; Lou et al., 2020), total polyphenols (Tarozzi et al., 2010), and certain vitamins (Breza-Boruta et al., 2022; Çakmakçı & Çakmakçı, 2023). While organic agriculture offers many advantages, aspects such as chemical content need further study for continuous improvement.

2. Organic Food Regulations

Organic farming is subject to strict regulations. In the EU, organic farming and production rules are enforced by Regulation (EC) N°848/2018 (European Parliament And Council, 2018). This regulation prohibits the use of chemically synthesized plant protection products and

requires the use of specific natural fertilizers and natural pest control methods, the exclusive use of organic seeds while avoiding GMOs, the recycling of organic matter, the use of crop rotation, the preference of plant varieties adapted to local conditions, special attention to animal welfare, and the limited use of drugs for therapeutic purposes ((EC) N°848/2018). In addition, these regulations provide extensive controls on labeling and a list of natural substances authorized in organic farming, such as azadirachtin, spinosad, copper and pyrethrin ((EC)N°2021/1165).

3. Chemical substances in Organic Food

Although of natural origin, these authorized substances may have potential effects on human health and/or the ecosystem.

- **Spinosad**, an insecticide used in organic farming, is derived from the industrial fermentation of an actinomycete bacterium naturally present in the soil called *Saccharopolyspora spinosa*. It is a neurotoxicant that is active through ingestion and contact but is not systemic. The 2018 peer review published by EFSA highlights the potential reprotoxic and endocrine-disrupting effects of spinosad (EFSA, 2018c).
- **Azadirachtin**, a metabolite found in the oil extracted from *Azadirachta indica* (neem tree) seeds, is used as an insecticide. This molecule poses a danger to bees and can cause endocrine disruption by feminizing male insects, as well as liver and lung damage in some mammals. The endocrine disrupting effect has led EFSA experts to propose categorizing this substance as suspected of being toxic for human reproduction (category 2 for reproduction) ((EFSA, 2018b).
- **Pyrethrins** are substances derived from *Chrysanthemum indicum*, commonly called Dalmatian pyrethrum flowers. They act by inhibiting sodium channel repolarization in organisms that ingest them. This mechanism targets the nervous system of insects but can also be toxic to fish (Anadón et al., 2009).

Therefore, these substances, which are specific to organic farming, require special monitoring.

Moreover, the literature showed that there are also ubiquitous environmental contaminations in organic food, over which we have no control, including heavy metals (Debnath et al., 2015; Malmauret et al., 2002), Persistent Organic Pollutants such as dioxins, and PCBs (Almeida-González et al., 2012; Dervilly-Pinel et al., 2017; Witczak & Abdel-

Gawad, 2012), inorganic contaminants (cadmium, lead and arsenic) (Hoefkens et al., 2009; Malmauret et al., 2002), brominated compounds (Dervilly-Pinel et al., 2017), and also pesticides authorized in conventional agriculture that persist in the environment, and consequently, contaminate organic products (Mao et al., 2021; Romero-González et al., 2011).

Some studies compared contaminants levels in between organic and conventional food. Results were quite variable depending on the matrices and the chemical substances. For example, in Almeida-González et al., medians for the sum of organochlorine (OC) pesticides were found to be three times lower in organic cheese (14.44 ng/g fat vs. 42.73 ng/g fat in conventional cheese, with p=0.001), while PCBs were found to be 2.4 times higher in the organic batch (22.55 ng/g fat vs. 9.57 ng/g fat in the conventional batch, p=0.074) (Almeida-González et al., 2012). Another study showed significantly higher levels of dioxins, PCBs, HBCD, zinc, copper, cadmium, lead, and arsenic found in organic bovine, porcine and chicken meat samples compared to conventional ones (Dervilly-Pinel et al., 2017). Moreover, two studies showed on cadmium, showed that levels were significantly higher (up to 2-fold) in organic spinach and carrots compared to conventional samples, but significantly lower (about 1.2-fold) in organic potato and lettuce samples (Hoefkens et al., 2009; Malmauret et al., 2002).

In conclusion, although regulations promote synthetic chemical-free production in organic food, there is still a potential for the presence of chemicals that may impact human health. Currently, studies investigating chemical content in organic foods are few. Further, the databases available in the literature or from health agencies, although very extensive in terms of the number of substances studied in conventional food, contain little or no data on the contamination of organically farmed foods. This was evidenced in the latest EFSA report, where organic foods represented only 6.5% of all the samples tested (EFSA, 2022). This scarcity underscores the need for further research, as findings from available literature encourage a thorough investigation into the potential impact of organic food consumption.

4. Impact of Organic Food Consumption on the exposure to chemicals

Although the French National Nutrition and Health program (PNNS) recommends that the general population consume at least 20% of their fruits, vegetables, cereals, and legumes in the form of organic food (Ministère des solidarités et de la santé, 2019), there have been **few studies conducted to assess the impact of organic food consumption on exposure to chemicals from food.**

Among the studies carried out is that of Baudry et al. in 2021 as part of the **BioNutriNet** study, which included 33,018 omnivores, 555 pescetarians, 501 vegetarians, and 368 vegans. The data on pesticide levels in plant-based products were obtained from an official food control laboratory in Germany, the CVUA of Stuttgart. Exposure levels were calculated based on two scenarios: a 100% organic scenario and a 100% conventional scenario. The findings revealed that adopting a 100% organic diet led to a nearly six-fold increase in exposure to azadirachtin, an eight-fold increase in exposure to spinosad, and a 0.5-fold increase in exposure to pyrethrin compared to a 100% conventional diet—these being substances permitted in organic production. Conversely, for other pesticides, including acetamiprid, chlorpyrifos, dimethoate, and glyphosate, exposures decreased by 3 to 10 times in the 100% organic scenarios (Baudry et al., 2021).

Moreover, the **ESTEBAN** study, published by Public Health France as part of the national biomonitoring program, measures exposure levels to 5 pesticide families as well as other contaminants. This study demonstrated that the consumption of products from organic farming resulted in a decrease in impregnation to organochlorines, DMTP (metabolite of organophosphates), and pyrethroids (SPF, 2021c, 2021b, 2021a).

With the lack of literature-based evidence, and with the increasing growth of organic food consumption, it necessary to increase research to understand the impact of this production from a chemical exposure point of view.

V. Total Diet Studies

1. Results from the French Total diet Study 2

The Total Diet Study (TDS) is a cost effective and reliable method for analyzing substances in food to assess exposure. In 2011, a guidance document on a harmonized approach for the assessment of dietary exposure to chemicals, known as the TDS, was jointly published by the European Food Safety Authority (EFSA), the Food and Agriculture Organization of the United Nations (FAO) and the World Health Organization (WHO) (EFSA et al., 2011b).

The design of a TDS includes three criteria: (i) representativeness of the dietary habits of a population, (ii) food preparation as consumed, and (iii) pooling of similar foods. Hence, for sample preparations, domestic cooking techniques are used to depict possible food composition effects, such as degradation/compound formation. Consequently, a TDS provides a

comprehensive database of average levels of substances in food that can be used to assess chronic dietary exposure (EFSA et al., 2011b).

In France, in addition to the previously mentioned TDSi targeting infants and toddlers, two TDSs have been conducted to date, TDS1 (EAT1) and TDS2 (EAT2).

TDS1 in 2005 considered children (3-14 years) and adults (15+ years) and assessed their exposure to 30 inorganic contaminants, minerals and mycotoxins (Leblanc et al., 2005). TDS2 in 2011 also considered a general population of children and adults aged 3-79 years and assessed them for 445 substances present in food using consumption data from the INCA 2 study (étude Individuelle Nationale des Consommations Alimentaires) conducted every seven years by the ANSES (ANSES, 2011b, 2011c).

TDS2 sampled and analyzed 212 types of foods that were selected on the basis of the food products that are most frequently consumed by the French population and/or the food products that are most likely to be contaminated. The samples were purchased and prepared as consumed for analysis. It should be noted that the prepared samples included a proportion of organic foods that reflected the population's consumption of organic foods, but the proportion of organic foods was not specified, making it impossible to distinguish exposure from different types of food production.

The results of these analyses were used to calculate exposure levels to the chemical contaminants studied and then compared with toxicological reference levels for risk assessment. As a result, a risk hierarchy scheme was defined, classifying the substance according to the level of concern:

- **Risk that can be excluded**: TRV not exceeded or high MOE (for substances with non-threshold effects)
- **Risk cannot be excluded**: TRV exceeded or low MOE (for substances with non-threshold effects)
- **Impossible to conclude:** No robust TRV or TRV exceeded under upper bound hypothesis, overestimating exposure.

Table 2, taken from the TDS document, summarizes the results for the substances classified as high risk for the French population.

Table 2. Substances for which toxicological risk cannot be ruled out for the French population, table retrieved from the French TDS document (ANSES, 2011b)

Category of substances	Substances	Population concerned	Main contributing foods
Inorganic contaminants	Lead	Adults and children most at risk	Adults: alcoholic beverages (14%), bread and dry bakery products (13%), water (11%) Children: water (11%), milk (11%), non-alcoholic soft drinks (10%)
	Cadmium	<1% of adults and 15% of children	Adults: bread and dry bakery products (22%), potatoes (12%) Children: potatoes (14%), bread and dry bakery products (13%)
	Inorganic arsenic	Adults and children most at risk	Adults: water (24-27%), coffee (14- 16%) Children: water (19-24%), milk (11- 17%), non-alcoholic soft drinks (10-12%)
	Aluminum	<1% of adults and 2% of children	Adults: Hot drinks excluding coffee (13%), Vegetables excluding potatoes (11%) Children: Vegetables excluding potatoes (8%), pasta (7%), pastries and cakes (6%)
	Organic mercury (methylmercury)	High consumers of tuna (<1% of adults and 1% of children)	-
Dioxins and PCBs	Dioxins and PCB-DL	<1% of adults and 1% of children	Adults: fish (20%), butter (20%) Children: butter (20°4), fish (14%)
	PCB-NDL	<1% of adults and 2% of children	Adults: fish (37%), butter (11%), cheese (11%), ultra-fine products (11%). fresh dairy products (11%) Children: fish (30%), butter (12%), meat (11%)
Additives	Sulfites	Heavy wine consumers (3% of adults)	-

Mycotoxins	DON and derivatives	<1% of adults and 5 to 10% of children	Adults: bread and dry bakery products (60%) Children: bread and dry bakery products (40%)
Neo-formed substances	Acrylamide	Adults and children most at risk	Adults: fried or sautéed potatoes (45%), coffee (30%) Children: fried or sautéed potatoes (61%), cookies (19%)
Pesticide Residues	Dimethoate	High consumers of cherries (<1% of adults and children)	-

2. Results from the European Total Diet Studies

Total Diet Studies have been conducted in over 33 countries worldwide (EFSA, FAO and WHO, 2011). In addition to France, several European countries have conducted TDSs, the most recent of which are Germany, Italy, and the United Kingdom.

A. The BfR MEAL Study (Germany)

Funded by the German Federal Ministry of Food and Agriculture (BMEL), the BfR MEAL study is the first Total Diet Study (TDS) conducted in Germany by the Federal Institute for Risk Assessment (BfR) (Sarvan et al., 2017).

Preparations for the BfR MEAL began in 2015. In total, the BfR MEAL study will examine around 356 different foods for almost 300 contaminants and residues. In particular, the German TDS distinguishes exposures between organic and conventional food matrices (Sarvan et al., 2017).

Risk assessment results are not yet complete, but a study on **chronic dietary exposure to organic and inorganic arsenic (iAs)** has been published based on the results of the German TDS. Dietary exposure was estimated using occurrence data from the BfR MEAL study combined with the corresponding consumption data of the German population. The exposure was calculated at the individual level related to the individual's body weight.

For chemical occurrence data, if the analytical technique did not detect or quantify a substance in a significant proportion of the food samples analyzed, two hypotheses were used to estimate exposure:

1- *The modified lower bound (MLB) hypothesis*, where results below the LOD were set to zero and results below LOQ and above LOD were replaced by the value reported as LOD.

2- *The upper bound (UB) hypothesis*, where results below the LOD are replaced by the value reported as the LOD. Results below the LOQ and above the LOD are replaced by the value reported as the LOQ.

As such, the MLB hypothesis underestimates levels and therefore exposure, whereas the UB hypothesis increases levels and therefore exposure and is therefore conservative in terms of risk assessment.

In the case of quantified substances, the MLB and UB represent the same value, which corresponds to the quantified value.

The results showed that a risk cannot be excluded for iAs due to the small margin between the estimated dietary exposure to iAs and the identified toxicological reference points for the German population. The highest median exposures to iAs were found in infants (0.5 to <1 year) under the MLB hypothesis and in young children (1 to <2 years) under the UB hypothesis (0.17-0.24 µg. kg^{-1} $BW.day^{-1}$ and 0.26-0.34 µg. kg^{-1} $BW.day^{-1}$, respectively). Cereals, especially rice, are the major contributors to total dietary exposure to arsenic and iAs for all age groups (Hackethal et al., 2023).

Another study assessed the dietary exposure of the German adult population to aluminum, copper, methylmercury, manganese, and lead based on BfR MEAL data. None of the exposure levels to the elements assessed exceeded the corresponding TRVs (Kolbaum et al., 2019).

In addition, three reports were published on the levels of contaminants found in the analyzed foods from the BfR MEAL for dioxins and dioxin-like PCBs (Stadion et al., 2022), mercury, cadmium, lead, and nickel (Fechner et al., 2022), and arsenic species (Hackethal et al., 2021).

B. The Italian Total Diet Study 2012-2014

The Italian TDS collected more than 3000 food samples to be representative of the whole diet of the population, prepared as it is consumed and grouped into 51 food groups, thus modeling the Italian diet. The samples were then analyzed for the presence of 65 substances, including 9 trace elements, 35 dioxins and PCBs, 9 mycotoxins and 4 radionuclides.

Similar to the French TDS study, risks could not be excluded for cadmium, methylmercury, lead, aluminum, inorganic arsenic, and dioxins and DL- PCBs. In addition, this study also showed a level of concern for nickel and some mycotoxins (Cubadda et al., 2016, 2020, 2020; D'Amato et al., 2013).

The results for trace elements and dioxins of high concern for the Italian population are summarized in Table 3.

Table 3. Trace elements and dioxins for which toxicological risk cannot be ruled out for the Italian population from TDS 2012-2014 (Cubadda, 2022)

Category of substances	Substances	Population concerned	Main contributing foods
Inorganic contaminants	Cadmium	21% of adults and 83% of children	Cereals and cereal products (35%), vegetables (29%), fish and seafood (15%), potatoes and tubers (13%)
	Aluminum	4% of adults and 34% of children	Vegetables (36%), Cereals and cereal products (29%), sweet products (9%)
	Lead	7% of population and children most at risk	N/A
	Inorganic arsenic	Adults and children most at risk	Cereals and cereal products (35%), water and non-alcoholic beverages (28%), vegetables (11%)
	Organic mercury (methylmercury)	22% of the population	Fish and sea food (100%)
	Nickel	12% of the population	Adults: cereals and cereal products (28%) (particularly bread (13%) & pasta (16%), sweet products (11%), water and non-alcoholic beverages (11%), potatoes and tubers (8%), pulses (6%), fruit (6%) and fish and seafood (6%) (Cubadda et al., 2020)
Dioxins and PCBs	Dioxins and DL- PCBs	0.6% of adults and 8% of children	Dioxins: cereals and cereal products (50%), meat and meat products (24%), fish (11%), Dairy products (mainly cheese) (7%)

C. The 2014 Total Diet Study in the United Kingdom

The TDS conducted on the population of the United Kingdom in 2014 considered 26 inorganic contaminants and minerals, acrylamide, and 12 mycotoxins that were measured in 28 food groups. The food groups cover many foods that were typical of the UK diet in 2014.

Similar to the French and Italian TDSs, cadmium, lead, and inorganic arsenic showed a level of concern, although not high. However, this was only for infants and young children (FSA, 2014c). In addition, a potential concern for increased lifetime cancer risk was identified for dietary acrylamide exposure for all age groups (FSA, 2014a). As for the mycotoxins, 10/12 did not pose a health risk, and for the remaining two (aflatoxins and citrinin) the risk was inconclusive (FSA, 2014b).

The results for the substances of high concern for the UK population are summarized in Table 4.

Table 4. Substances of high concern for the UK population

Category of substances	Substances	Population concerned	Main contributing foods
Inorganic contaminants	Cadmium	Toddlers and young children (1.5 to 3 years)	Miscellaneous cereals
	Lead	Toddlers and young children (1.5 to 3 years)	Milk
	Inorganic arsenic	Toddlers and young children (1.5 to 3 years)	Miscellaneous cereals
Neo-formed substances	Acrylamide	Adults and children at risk	Potatoes (particularly fried potatoes) and cereals (such as breakfast cereals and sweet biscuits).

To summarize, the French TDS included the largest number of substances examined. However, similar to the French TDS, none of the TDSs under discussion had a focus on specific

populations, nor did they differentiate between types of food production, with the exception of the German TDS.

VI. Focus on University students

1. Food Insecurity Level among University Students

As mentioned in chapter I (3- Economic Disparities and Food Insecurity), one of the vulnerable populations at risk of being food insecure are university students (Abbey et al., 2022; Davitt et al., 2021; DeBate et al., 2021; Nikolaus et al., 2019). This vulnerability can be attributed to limited financial resources, reduced purchasing power, and the rising cost of housing and food (El Zein et al., 2019).The transition from high school to university brings with it new responsibilities and financial burdens, often resulting in limited resources, especially for students who live away from home (Hafiz et al., 2023; Papadaki et al., 2007), have tuition debts (Phillips et al., 2018), or have a family history of financial difficulties (Zigmont et al., 2019).

In Europe, the prevalence of food insecurity among university students has been little studied. In a study conducted among Greek university students, results showed a relatively low proportion of food secure participants (17.8%), with 45.3% experiencing severe food insecurity, 22.0% experiencing moderate food insecurity and the remaining 14.8% experiencing low food insecurity. Notably, students studying in their home city had lower levels of food insecurity than those studying in other cities ($p=0.009$), and there was a clear tendency towards increased food insecurity among students with an unemployed family member ($p=0.05$) (Theodoridis et al., 2018). In comparison to studies conducted in Western societies, where food insecurity ranged from 14.8% to 58.8% (Berg & Raubenheimer, 2015; Bruening et al., 2016; Chaparro et al., 2009; Freudenberg et al., 2009; Gaines et al., 2014; Hughes et al., 2011; Micevski et al., 2013; Morris et al., 2016; Patton-López et al., 2014; Payne-Sturges et al., 2018), the prevalence of food insecurity among Greek students was particularly high. These results may be linked to the severe downturn that happened in Greece, leaving the Greek population with economic challenges (Theodoridis et al., 2018).

In France, nearly 20% of university students are reported to live below the poverty line, according to the General Inspection of Social Affairs (IGAS) (Marie E et al., 2015). This highlights the importance of studying food insecurity in the university student population, in order to understand its prevalence and its impact on the overall well-being of French students.

A pioneering study conducted at the University of Grenoble Alpes in France examined the links between food insecurity status, along with other variables, and the well-being of university students (Isoard-Gautheur et al., 2023). This research was part of a larger initiative called PEANUTS (Précarité Etudiant-es Alimentation Nutrition UniversiTé Santé), which has several objectives, including: describing food insecurity and dietary behaviour, describing the nature of physical activity, analysing information and communication mechanisms in the field of nutrition, and studying information practices on food and health issues among university students (GRESEC, 2021). The results of this study concerning the food insecurity prevalence, are discussed in Chapter 5. This part of our work is included in PEANUTS, as it assessed students' food insecurity and their adherence to the French National Nutrition and Health Programme (PNNS).

Food insecurity has been shown to have a direct impact on the diets of university students, leading to the development of unfavourable eating habits and a significant shift towards degraded, unbalanced behaviours (Chourdakis et al., 2010). Furthermore, these unhealthy eating habits observed during the university years have been associated with increased weight gain, which may persist throughout life (Sogari et al., 2018), causing adverse health outcomes (Djalalinia et al., 2015).

2. Dietary habits among university students

As shown in the literature, the typical diet of university students is generally low in foods that are likely to contribute to optimal eating patterns and are perceived to be healthier, such as vegetables, fruit and dairy products, and high in fats, sugars, salt and alcohol, which are perceived to be less healthy (Althubaiti, 2022; Bernardo et al., 2017; Deliens et al., 2014; Henley et al., 2023; Hilger et al., 2017; Kelly et al., 2013; Ramón-Arbués et al., 2021; Yun et al., 2018), which is unlikely to be conducive to good health (Sprake et al., 2018).

In addition to food insecurity, many other factors can influence the dietary habits of university students. These include the independence to make food choices, engagement with new social groups, cooking skills and facilities, nutritional knowledge and academic stress (Hafiz et al., 2023; Sprake et al., 2018).

Research on the dietary habits of French university students is limited. To our knowledge, only two studies have examined the dietary habits of French university students. The first, conducted in Rouen in 2021, examined university students to assess changes in their

diet quality before and during the COVID-19 period, as well as factors associated with the unfavourable changes, including food insecurity. This study assessed only the consumption of six components: fruit and vegetables, nuts, legumes, cereals, dairy products and fish, using a score based on the PNNS guidelines, the PNNS-GS2, which will be discussed in more detail in the following part (3-A PNNS scores). The results of this study showed a decrease in the PNNS score between the pre- and the COVID-19 pandemic period (mean score pre-COVID 5.0±2.3 versus 4.7±2.3 in the COVID-19 period ($p < 0.0001$)) for 33.1% of the university students (L. Miller et al., 2022), indicating a lower compliance with the PNNS recommendations and thus a deterioration in the dietary quality of the concerned population. It should be noted that the PNNS-GS2 score ranged from 0 to 14. This highlights the fact that **students did not meet the recommendations of the French guidelines even before the COVID-19 pandemic**.

The other study, carried out in 2022, aimed to describe the diets of French students and to identify groups of diets that might differ in terms of nutritional quality and environmental impact.The nutrition quality of the diets was assessed in terms of their compliance with the French dietary standards (sPNNS-GS2 score, ranging from -17 to 11.5) and their environmental impact in terms of greenhouse gas emissions (GHGE). With regard to the results for the nutritional quality aspect, the average sPNNS-GS2 score of the students' diets was -0.8 ± 2.8 overall, i.e. **57% coverage of the French nutritional recommendations**. Furthermore, 3 dietary groups were identified:

- **A healthy diet group with a significantly higher nutritional quality, representing only 20% of the population**
- A Western diet group, characterized by a reported high-energy diet of poor nutritional quality, representing 40% of the population
- A Frugal diet group characterized by a significantly lower energy intake, representing 40% of the population (Arrazat et al., 2023).

These results highlight the importance of conducting more studies on this topic, as they are essential to inform public policies on how to initiate transitions towards healthy and sustainable diets in this population.

3. Evaluation of Nutritional Quality of a Diet

One way of assessing the nutritional quality of the diets of a population is to use an approach known as "a priori", in which diet patterns are defined based on current nutritional knowledge, expressed mainly in terms of food- and nutrition-based dietary guidelines (Verger et al., 2012). Diet quality indices are constructed based on overall adherence or proximity to these dietary patterns. Most existing indices are based on traditional Mediterranean diets or national food-based dietary guidelines (Verger et al., 2012), such as Healthy Eating Index (HEI), developed by United States Department of Agriculture (USDA) to evaluate how well a set of foods corresponds to recommendations of Dietary Guidelines for Americans (Reedy), or PNNS - guidelines score (PNNS-GS), an index previously developed by Chaltiel et al. to reflect compliance with French dietary recommendations (Chaltiel et al., 2019).

A. Programme National Nutrition Santé – guidelines scores

The guidelines of the French national nutrition and health program (PNNS) are a key reference for the promotion of healthy eating habits and the prevention of diet-related diseases (Manger Bouger, 2022). These guidelines provide evidence-based recommendations on various aspects of diet, including consumption of fruit and vegetables, whole grain cereals, dairy, and protein, and avoiding excess sugars, salt and saturated fat.

To date, four PNNSs have been established as dietary guidelines evolve over time in responsiveness to advances in scientific knowledge: PNNS 1 (2001 - 2005), PNNS 2 (2006 - 2010), then PNNS 3 (2011- 2015), and finally PNNS 4 (2019-2023), the current one. Based on these guidelines, two dietary indices were developed to assess guideline compliance and thus population dietary quality.

The first was **the PNNS-Guidelines Score (PNNS-GS),** based on the first PNNS 1 (Estaquio et al., 2009). It included: 8 components that measured the overall compliance with the French portion recommendations (for fruit and vegetables, bread, cereals, potatoes and legumes, milk and dairy products, meat, poultry, seafood and eggs, and non-alcoholic drinks and alcohol consumption), and 4 components that marked the limits of consumption as quantified frequencies (for added fats, added fat ratio, added sugars, and salt). The last component estimated the adherence to physical activity recommendations on a daily basis (Estaquio et al.,

2009). Then, with the new PNNS, the nutritional indices had to evolve in parallel leading to the development of the second PNNS score**, PNNS-GS2**.

In comparison with the 2001 recommendations, the recent recommendations are more extensive, include more specific food groups and, for some groups, have been updated according to an actualization of the scientific literature (Chaltiel, Adjibade, et al., 2019). The main changes to the PNNS-GS2 include:

- An addition of a recommendation for the consumption of organic foods for fruit, vegetables, bread, pulses and cereals as a measure to reduce exposure to contaminants.
- An addition of a refined food specification for bread and cereals
- An addition of two recommendations for moderation in consumption of red meat and processed meat.
- An addition of a weighting factor for each component in accordance with the level of evidence of the association between consumption of food groups and health, in order to avoid over-estimation of multi-item components.

Figure 1 retrieved from Chaltiel et al. is a table detailing the PNNS-GS2: components and scoring (Chaltiel et al., 2019).

Dietary components	Recommendation	Criteria*	Score
Fruits and vegetables (weight = 3)	At least 5 servings/d, with 1 maximum as juice and 1 maximum as dried†	0–3·5	0
		3·5–5	0·5
		5–7·5	1
		≥7·5	2
	Prefer organic fruits	Most of the time	0·5
		Occasionally	0·25
		Never	0
	Prefer organic vegetables	Most of the time	0·5
		Occasionally	0·25
		Never	0
Nuts (weight = 1)	A handful/d†	0	0
		0–0·5	0·5
		0·5–1·5	1
		≥1·5	0
Legumes (weight = 1)	At least 2 servings/week†	0/week	0
		0–2/week	0·5
		≥2/week	1
	Prefer organic legumes	Most of time	0·5
		Occasionally	0·25
		Never	0
Whole-grain food (weight = 2)	Every day†	0	0
		0–1	0·5
		1–2	1
		≥2	1·5
	Prefer organic bread	Most of the time	0·5
		Occasionally	0·25
		Never	0
	Prefer organic grains	Most of the time	0·5
		Occasionally	0·25
		Never	0
Milk and dairy products (weight = 1)	2 servings/d†	0–0·5	0
		0·5–1·5	0·5
		1·5–2·5	1
		≥2·5	0
Red meat (weight = 2)	Limit consumption†	0–500 g/week	0
		500–750 g/week	−1
		≥750 g/week	−2
Processed meat (weight = 3)	Limit consumption†	0–150 g/week	0
		150–300 g/week	−1
		≥300 g/week	−2
	Prefer white ham over other processed meat‡	Ratio < 50 %	0
		Ratio ≥ 50 %	0·5
Fish and seafood (weight = 2)	2 servings/week†	0–1·5 servings/week	0
		1·5–2·5 servings/week	1
		2·5–3·5 servings/week	0·5
		≥3·5 servings/week	0
	Fatty fish 1 serving/week	0–0·5 servings/week	0
		0·5–1·5 servings/week	1
		≥1·5 servings/week	0
Added fat (weight = 2)	Avoid overeating†	>16 % of EIWA	0
		≤16 % of EIWA	1·5
	Prefer ALA-rich and olive oil over other oils	Ratio < 50 %	0
		Ratio ≥ 50 %	1
	Prefer plant fat over animal fat	Ratio > 50 %	0
		Ratio ≤ 50 %	1
Sugary foods (weight = 3)	Limit consumption†	<10 % of EIWA	0
		10–15 % of EIWA	−1
		≥15 % of EIWA	−2
Sweet-tasting beverages§ (weight = 3)	Limit consumption†	0 ml/d	0
		0–250 ml/d	−0·5
		250–750 ml/d	−1
		≥ 750 ml/d	−2
Alcoholic beverages (weight = 3)	Limit consumption†	0 g/week	0·5
		0–100 g/week	0
		100–200 g/week	−1
		>200 g/week	−2

(*Continued*)

Dietary components	Recommendation	Criteria*	Score
Salt (weight = 3)	Limit consumption†	≤6 g/d	1
		6–8 g/d	0
		8–10 g/d	−0·5
		10–12 g/d	−1
		>12 g/d	−2

EIWA, energy intake without alcohol; ALA, α-linolenic acid.
* Servings per d unless otherwise stated.
† Principal recommendations.
‡ Conditional: the 0·5 bonus point only occurs if total processed meat consumption is more than 150 g/week.
§ Sweet-tasting beverages are specifically sugary beverages, artificially sweetened beverages and fruit juices.

Figure 1. Programme National Nutrition Santé – guidelines score 2 (PNNS-GS2): components and scoring retrieved from Chaltiel et al., 2019

The PNNS-GS2 has been used in different studies to evaluate the nutritional quality of the diet for the general population (Chaltiel et al., 2019; Marty et al., 2022), as well as for specific populations such as university students (Arrazat et al., 2023; L. Miller et al., 2022).

B. Probability of Adequate Nutrient Intake

The Probability of Adequate Nutrient Intake (PANDiet) is an index of dietary quality based on the intake of nutrients, using a probabilistic approach to estimate the adequacy of intake for 24 nutrients, including : proteins, total carbohydrates, fibres, total fat, saturated and polyunsaturated fatty acids, cholesterol, thiamin, riboflavin, niacin, folate, vitamins A, B-6, B-12, C, D and E, calcium, magnesium, zinc, phosphorus, potassium, iron and sodium (Verger et al., 2012).

The advantage of the PANDiet over food-based dietary guideline-based indices is that it can be adapted (applied to populations with different dietary habits) and it is supported by evidence on nutrient intakes, including recommended dietary intakes and lower and upper dietary limits (Verger et al., 2012).

The PANDiet dietary index can be used alone in a study to assess the dietary quality of a population (de Gavelle et al., 2016; Verger et al., 2012) as it can be used together with another food-based dietary guideline for better and more sensitive analyses (Arrazat et al., 2023; Assmann et al., 2016; Berthy et al., 2023).

Chapter 2 - Project Context and Objectives

As mentioned in the previous chapter, the main challenges related to nutrition include ensuring an optimal diet in terms of both quantity and quality, ensuring a good environmental impact of the food system, and ensuring food safety for consumers, including exposure to chemicals, especially since food is a major source of exposure to environmental pollutants for the general population.

Consequently, this thesis embarks on a comprehensive analysis to address these challenges on the following levels:

To meet environmental challenges, new methods of agricultural production are being developed, including organic farming. This method of production is strongly encouraged due its sustainable impact on the environment (Reganold & Wachter, 2016; Shubha et al., 2021), as well as many studies have shown a higher nutritional value for organic produced food compared to conventional ones (Zheng et al., 2019). Moreover, the French National Nutrition and health program (PNNS) recommends 20% of total consumptions of fruits and vegetables, cereals, and legumes from organic productions (Ministère des solidarités et de la santé, 2019). However, this expansion raises a number of questions. It has been shown in the literature that, despite the ban on the use of chemically synthesized molecules, which reduces exposure to certain pesticides, there are specific problems associated with this method of production (e.g., (Dervilly-Pinel et al., 2017)). The risk assessments carried out to date do not take these issues into account. Additionally, to date no comprehensive databases has been found in the literature for chemical substances in organic food. Hence the first objective of this thesis was **(1) to develop and implement a methodology for assessing the impact of organic food consumption versus conventional food on the consumer's exposure to environmental pollutants in Europe.**

To meet the challenge of ensuring an optimal diet for all, a specific population vulnerable to food insecurity (university students) has been included in this thesis, to evaluate their dietary habits. Hence, the second objective of this thesis was **(2) to assess the nutritional quality of the young adult population via a PNNS score and to study the impact of food insecurity on the adherence national nutritional recommendations**.

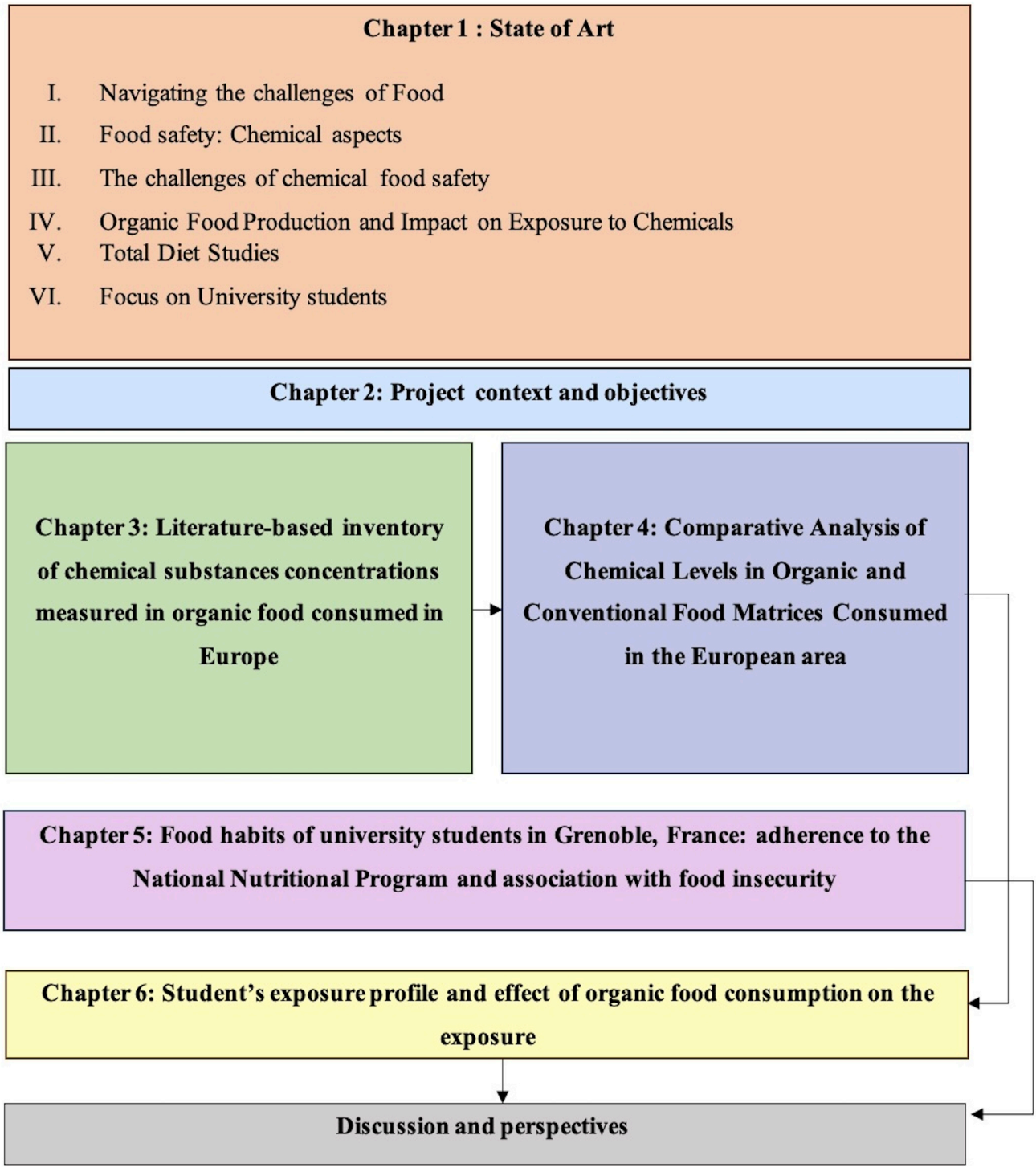
To achieve these goals, the first step was to create an exhaustive database collecting data from the literature on contamination levels in organic agriculture in Europe. Therefore, a comprehensive search was carried out in 4 different databases (Pubmed, WoS, Embase and Agricola) and EFSA, and the results were subjected to a selection process and data were retrieved and described **(Chapter 3)**.

Subsequently, as the data from the database presented limitations (quantitative and units limitations), disabling the direct calculation of exposure levels from the available data, an Excess Ratio (ER) approach was developed, enabling first the evaluation of chemical content in between the different agriculture foodstuff, while overcoming the database's limitations, and serving as a base for the methodology of assessing the impact of organic food consumption on the level of exposure to chemicals compared to conventional food **(Chapter 4)**.

Furthermore, in the context of a study on food insecurity among students at the University of Grenoble Alpes (PEANUTS), this population was selected to assess their dietary habits using a score that was developed to measure students' adherence to the PNNS, thus assessing the meeting on an optimal diet. Subsequently, the association between food insecurity and compliance with the PNNs was studied in order to better understand the impact of FI on the quality of students' diets **(Chapter 5)**.

Finally, the exposure values of the students were calculated using the EAT2 contamination database for selected substances and were compared to that of the general population from the EAT2 study. Moreover, the developed methodology was applied based on the calculation of a percentage relative exposure illustrating the impact of increased organic food consumption on exposures specific to a couple: substance, matrix **(Chapter 6)**.

The following manuscript reading guide is a reminder of the manuscript's outline.


Chapter 1 : State of Art
I. Navigating the challenges of Food
II. Food safety: Chemical aspects
III. The challenges of chemical food safety
IV. Organic Food Production and Impact on Exposure to Chemicals
V. Total Diet Studies
VI. Focus on University students
Chapter 2: Project context and objectives
Chapter 3: Literature-based inventory of chemical substances concentrations measured in organic food consumed in Europe
Chapter 4: Comparative Analysis of Chemical Levels in Organic and Conventional Food Matrices Consumed in the European area
Chapter 5: Food habits of university students in Grenoble, France: adherence to the National Nutritional Program and association with food insecurity
Chapter 6: Student's exposure profile and effect of organic food consumption on the exposure
Discussion and perspectives

Chapter 3 - Literature-based inventory of chemical substances concentrations measured in organic food consumed in Europe

In 2011, the European Food Safety Authority (EFSA), the Food and Agriculture Organization of the United Nations (FAO), and the World Health Organization (WHO) issued joint guidance on a harmonized approach known as the 'Total Diet Study' (TDS). This approach involves analyzing chemicals in foods and combining the data with dietary consumption information to characterize exposure profiles of studied populations (EFSA, FAO, and WHO, 2011).

In France, the French Agency for Food, Environmental and Occupational Health and Safety (ANSES) has conducted three Total Diet (EAT) studies on the French population in 2005, 2011 and 2016, including one specifically focused on children (EATi). **However, these studies do not differentiate contamination levels based on the production method of foods.** Notably, Europe has seen a significant increase in organic agricultural area, reaching 14.7 million hectares in 2020 and increasing to 15.9 million hectares in 2021, representing 9.9% of the total agricultural area. With 2.8 million hectares in 2021, France has the largest organic production area in the EU (Eurostat, 2023).

Currently, **the assessment of exposure through the consumption of organic products is limited via the Total Diet Study method, mainly due to the lack of European databases containing contamination levels for organic foods**.

This Chapter’s aim was to construct an exhaustive database on chemical and their levels in OF consumed in Europe. The chapter provides a comprehensive description of the database reflecting what was found in the literature to this concern.

Literature-based inventory of chemical substances concentrations measured in organic food consumed in Europe

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Abstract:

Populations are exposed daily to numerous environmental pollutants, particularly through food. Because of environmental issues, many agricultural production methods have been developed, including organic farming. To date, there is no exhaustive inventory of the contamination of organic food (OF) as it exists for conventional food. The main objective of this work was to construct a database on chemical substances and their levels in OF consumed in Europe. To this end, a literature search was conducted resulting in a total of 1207 concentration values from 823 food–substances couples involving 166 food matrices and 209 chemical substances among which 95% were not authorised in organic farming and 80% were pesticides. The most encountered substance groups are “inorganic contaminants” and “organophosphate” and the most studied food groups are “fruit used as fruit” and “Cereals and cereal primary derivatives”. Additional studies are necessary to evolve towards an exhaustive database on OF contaminations.

Keywords: organic food, contaminants, residues, metals, persistent organic pollutants, pesticides

I. Introduction

Today, following the evolution of the concept, exposome is defined as the cumulative measure of environmental influences (positive or negative) and associated biological responses that living organisms undergo from conception and throughout life, and which can influence their state of health. This includes environmental exposure to chemical, biological and physical agents, but also lifestyle, socio-economic and cultural determinants and psycho-social context (ANSES 2023; Miller & Jones, 2014; Rappaport, 2011; Vermeulen, 2020; Wild, 2005).

Because of the contamination of the food chain by a multitude of environmental pollutants (Abraham et al., 2018), food is a major source of exposure on humans and therefore a significant part of its chemical exposome. On the other hand, in order to meet the food needs of the world's ever-growing population while simultaneously responding to the challenges of sustainable development, several agricultural production methods have been developed and coexist, such as for example organic, biodynamic or sustainable farming (Reganold & Wachter, 2016; Turinek et al., 2009; Velten et al., 2015).

As far as organic farming is concerned, the European Union (EU) has seen a steady increase, with 14.7 million hectares in 2020 up to 15.9 million hectares in 2021, i.e., 9.9% of the total agricultural area, with France holding the largest organic production area in the EU, with 2.8 million hectares in 2021 (Eurostat, 2023).

Organic farming complies with strict regulations. In the EU, organic farming and production rules are enforced by Regulations (EC) N°848/2018 (European Parliament And Council, 2018). This regulation includes in particular a ban on the use of chemically synthesized plant protection products, the use of specific natural fertilizers and natural pest control methods, the exclusive use of organically-farmed seeds while avoiding GMOs, the recycling of organic matter, the use of crop rotation, a preference for plant varieties adapted to local conditions, particular attention to animal welfare and the limited use of medicinal products for therapeutic purposes ((EC) N°848/2018). Additionally, these regulations specify extensive controls on labelling, and provide a list of natural substances authorized in organic farming such as azadirachtin derived from *the Azadirachta indica* (neem trees seeds), Spinosad produced by *Saccharopolyspora spinosa* (soil bacterium), copper, and pyrethroids derived from *Chrysanthemum indicum* (Chrysanthemum) ((EC)N°2021/1165).

Moreover, despite all the regulations, there are also ubiquitous environmental contaminations over which we have no particular control in biological products, including heavy metals (Debnath et al., 2015; Malmauret et al., 2002), Persistent Organic Pollutants (POP) such as dioxins, PolyChloroBiphenyls (PCBs) (Almeida-González et al., 2012; Dervilly-Pinel et al., 2017; Witczak & Abdel-Gawad, 2012), inorganic components as cadmium, lead, arsenic (Hoefkens et al., 2009; Malmauret et al., 2002) and brominated compounds (Dervilly-Pinel et al., 2017), but also pesticides authorized in conventional agriculture that can circulate in the biosphere and contaminate organic products (Mao et al., 2021; Romero-González et al., 2011). The results obtained are quite variable and depend on the chemical substance studied but also on the food matrices. For example, in Almeida-González et al, medians for the sum of organochlorine (OC) pesticides were found to be three times lower in organic cheese (14.44 ng/g fat vs. 42.73 ng/g fat in conventional cheese, with p=0.001), while PCBs were found to be 2.4 times higher in the organic batch (22.55 ng/g fat vs. 9.57 ng/g fat in the conventional batch), but with p=0.074 (Almeida-González et al., 2012). Significantly higher levels of dioxins and PCBs were also found in organic pork and chicken, up to 3 times higher for the 6 non-dioxin like PCBs in pork (Dervilly-Pinel et al., 2017). Concerning cadmium, some studies show significantly higher levels in certain vegetables compared to conventional (up to 2 times higher in spinach and carrots), while they are around 1.2 times lower in other matrices (potatoes, lettuce) (Hoefkens et al., 2009; Malmauret et al., 2002). With regard to levels of impregnation, a study of 13 adults showed that switching to a diet consisting of 80% organic food for one week reduced their exposure to organophosphate pesticides. Indeed, urinary levels of total dialkylphosphate metabolites were significantly reduced by 89% compared to others on a diet consisting solely of conventionally farmed foods (mean=0.032 µg/g creatinine [SD=0.038] and 0.294 µg/g creatinine [SD=0.435] respectively, p=0.013) (Oates et al., 2014). Other similar studies carried out on groups of primary school-age children have also shown that median urinary concentrations of metabolites of certain pesticides were significantly reduced immediately after the introduction of organic food (Curl et al., 2003; Lu et al., 2006, 2008).

In 2011, the European Food Safety Authority (EFSA), the Food and Agriculture Organization of the United Nations (FAO) and the World Health Organization (WHO) jointly published a guidance document on a harmonized approach to the assessment of dietary exposure to chemicals, known as the Total Diet Study (TDS) (EFSA et al., 2011b). This methodology is based on the analysis of chemicals substances present in food combined with data on food consumption and therefore requires data on food contamination. However, the

databases available in the literature or from health agencies, although very extensive in terms of the number of substances studied in conventionally farmed foods, contain little or no data on the contamination of organically farmed foods. This scarcity is evident in the latest EFSA report, in which organic foods represent only 6.5% of all the samples tested (EFSA, 2022). At national level, the database of the Chemical and Veterinary Investigation Offices (CVUA) in Stuttgart, Germany, provides concentration values for pesticides in organic food (CVUA Stuttgart, 2019), which were included in the EFSA report. In France, as part of its studies on total diet, the French Agency for Food, Environmental and Occupational Health and Safety (ANSES) offers a database on the contamination of foods as consumed by the population. It covers 455 chemical substances (pesticides, dioxins, PCBs, heavy metals, etc.) and 212 food matrices, but does not present any specific data to organically produced foods (ANSES, 2011a).

The aim of this study was to compile a database containing all the data available in the literature on the contamination of organically produced foodstuffs in the EU region. To this end, an exhaustive literature search was carried out from June 10 to June 15, 2021. The steps involved in the search and selection of articles are described in detail in the sections on materials and methods. The results were presented in two parts: one describing the database qualitatively in terms of content (matrices, chemical substances, matrix – chemical couples), and one describing the distribution of data collected. Although with certain limitations that will be discussed in the article, the obtained database provides an exhaustive overview of all the contamination measurements that have been carried out in Europe on organically-farmed foods.

II. Methods

Literature search and identification of articles of interest

Literature search was conducted from June 10 to June 15, 2021, in four electronic databases: PubMed, Web of Science (WoS), Embase and Agricola. The phrase “contaminants and residues in organic food” was used for the search in all databases except for PubMed where the following MeSH (Medical Subject Headings) was used: pesticide* OR contaminant* OR pollutant* OR chemical* OR herbicide* OR fertilizer* OR fungicide* OR insecticides OR metals* OR antibiotics*) AND (residues OR contamination* OR exposure*) AND organic* AND (food* OR product*). All articles published from January 1, 2000 until June 15, 2021 in PubMed were considered whereas all articles until the end of the search period (June 15, 2021) were considered in the other databases. Furthermore, in order to facilitate the sorting step,

resulting articles were merged into Zotero v5.0.96.3 (Zotero.org) and exported to an Excel sheet.

In addition to the literature search, EFSA reports were targeted. Indeed, EFSA is the major source of data and regulations related to food in the EU Region. Only reports on pesticide residues in food from 2008 until to 2021 considered chemicals in organically farmed food (OF) and hence were included in the study.

Study selection and eligibility criteria

The articles were selected in four steps using criteria based on the language, title, abstract and data availability (see Table A3-1 in Supplemental Material). Briefly, the selected studies had to be written either in English or in French and had to deal with OF and any chemicals substances they might contain. Finally, the article had to contain concentration values corresponding to OF from EU markets.

All the articles that did not match the scores were excluded; the remainder were scanned for concentration values. Articles that did not have any data were examined to see if they contained a supplemental material for concentration records, otherwise they were excluded. Furthermore, articles with concentration values that did not correspond to food matrices from the EU market were also excluded, as our aim is to create an inventory on chemical substances in organic food consumed in the EU region.

Data extraction

The concentration values (mean ± SD and/or distribution as appropriate) were grouped with their corresponding food matrix/chemical substance and an excel table was created. The number of matrices tested, the number of positive samples, the limits of quantification (LOQ), the limits of detection (LOD), the minimum and the maximum values were added to the table whenever they were specified in the studies. Furthermore, additional columns were added, including the country of origin, the continent of origin, the foodEx2 group from level 2 nomenclature to which belongs the matrix (FX-L2) (detailed in appendix A3 Table A3-3), the code of the foodEx2 group (code_FX), the food matrices, the chemical substances, the substance groups, the general groups, the complementary information, the couple frequency, the LOD/LOQ units, the measurement types, the chemical types, and the references (elements detailed in Appendix A3 Table A3-2).

Food matrices and chemical substances classifications

To facilitate the analysis of the database, the OF matrices and the chemical substances were aggregated into broader groups, allowing a description on a wider scale of what has been more represented in the database table. For each food matrix, a group nomenclature and a code were assigned following the food classification and description system FoodEx2 L2 (European Food Safety Authority, 2015). For the chemical substances, substance groups (the chemical family group of the substance), general groups (the largest category to which belong this chemical substance) and complementary information were assigned for each substance. For the pesticides, the classifications were based on the Pesticide Properties DataBase (PPDB) (University of Hertfordshire, 2020).

The Scientific Interest

The scientific interest (I) indicates the significance of an item (substance, matrix, or matrix-substance couple) based on the frequency or number of values in the database. Items were then classified as high I when twenty or more concentration values are reported, medium I when the number of concentration values lies between ten (included) and twenty (exclude), and low I for the number of concentration values less than ten. This classification was inspired from an article done by Rieutort et al. (Rieutort et al., 2016).

Data Processing and Analysis

The database was implemented using an Excel spreadsheet. All statistical processing and analyses were done using Excel and R software.

III. Results

Identification of concentration values of chemical substances in organic food

The literature search resulted in a total of 15 455 articles out of which 22 articles were considered after the exclusion of duplicate reports and those not meeting the selection criteria (Table A3-1) based on language, title, abstract, the database availability and the region of consumption. Furthermore, 10 articles withdrawn from EFSA were added, including databases on concentration values of chemical substances in OF, resulting in 32 articles in total (Fig.1). These studies resulted in a database table of 1207 concentration values for chemical substances in OF consumed in EU. The database table “Pollutants in Organic Food consumed in Europe (POFE)” can be found at (Choueiri et al., 2023, or see attached Excel file).

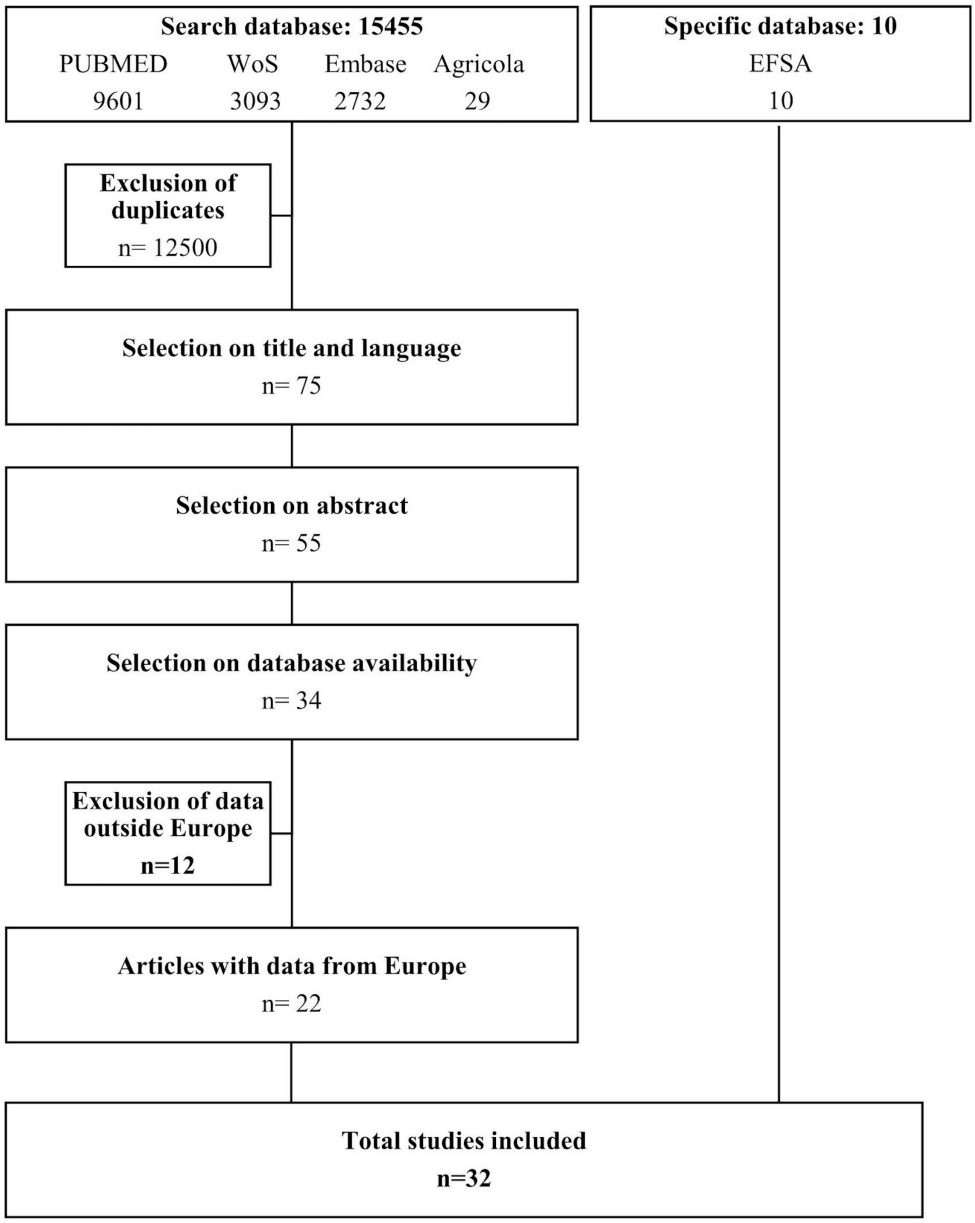


Figure 2. Selection of studies with concentration values for chemical substances in organic food matrices consumed in Europe

1. Descriptive analysis of POFE

Description of the database table

A total of 166 matrices and 209 chemical substances were found, resulting in 823 couples (matrix, substance). All food matrices were retrieved from the EU market, but they originated from 63 different EU and non-EU countries worldwide. The food matrices were grouped into 34 different FoodEx 2 level 2 categories. The chemical substances, also grouped into larger categories, resulted in 74 substance groups and 6 general groups (pesticides, inorganic contaminants, minerals, dioxins, PCBs, and brominated flame retardants). Concerning the data sources, 58% of concentration values were from EFSA and 48% from the rest of the data sources (non-EFSA). Figure 3 shows the summary of the overall distribution of the parameters in the database table.

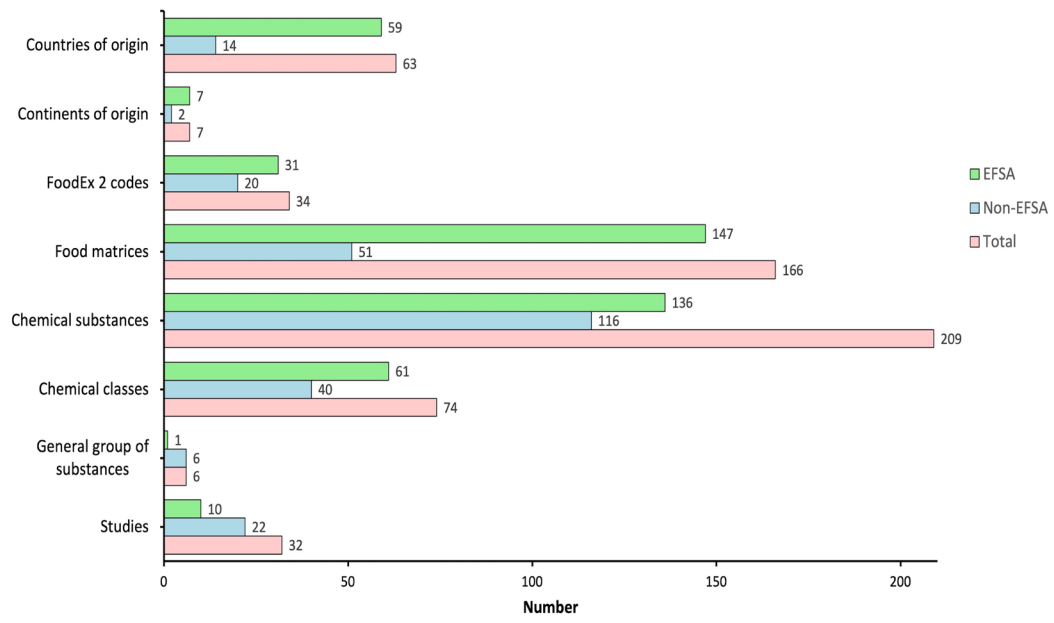


Figure 3. Distribution of the variables in POFE. Quoted numbers to the histogram bars correspond to the total number of distinct variables. The non-EFSA columns represent all the data of studies from PUBMED, WoS, Embase, and Agricola

Description of food matrices

The number of concentration values varied among the OF matrices: 132 food matrices, i.e., 79,5% of the OFs have a low I (number of values in the database less than 10) while 13 and 21 food matrices have high (more than 20 values in the database) and medium (between 10 and 20 values in the database) I, respectively (Figure 4A). The OFs "Baby foods other than processed cereal-based foods" and "Tea (green, black)" have the highest I with number of values of 80 (from 6 studies) and 76 (from 8 studies), respectively (Figure 4B). However, several OFs are less represented in terms of number of values, but with more studies. For example, the OFs "wheat" and "tomato" have respectively 53 and 40 values but each comes from 12 studies.

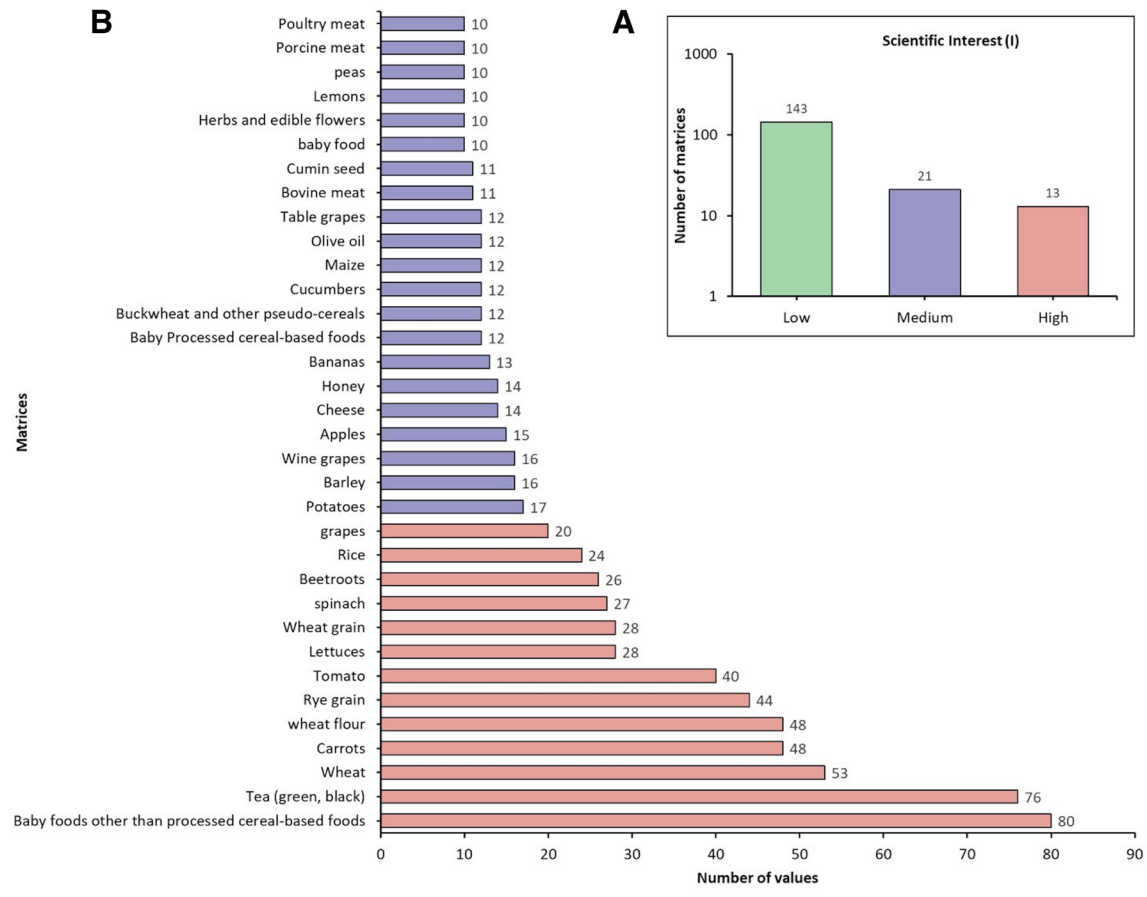


Figure 4. (A) Distribution of organic food matrices in low, medium, and high scientific interest (I). Low (green), medium (purple) and high (pink) I correspond to matrices with number of values < 10, ≥ 10 and < 20, and ≥ 20, respectively. (B) Matrices ranked by number of values (number of studies) for matrices with medium (purple) and high (pink) I

The OFs also varied in terms of association to chemical substances: while 48% of OFs are related to a single chemical, the remaining involved multiple chemicals. Figure 5 shows the 18 OFs with contamination values for 10 or more different chemicals substances. For example, "rye grain" has contamination values for 38 different chemicals substances (Figure 5), while "poultry meat" has contamination values for 10 different chemicals substances (Figure 5) whereas "pear" has values only for one chemical substance (not shown in Figure 5).

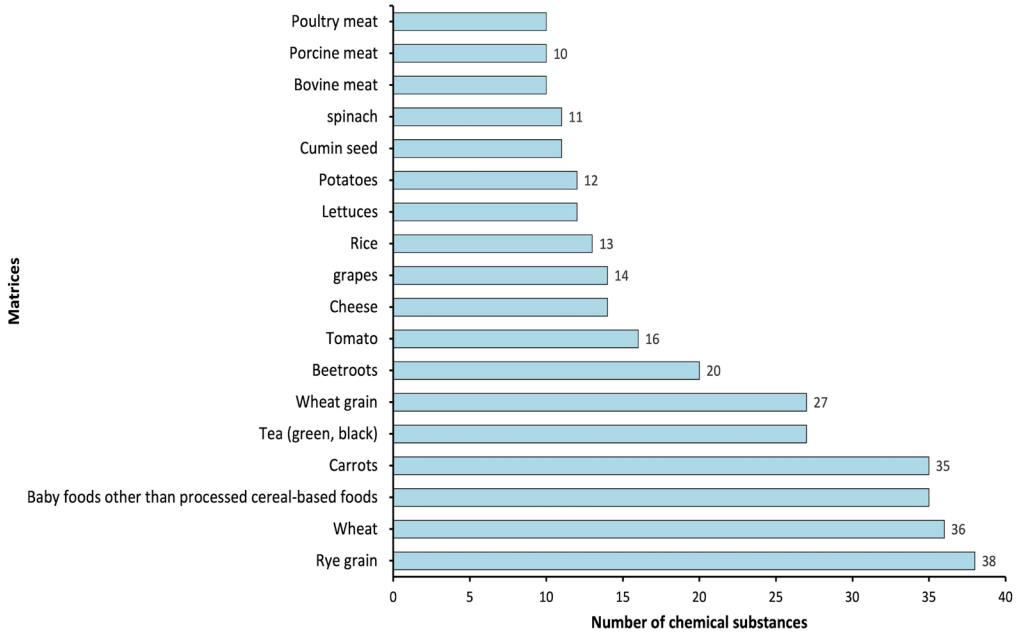


Figure 5. Organic food matrices with 10 or more quantified distinct chemical substances

Description of chemical substances

The current POFE database includes in total 209 different chemical substances. The number of values for each substance is distributed as follows: 182 (86%) chemical substances have a low I, 19 (9%) a medium I and 10 (4.8%) with high I (Figure 6A). The highest I is observed for “Chlorate” and “Fosetyl-Al” with 117 and 76 values, respectively, from 4 and 6 studies, respectively. Other substances with medium I like "Imidacloprid", "Acetamiprid" and "boscalid" (with 19, 17 and 13 values, respectively) are considered in larger number of studies, 11 for each of them (Figure 6B).

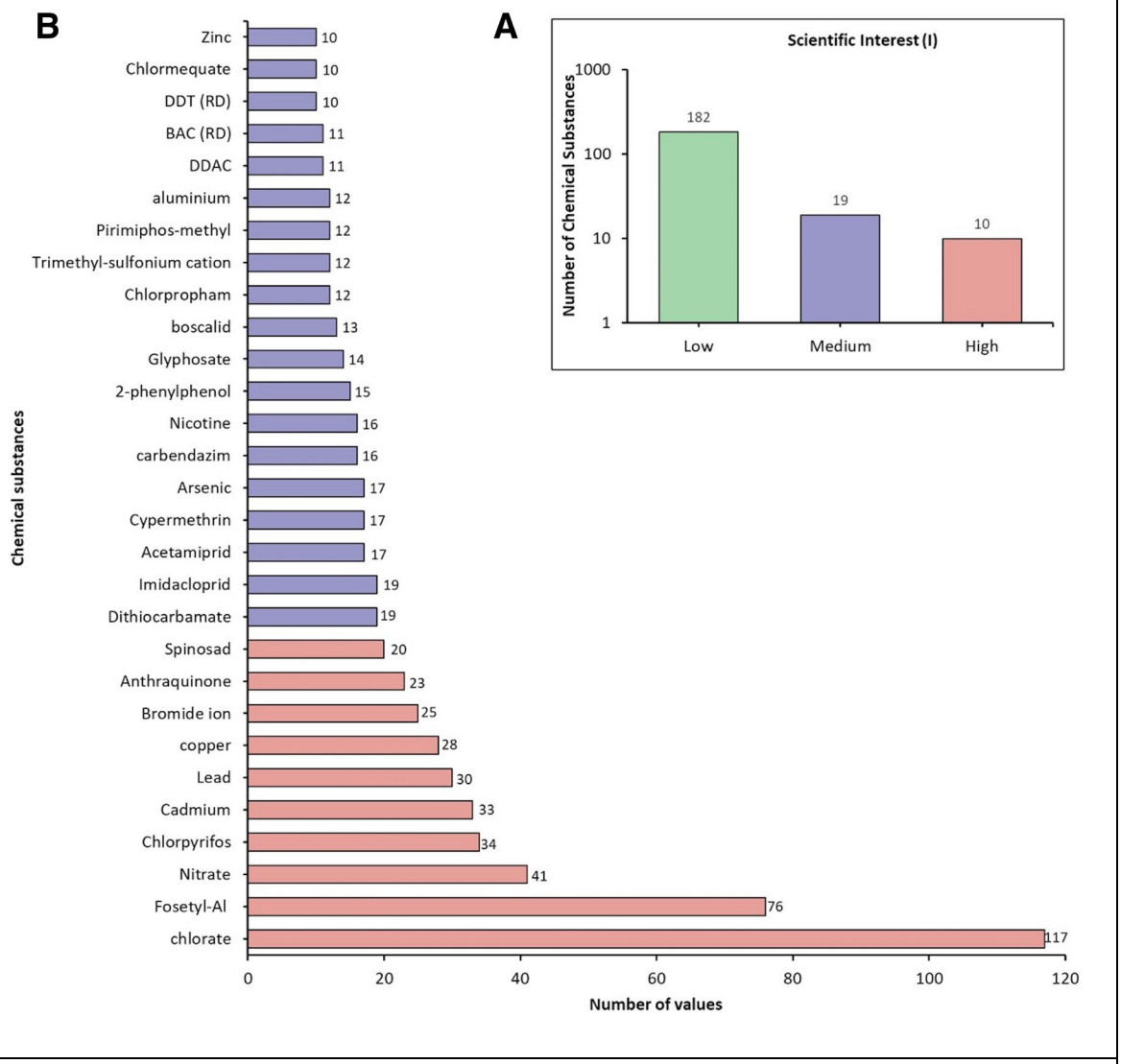


Figure 6. (A) Distribution of chemical substances in low, medium, and high scientific interest (I). Low (green), medium (purple) and high (pink) I correspond to chemical substances with number of values < 10, ≥ 10 and < 20, and ≥ 20, respectively. (B) Chemical substances ranked by number of values (number of studies) for substances with medium (purple) and high (pink) I.

On the other hand, the number of OFs analyzed per chemical substance varies greatly. 58% of the substances studied were analyzed in several OFs. Figure 7 identifies the chemicals substances detected in at least 10 different OF matrices. Chlorate is the chemical substance that was analyzed in the most different OFs with 52 different OFs analyzed (Figure 7).

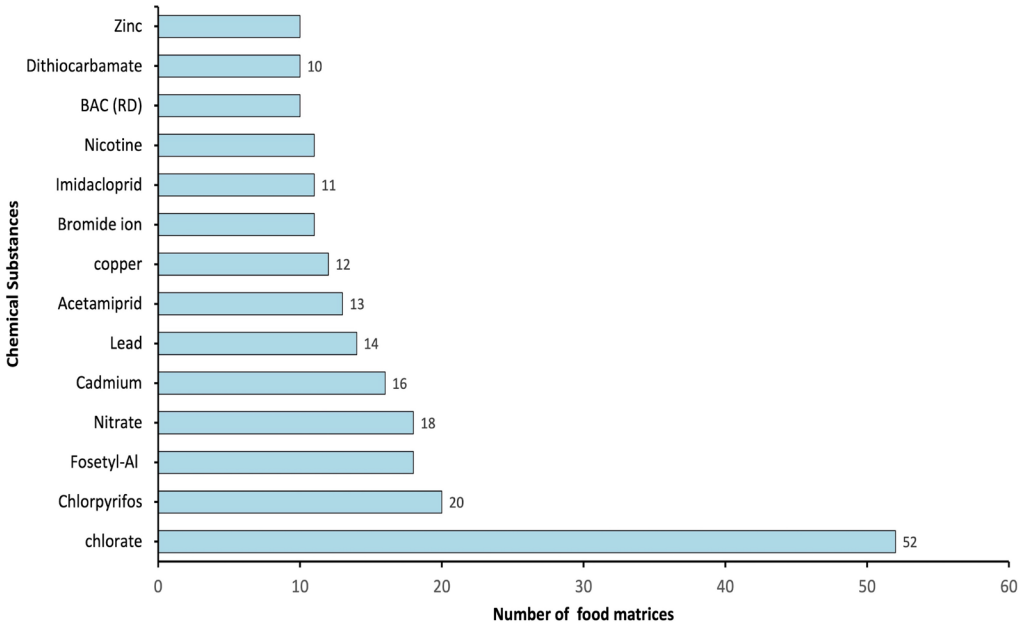


Figure 7. Chemical substances detected in at least 10 distinct organic food matrices

Moreover, more variation was perceived at the "general groups" level, where the majority of the chemical substances were pesticides (80%), while the rest were inorganic contaminants (9%), minerals (5%), PCBs (2%), dioxins (4%) and brominated flame-retardants (0,25%). Also, only 5% of the detected pesticides corresponded to authorized substances in OF and included spinosad, azadirachtin, pyrethrins and copper. They were referred to as residues, while the rest were labeled as contaminants.

Description of food matrix - chemical substance couples

While the database consists of 823 different couples, only 8 couples have more than 10 values (medium I) (Figure 8A) and only a single couple "Baby food other than processed cereal-based foods, Fosetyl-Al" is ranked as high I with 66 values from 6 different studies (Figure 8B). The couples "Baby foods other than processed cereal-based foods, Fosetyl-Al" and "Tea (green, black), Anthraquinone" were the most studied in terms of number of articles (6 articles for each). 88.4% (783/823) of couples were from a single study.

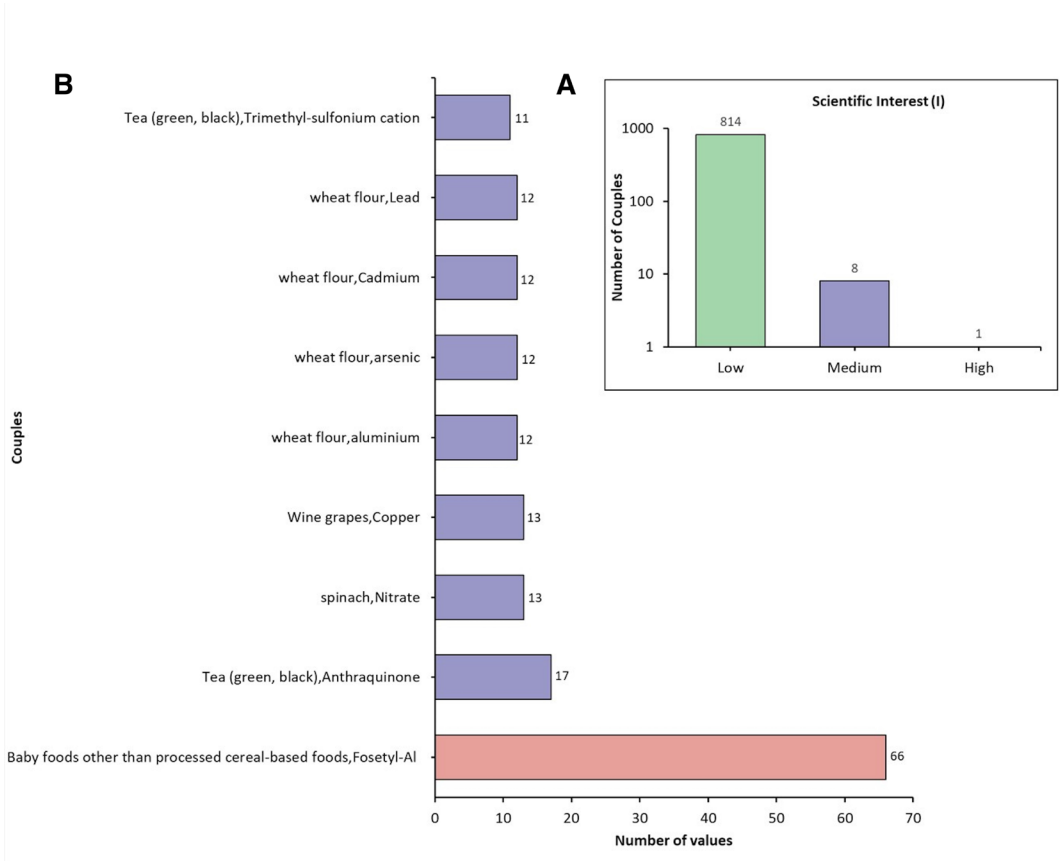


Figure 8. (A) Distribution of food matrix - chemical substance couples in low (green), medium (purple), and high (pink) scientific interest (I). Low, medium and high I correspond to chemical substances with number of values < 10, ≥ 10 and < 20, and ≥ 20, respectively. (B) Couples ranked by number of values (number of studies) with medium (purple) and high (pink) I

Description of the POFE database according to FoodEx groups

To obtain groups of OF matrices with a substantial number of values and subsequently allow for statistical analysis, the 166 matrices were grouped according the FoodEx2 group nomenclature, thus resulting in 34 FoodEx2 groups. Table A3-3 in the Supplementary Materials lists the food matrices in FoodEx2 groups. The distribution in terms of scientific interest I was as follows: 12, 10 and 12 FoodEx groups were classified as low, medium and high, respectively (data not shown). The groups with most data (high I) include “Fruit used as fruit”, “Ready-to-eat meal for infants and young children”, “Cereals and cereal primary derivatives”, “Herbs and edible flowers”, “Legumes”, “Root and tuber vegetables (excluding starchy- and sugar-)”, “Mammals and birds meat”, “Nuts, oilseeds and oilfruits”, “Leafy vegetables”, “Coffee, cocoa, tea and herbal ingredients”, “Fruiting vegetables”, and “Spices”,

Figure 9 represents the distribution of the FoodEx groups according to the total number of chemicals substances studied in these groups. The three groups with the highest number of chemical substances were "Cereals and Primary Cereal Derivatives" with 83 substances (78 contaminants and 5 residues) followed by "Fruit used as fruit" with 65 substances (63 contaminants and 2 residues) and "Root and tuber vegetables (excluding starch and sugar)" with 42 substances (42 contaminants and 0 residue) (Figure 9). The groups "infant and follow-on formulae", "savoury extracts and sauce ingredients" and "sprouts, shoots and similar" comprised only 1 chemical substance, i.e., involving 0.4% of all chemical substances in the database (Figure 9).

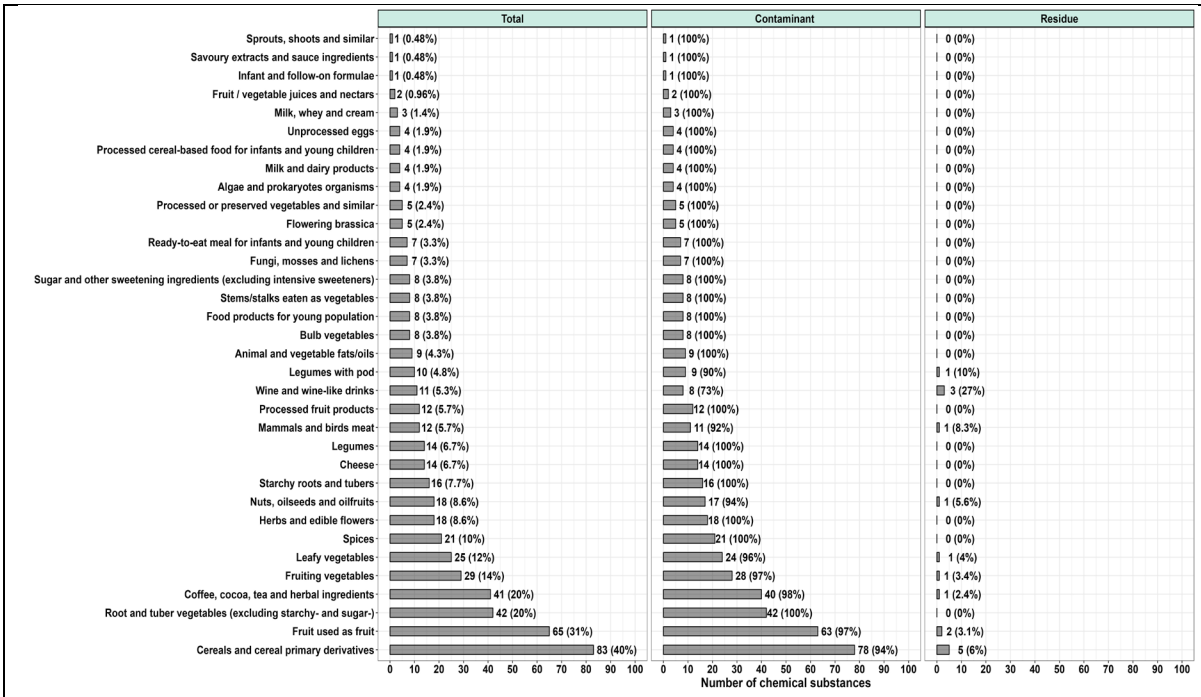
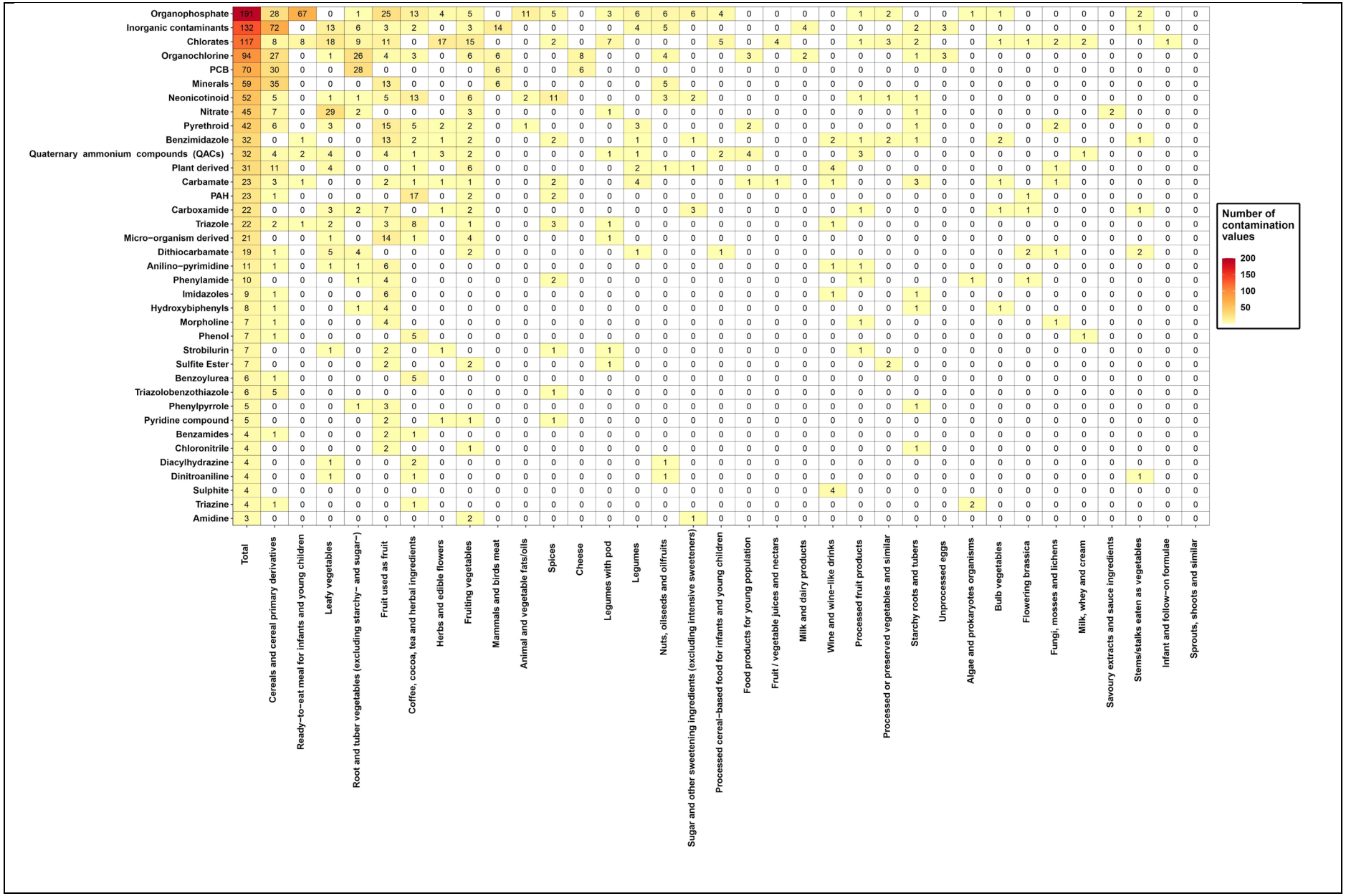


Figure 9. FoodEx groups ranked by number of chemical substances. “Total” stands for the total number of chemical substances (% of the total chemical substances), “Contaminants” for the number of contaminants, i.e., not allowed in organic agriculture (% of the total contaminants), and “Residues” for the number of residues, i.e., allowed in organic agriculture (% of the total residues)

Substance group versus FoodEx group

Likewise, chemical substances were classified according to a generic group “substance groups” as described in Table A3-4 in the Supplemental Material. The seventy-four substance groups obtained are shown in Figure 10.


Organophosphate 191 28 67 0 1 25 13 4 5 0 11 5 0 3 6 6 6 4 0 0 0 0 1 2 0 0 1 1 0 0 0 0 2 0 0
Inorganic contaminants 132 72 0 13 6 3 2 0 3 14 0 0 0 0 4 5 0 0 0 0 4 0 0 0 2 3 0 0 0 0 0 0 1 0 0
Chlorates 117 8 8 18 9 11 0 17 15 0 0 2 0 7 0 0 0 5 0 4 0 0 1 3 2 0 0 1 1 2 2 0 0 1 0
Organochlorine 94 27 0 1 26 4 3 0 6 6 0 0 8 0 0 4 0 0 3 0 2 0 0 0 1 3 0 0 0 0 0 0 0 0 0
PCB 70 30 0 0 28 0 0 0 0 6 0 0 6 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Minerals 59 35 0 0 0 13 0 0 0 6 0 0 0 0 0 5 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Neonicotinoid 52 5 0 1 1 5 13 0 6 0 2 11 0 0 0 3 2 0 0 0 0 0 1 1 1 0 0 0 0 0 0 0 0 0 0
Nitrate 45 7 0 29 2 0 0 0 3 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 2 0 0 0
Pyrethroid 42 6 0 3 0 15 5 2 2 0 1 0 0 0 3 0 0 0 2 0 0 0 0 0 1 0 0 0 0 2 0 0 0 0 0
Benzimidazole 32 0 1 0 0 13 2 1 2 0 0 2 0 0 1 0 1 0 0 0 0 2 1 2 1 0 0 2 0 0 0 0 1 0 0
Quaternary ammonium compounds (QACs) 32 4 2 4 0 4 1 3 2 0 0 0 0 1 1 0 0 2 4 0 0 0 3 0 0 0 0 0 0 0 1 0 0 0 0
Plant derived 31 11 0 4 0 0 1 0 6 0 0 0 0 0 2 1 1 0 0 0 0 4 0 0 0 0 0 0 0 1 0 0 0 0 0
Carbamate 23 3 1 0 0 2 1 1 1 0 0 2 0 0 4 0 0 0 1 1 0 1 0 0 3 0 0 1 0 1 0 0 0 0 0
PAH 23 1 0 0 0 0 17 0 2 0 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0
Carboxamide 22 0 0 3 2 7 0 1 2 0 0 0 0 0 0 0 3 0 0 0 0 0 1 0 0 0 0 1 1 0 0 0 1 0 0
Triazole 22 2 1 2 0 3 8 0 1 0 0 3 0 1 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0
Micro−organism derived 21 0 0 1 0 14 1 0 4 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Dithiocarbamate 19 1 0 5 4 0 0 0 2 0 0 0 0 0 1 0 0 1 0 0 0 0 0 0 0 0 0 0 2 1 0 0 2 0 0
Anilino−pyrimidine 11 1 0 1 1 6 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 1 0 0 0 0 0 0 0 0 0 0 0 0
Phenylamide 10 0 0 0 1 4 0 0 0 0 0 2 0 0 0 0 0 0 0 0 0 0 1 0 0 0 1 0 1 0 0 0 0 0 0
Imidazoles 9 1 0 0 0 6 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 1 0 0 0 0 0 0 0 0 0 0
Hydroxybiphenyls 8 1 0 0 1 4 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 1 0 0 0 0 0 0 0
Morpholine 7 1 0 0 0 4 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 1 0 0 0 0 0
Phenol 7 1 0 0 0 0 5 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0
Strobilurin 7 0 0 1 0 2 0 1 0 0 0 1 0 1 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0
Sulfite Ester 7 0 0 0 0 2 0 0 2 0 0 0 0 1 0 0 0 0 0 0 0 0 0 2 0 0 0 0 0 0 0 0 0 0 0
Benzoylurea 6 1 0 0 0 0 5 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Triazolobenzothiazole 6 5 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Phenylpyrrole 5 0 0 0 1 3 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0
Pyridine compound 5 0 0 0 0 2 0 1 1 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Benzamides 4 1 0 0 0 2 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Chloronitrile 4 0 0 0 0 2 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0
Diacylhydrazine 4 0 0 1 0 0 2 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Dinitroaniline 4 0 0 1 0 0 1 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0
Sulphite 4 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 4 0 0 0 0 0 0 0 0 0 0 0 0 0
Triazine 4 1 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 2 0 0 0 0 0 0 0 0
Amidine 3 0 0 0 0 0 0 0 2 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Total
Cereals and cereal primary derivatives
Ready−to−eat meal for infants and young children
Leafy vegetables
Root and tuber vegetables (excluding starchy− and sugar−)
Fruit used as fruit
Coffee, cocoa, tea and herbal ingredients
Herbs and edible flowers
Fruiting vegetables
Mammals and birds meat
Animal and vegetable fats/oils
Spices
Cheese
Legumes with pod
Legumes
Nuts, oilseeds and oilfruits
Sugar and other sweetening ingredients (excluding intensive sweeteners)
Processed cereal−based food for infants and young children
Food products for young population
Fruit / vegetable juices and nectars
Milk and dairy products
Wine and wine−like drinks
Processed fruit products
Processed or preserved vegetables and similar
Starchy roots and tubers
Unprocessed eggs
Algae and prokaryotes organisms
Bulb vegetables
Flowering brassica
Fungi, mosses and lichens
Milk, whey and cream
Savoury extracts and sauce ingredients
Stems/stalks eaten as vegetables
Infant and follow−on formulae
Sprouts, shoots and similar
Number of contamination values
200
150
100
50

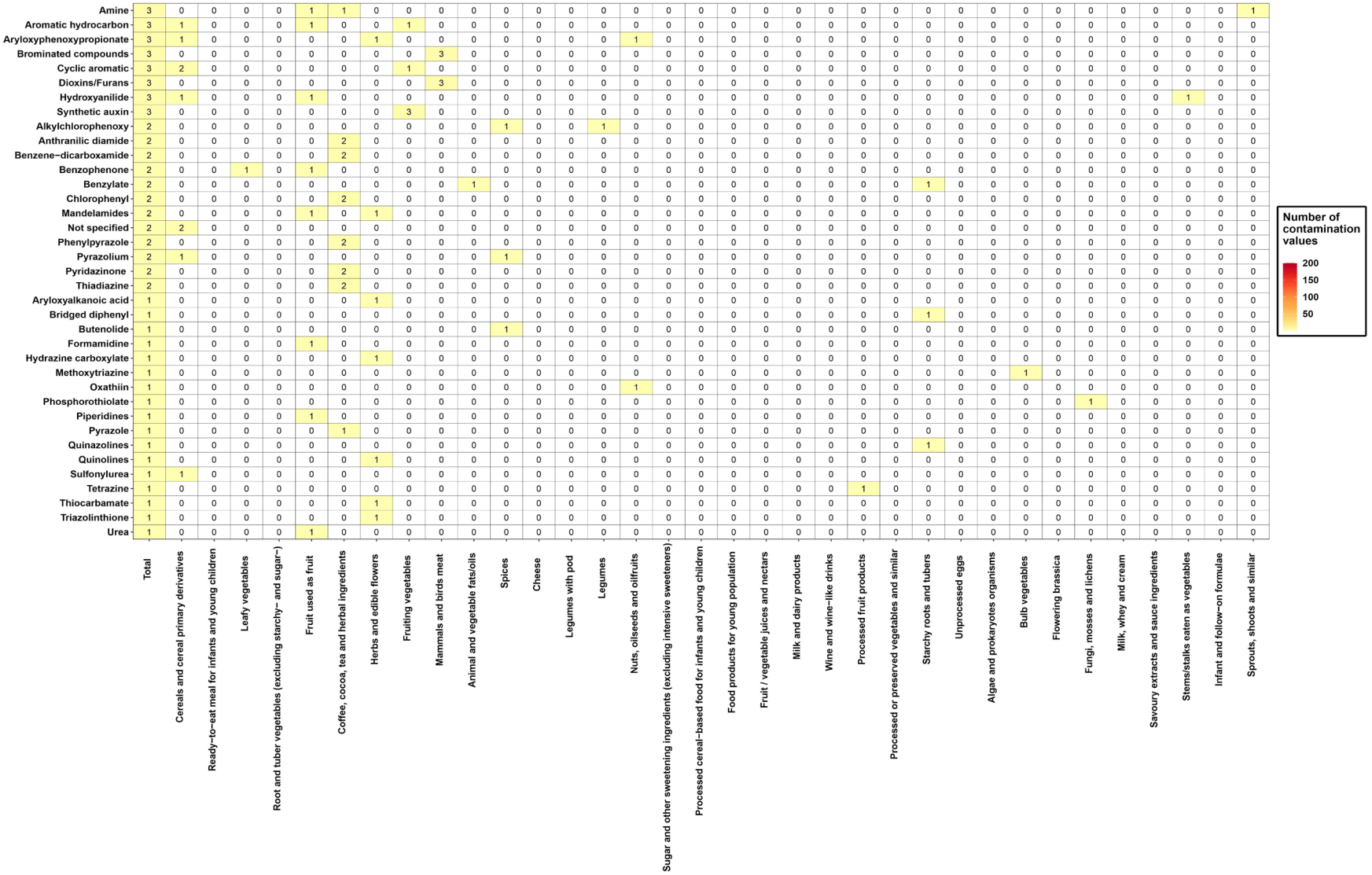


Figure 10. Matrix table of substance groups (rows) versus FoodEx groups (columns). Numbers within cells represent the number of concentration values for the substance group - FoodEx group pair

The most prevalent groups were Organophosphates (191), Inorganic contaminants (132), chlorates (117), organochlorine (94) and PCBs (70) (column "Total" in Figure 10).

Figure 10 also provides the number of concentration values for substance group - FoodEx group pairs in the POFE database. With a total of 311 distinct pairs in the database, only 16 and 10 pairs had medium and high I value, respectively. Organophosphates in 3 groups, "Ready-to-eat meal for infants and young children" (67), "Cereals and cereal primary derivatives" (28) and "fruit used as fruit" (25), along with inorganic contaminants in "Cereals and cereal primary derivatives" (72), organochlorines and PCBs in the two groups "Cereals and cereal primary derivatives" (27 and 30, respectively) and "Root and tuber vegetables (excluding starchy- and sugar-)" (30 and 28, respectively) and nitrates in "Leafy vegetables" (29) were the pairs with the highest number of values in the table.

A figure similar to Figure 10 but dealing with the number of studies is presented in Figure A3-1 in the Supplemental Material. Inorganic contaminants and organophosphates were the most considered by the literature in terms of number of studies (17 studies each). Organophosphates in "Fruit used as fruit" were the most considered (11 studies), followed by organophosphates in "Cereals and cereal primary derivatives" (10 studies).

2. Quantitative analysis of POFE

Table 5. Summary of the descriptive statistics of the substance groups in substance group-FoodEx group pairs with medium and high scientific interest

Substance group	FoodEx	n	Min	Q1	Mean	Median	Q3	Max	unit
Organophosphates	Cereals and cereal primary derivatives	28	5.10	14.00	287.81	16.00	395.00	2000.00	$\mu g.kg^{-1}$
	Ready-to-eat meal for infants and young children	67	14.77	72.52	224.72	111.47	228.31	2583.93	$\mu g.kg^{-1}$
	Fruit used as fruit	25	11.00	38.00	768.43	66.00	299.00	5000.00	$\mu g.kg^{-1}$
	Coffee. cocoa. tea and herbal ingredients	13	61.00	69.00	200.92	120.00	305.00	650.00	$\mu g.kg^{-1}$
	Animal and vegetable fats/oils	11	0.001	0.0068	0.02	0.01	0.019	0.08	$mg.kg^{-1}$
Inorganic Contaminants	Cereals and cereal primary derivatives	56	2×10^{-4}	0.014	8.226	0.032	1.69	120	$\mu g.kg^{-1}$
		9	0.22	20	50.78	50	66.8	100	$\mu g.kg^{-1}$ fw
		7	0.04	0.05	0.076	0.08	0.1	0.11	$mg.kg^{-1}$ ww
	Mammals and birds meat	12	0.0003	0.001	0.006	0.0035	0.0095	0.023	$mg.kg^{-1}$ fw
		2	0.00	0.00	7.098	1.495	8.593	25.40	$\mu g.kg^{-1}$
	Leafy vegetables	4	0.06	0.27	24.02	3.2	50	64.36	$mg.kg^{-1}$
		9	14	20	53,44	55	79	100	$\mu g.kg^{-1}$ fw

Chlorates	Leafy vegetables	18	11.00	13.50	60.44	21.50	98.50	320.00	$\mu g.kg^{-1}$
	Herbs and edible flowers	17	11.00	16.00	66.82	24.00	55.00	340.00	$\mu g.kg^{-1}$
	Fruiting vegetables	15	11.00	14.00	26.47	18.00	42.00	57.00	$\mu g.kg^{-1}$
	Fruit used as fruit	11	0.01	0.013	0.05	0.02	0.017	0.38	$mg.kg^{-1}$
Organochlorines	Cereals and cereal primary derivatives	27	0.005	0.10	1,692	0.44	2,39	6,44	$ng.g^{-1}dw$
	Root and tuber vegetables (excluding starchy- and sugar-)	26	0.02	1.22	11.02	2.25	8.27	90.69	$ng.g^{-1}dw$
PCBs	Cereals and cereal primary derivatives	30	2×10^{-6}	0.005	0.023	0.01	0.02	0.160	$ng.g^{-1}dw$
	Root and tuber vegetables (excluding starchy- and sugar-)	28	0.02	0.05	0.20	0.11	0.22	0.85	$ng.g^{-1}dw$
Minerals	Cereals and cereal primary derivatives	35	0.890	2.39	10.16 5	6.66	14.33	36.48	$mg.kg^{-1}$ ww
	Fruit used as fruit	13	3.310	5.81	8.538	9.32	11.26	14.02	$mg.kg^{-1}$ fw
Neonicotinoids	Coffee. cocoa. tea and herbal ingredients	13	0.01	0.069	0.17	0.093	0.232	0.49	$mg.kg^{-1}$
	Spices	11	0.05	0.13	0.89	0.21	1.6	3.10	$mg.kg^{-1}$
Nitrates	Leafy vegetables	26	0.300	81.25	967.58	855	1707.8	3074.90	$mg.kg^{-1}$ fw
		3	0.200	567.6	785.40	1135	1178	1221	$mg.kg^{-1}$
Pyrethroids	Fruit used as fruit	15	0.002	0.003	0.265	0.056	0.115	1.1	$mg.kg^{-1}$
Benzimidazoles	Fruit used as fruit	13	55	55	517.5	630	893.50	1300	$\mu g.kg^{-1}$

PAHs	Coffee. cocoa. tea and herbal ingredients	17	20.00	24.00	39.97	27.00	37.00	180.00	$\mu g.kg^{-1}$
Micoorganisms derived	Fruit used as fruit	14	2.00	5.00	42.50	37.50	79.50	79.50	$\mu g.kg^{-1}$
Plant derived	Cereals and cereal primary derivatives	11	10.4	13.85	18.85	19.1	29.6	94.8	$\mu g.kg^{-1}$

"n" stands for the number of values for the substance group-**FoodEx group** pair**, "Min" for the minimum value, "Q1" for the 1rst quartile, "Mean" for the mean value, "Median" for the median, "Q3" for the 3rd quartile, "Max" for the maximum value, and "unit" for the unit of measurement of the concentrations. Units are expressed in weight per fresh weight (fw), weight per wet weight (ww), weight per lipid weight (lw), weight per dry weight (dw), or just the weight without details.**

Table 5 shows the descriptive statistics of the concentration levels of substance groups in the pairs with medium and high I. Large variations can be observed when comparing the minimum and the maximum values for a pair in Table 5. For example, there is an order of magnitude between the min and max values for the pair “organophosphates - coffee. cocoa. tea and herbal ingredients” and five orders of magnitude for “PCBs - cereals and cereal primary derivatives” while zero order of magnitude for pairs “chlorates - fruit used as fruit” and “plant derived - cereals and cereal primary derivatives”.

IV. Discussion

Organic farming is an environmentally friendly agricultural system that favors natural practices, notably avoiding the use of synthetic chemical plant protection products. It does, however, authorize the use of substances of natural origin, such as certain insecticides (e.g., pyrethroids from chrysanthemums or azadirachtin from neem trees) ((EC)N°2021/1165). Chemical substances that are ubiquitous in the environment are also likely to be found in OF (Almeida-González et al., 2012; Debnath et al., 2015; Dervilly-Pinel et al., 2017; Malmauret et al., 2002; Mao et al., 2021; Romero-González et al., 2011; Witczak & Abdel-Gawad, 2012). The growth of organic farming is remarkable both in Europe and worldwide, probably driven by growing consumer demand for food that meets environmental and sustainable development challenges. According to the Eurostat, the share of EU farmland devoted to organic farming has been increasing annually, with an increase reaching 9.9% of the EU's agricultural area in 2021(Eurostat, 2023). In addition, the United Nations Food and Agriculture Organization (FAO) reports that organic farming is practiced in 187 countries, with 72.3 million hectares of farmland for at least 3.1 million farmers (FiBL & IFOAM – Organics International (2023). In view of this rapid expansion and the growing importance of OF consumption, it is important to take account of this in the characterization of population exposure to substances present in food, as well as in food-related health risk assessments. However, unlike conventional agriculture, where there are quantitative databases on chemical substances present in foodstuffs, there is currently no equivalent database for substances present in OF.

Thus, the main objective of this work was to conduct an exhaustive literature search to collect levels of chemical substances present in OF in the EU region. These data have been organized in a database and have been described in such a way as to provide an overview of the contents of this database. At this stage, the study does not aim to compare the collected data with other modes of agricultural production, nor even to assess potential health impacts.

The results in this paper are based on the literature search as described above. Descriptive analyses were carried out including a ranking according to the degree of scientific interest to understand which items were the most described in the literature and which need to be studied further. FoodEx2 was chosen as the standardized system for categorizing and describing foods in this study because it has been adopted in EFSA and because 58% of the concentration values selected in this study come from EFSA report. As previously noted, the database was described on a large scale by grouping chemical substances into more general groups (pesticides, inorganic contaminants, minerals, dioxins, PCBs and brominated compounds).

During the data cleaning process, only 32 studies (22 scientific articles and 10 EFSA database) out of 12500 articles met the inclusion criteria, i.e., 0.25% of inclusion. Such a small proportion of results indicates that the presence of chemical substances in OFs is not much studied in the literature, particularly in the EU region, and that the database will need to be supplemented with future studies. However, despite the small number of selected articles, we have collected data for 166 matrices, 209 substances and 823 matrix - substance couples already constituting an interesting database that could be continuously updated as research in this field progresses.

The number of values for each variable (also characterized using the interest I) are very heterogeneous. The majority of variables were classified as having a low I, i.e., less than 10 values in the database. Indeed, 132 matrices out of 166, 180 chemicals out of 209 and 814 matrix - substance couples out of 823 have a low I. However, 13 matrices (including baby foods other than processed cereal-based foods, tea and wheat), 10 chemicals (including chlorate and fosetyl-Al) and 1 matrix - substance couple (baby foods other than processed cereal-based foods - Fosetyl-Al) have a high I (Figures 4, 6, 8). The origin of the values is also very heterogeneous. Indeed, 2 matrices (tomato and wheat) were processed in 12 articles out of 32, 5 chemical substances (chlorpyrifos, spinosad, imidacloprid, acetamiprid and boscalid) in 11 articles out of 32 and 2 couples (Baby food other than processed cereal based food, fosetyl-Al, and tea, anthraquinone) in 6 articles out of 32, while all the others (74 matrices, 110 chemical substances and 738 matrix - substance couples) were considered in just one study (Figures 4, 6, 8). This highlights the inadequacy and heterogeneity of the existing literature, and underscores the need for further, more exhaustive studies.

In terms of OF matrices, "Baby foods other than cereal-based foods" has the highest I with 80 values (Figure 4B). This can be explained simply by the fact that this food matrix is aimed at a particularly vulnerable population (Mandy & Nyirenda, 2018). In addition, the inventory showed that the rest of the food matrices ranked with a high I in the database were plant-based (Figure 4B). This is consistent with the fact that most of the values in the database correspond to pesticides (80% of total values) that are mainly related to plant products. In addition, particular attention should be paid to wheat and wheat products (wheat (undefined form), wheat flour and wheat grain), which were included in the high I category. This focus of the literature may be due to the fact that cereals are the main contributors to the world's food supply, accounting for more than half of man's total caloric requirements and the EU, with a population of 450 million, is the second largest consumer of wheat in the world, after China (Abis, 2023).

As far as chemical substances are concerned, the analysis of our database showed that the literature focused mainly on the study of the presence of pesticides in organic foods. In fact, 965 of the 1207 values in the database correspond to pesticides, in particular organophosphates (191 values), chlorates (117 values), organochlorines (94 values) and neonicotinoids (52 values) (Figure 10), which are pesticides not authorized for use in organic farming. Plant protection products of natural origin authorized for organic farming received less attention: only 31 values were found in the group of plant-derived substances (pyrethrins and azadirachtin) and 21 in the group of micro-organism-derived substances (spinosad). This highlights the lack of studies on the presence of residues substances authorized in OF, especially as the natural origin of these substances does not exempt them from a potentially toxic impact on human health (Mossa et al., 2018).

After organophosphate pesticides, inorganic contaminants had the most values in the database (132 values) (Figure 10), with cadmium and lead having the highest number of values (33 and 30 values respectively) (Figure 6B). This could be explained by the fact that lead and cadmium are among the 10 chemicals of greatest public health concern (WHO, 2019, 2023b). What's more, almost half of the values retrieved for cadmium and lead corresponded to cereals and cereal products (32 values out of 63 retrieved, data not shown). This is consistent with the fact that these substances accumulate particularly in cereal grains (Aslam et al., 2021; Zhou & Li, 2022).

Other chemical groups, such as PCBs, dioxins, minerals, and brominated compounds, were given little consideration in the literature reviewed (number of values 70, 3, 59 and 3 respectively) (Figure 10). However, this does not mean that they are of lesser health concern, or that their presence in OF is less likely. For example, PCBs are ubiquitous and long-lasting environmental contaminants, which have been shown to be present in higher concentrations in OF matrices to matrices from conventional agriculture, particularly in meat (Dervilly-Pinel et al., 2017) or other fat matrices such as cheese (Almeida-González et al., 2012). In addition, several European TDSs have shown that the risk associated with presence in food cannot be ruled out for a number of these substances. The French and UK TDS concluded that there were health concerns for certain inorganic contaminants, in particular lead, cadmium and inorganic arsenic (ANSES, 2011b; FSA, 2014c). The French TDS also showed concerns for PCBs and dioxins, certain minerals (e.g. copper), some mycotoxins (15-ac-DON and 3-Ac-DON), sulfites, acrylamide but also aluminum and methylmercury (ANSES, 2011b). Furthermore, a TDS conducted in Catalonia in food samples collected in 2008 revealed that weekly aluminum intake in children exceeded EFSA recommendations as a consequence of the higher intake of cereals (Perelló et al., 2015). There is therefore a real challenge to be able to consider the level of presence of all these substances in organic foods when assessing population exposures to chemicals in food, which will require additional studies.

Finally, with regard to the number of chemical substances found in the different matrices, it was observed that some matrices, such as rye grain, showed values for 38 chemical substances (Figure 5), while others, such as cabbage, showed values for only one chemical substance (POFE). This does not mean that some matrices are more contaminated than others, only that they have not been studied in the same way in the literature. The same applies to chemical substances. Some, such as chlorate, are found in 52 different matrices (Figure 7), while others have only been detected in 1 matrix (e.g., urea) (Figure 10).

In addition, other substances likely to have harmful effects on human health have not been found from the literature: compounds newly formed during processing (acrylamide, PAHs) or substances migrating from materials in contact with foodstuffs (bisphenols, phthalates, per- and polyfluoroalkylates), remain largely unexplored.

As far as quantitative values are concerned, Table 5 shows a large difference between the minimum and maximum values obtained for most of the substance group-FoodEx group pairs represented, with orders of magnitude ranging from one (e.g. for the

organophosphorus/coffee, cocoa, tea, herbal ingredients pair) to five (e.g. for the PCB/cereals, primary cereal derivatives pair). This could be explained by the different origins of the matrices, the different methods of sample preparation which were not always specified (peeled, washed, etc.), and the difference between analytical methods. In addition, as the units were not all the same and were not interconvertible, it was sometimes impossible to compare values for the same pair.

Another limitation in the database is that the number of analyzed samples, as well as limits of detection and quantification, are not always indicated in studies. In addition, some concentrations are reported by ranges, others may be means, medians, a single value or sometimes this is not specified.

It should be emphasized that our approach to data search and selection, as well as the structure of the database, allows for the addition of further data, studies or articles as they become available in order to keep the table up to date. It will also be possible to include data on other types of chemical contaminants that have not yet been studied, or even to include contaminants of biological origin such as mycotoxins.

As it stands, this database cannot be used yet to draw conclusions on the concentration levels obtained in relation to other agricultural production modes. On the other hand, it brings together all the data published to date on OF contamination and could be an interesting tool for identifying the efforts needed to study and consider the consumption of products from organic farming in studies of population exposure to environmental pollutants. This will enable us to better understand and assess the potential benefits and points of vigilance to be had, regarding the consumption of OF.

Author contributions

JC: methods, data extraction, database formulation, data analysis, article writing

PP: design of the figures

FB: database analysis support

DJB: supervision, methods, validation, article proofreading and correction

CD: supervision, validation article proofreading and correction, responsibility for the project

Disclosure statement

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

Data availability statement

The data that support the findings of this study are openly available in Mendeley Data at https://doi.org/10.17632/sxtbf876xs.1.

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Chapter 4 - Comparative Analysis of Chemical Levels in Organic and Conventional Food Matrices Consumed in the European area

In this thesis, the evaluation of human exposure to environmental pollutants through organic and conventional food was initially intended. As no databases encompassing various chemical substances in organic food were identified in the literature, the decision was made to create a database on organic food (OF) in the European Union by gathering all data found in the literature. This undertaking was designed to facilitate the following steps:

1) Assessing the exposure levels of population from organic food

2) Comparing values from 1) with the exposures calculated using contamination values from EAT to evaluate the contribution coming from organic food in the total exposure.

Unfortunately, this plan couldn't be done due to the limitations of the database. These include:

1. Data heterogeneity

- While some pairs (chemical – food matrix) were single reported in the database (one value) others were found several times and in different articles. This posed a challenge in analyzing the gathered data: for pairs studied many times, measurement units from one article to another varied, making it impossible to interconvert units to homogenize the database and use the values for statistics to use in the calculation of exposure levels. As some measurements were done per fresh product weight, others were done per dry, wet or lipid weight; and some other times this specification was not mentioned.

- Another limitation was the difference in quantification methods, sample preps and other quantitative parameters including the number of samples included in each study.

This made it difficult to calculate the exposure level from organic food using these concentration levels.

2. Even if exposure levels could be calculated through the organic values from the constructed database, the samples tested in all included articles, were raw samples (or in their primitive forms). As for the values from the EAT database, those were for samples that were prepared as consumed. This limits the possibility of comparing the two exposure levels (ORG versus CONV), especially that the processing and transformation of food can lead changes in the levels of pollutants.

3. It is said that the tested samples from the EAT study include a portion of organic batch (≅10%) which is not mentioned in the study report but said to be estimated from the proportion of organic food consumed by the French population, as declared in the INCA2 study. Given this, even if we were able to calculate and compare the exposures from both modes of productions (ORG versus CONV), comparison wouldn't have been accurate due to the unknown proportion of organic food included in the tested samples from the EAT study.

To solve this, the first step we did was:

1. We sorted all the articles included in the database which presented values for the same pair from organic versus conventional production (22/32 articles). This will enable the accurate comparison in between the contaminants' level in ORG versus CONV, through removing all the limitations related to the differences in quantification methods, parameters, and measurement units.

2. We calculated an excess ratio (ER) as the reduced difference between the concentration of the contaminant in the organic batch and in the conventional batch from the same study. As such, an ER was calculated for every pair from the same study. With this ratio, we were able to overcome the limitation related to measurement unit differences in between the same couples from different studies and the results with ER for pairs were represented as distributions.

In a first step, this approach enabled us to evaluate the contaminant levels in an organic versus conventional matrix from what was found in the literature.

In a second step, the ER were used to calculate the relative exposure as a percentage to the proportion of organic food consumed. This portrayed the effect of organic food consumption on the exposure profile of the population.

This part will be covered in chapter 6.

In this chapter, only the first step is covered. Results will be expressed as mean ER (95%CI), with the distribution of the ER percentiles (2.5, 50, 97.5) for each pair.

This chapter is presented in an article format as it can be further submitted for publication.

Comparative Analysis of Chemical Levels in Organic and Conventional Food Matrices Consumed in European area

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Abstract

Objective: The study aimed to assess relative levels of chemicals in organic and conventional matrices consumed in Europe. **Methods:** Building upon a previous study, this research selected articles that included data on both organic and conventional products, enabling the evaluation of chemical levels in these food types. An excess ratio (ER) was calculated for each chemical – food matrix pair, using concentrations from both organic and conventional food. This method overcame differences in chemical levels between different studies, thus facilitating a comparison of contaminant levels. **Results**: Overall, chemical levels are lower in organic products than in conventional ones for contaminants (ER: -0.11) and authorized chemicals in conventional production (ER: -0.16). However, substances authorized in organic production showed higher organic product contamination (ER: 0.07). Limitations were found in drawing conclusions due to low number of data on some chemical categories including organic production-authorized substances. **Conclusion:** Contamination levels in organic food vary with the chemical substance and food matrix. Further comprehensive studies are necessary for better comprehension.

I. Introduction

Amid increasing concerns about the harmful effects of exposure to chemicals from food, especially pesticides, the demand for organic food has increased as a means of reducing exposure to these chemicals (Hyland et al., 2019). This has led to a steady growth of organic farming across Europe, expanding from 14.7 million hectares in 2020 to 15.9 million hectares in 2021, equivalent to 9.9% of the total agricultural area. Remarkably, France leads the European Union (EU) with the largest organic production area, reaching 2.8 million hectares in 2021(Eurostat, 2023).

Organic farming adheres to stringent regulations enforced by the European Union, primarily governed by Regulations (EC) No. 848/2018. These regulations encompass prohibitions on chemically synthesized plant protection products, mandates the use of specific natural fertilizers and natural pest control methods, the exclusive use of organically-farmed seeds while avoiding genetically modified organisms (GMOs), the promotion of organic matter recycling, the implementation of crop rotation, the preference for locally adapted plant varieties, dedicated attention to animal welfare, and the limited use of medicinal products for therapeutic purposes (European Parliament And Council, 2018). Furthermore, these regulations outline rigorous controls for labeling and provide an inventory of approved natural substances for organic farming, including azadirachtin from Azadirachta indica (neem tree seeds), Spinosad produced by Saccharopolyspora spinosa (a soil bacterium), copper, and pyrethroids derived from Chrysanthemum indicum (Chrysanthemum) (Regulation (EC) No. 2021/1165).

Yet, despite all the regulations, there are also ubiquitous environmental contaminations over which we have no particular control in organic products (Mie et al., 2017). Not to forget that, authorized substances, although of natural origins, can still have a negative impact on human health (Mossa et al., 2018). This raises the question of investigating the levels of chemicals in organic food.

As for the effect of organic food consumption on exposure levels, little is known. Limited studies especially those based on the questionnaire method were done to assess the exposure levels from organic food. The questionnaire method involves evaluating a population's dietary habits and estimating exposure to specific pollutants by multiplying the food intake with contamination levels obtained from national or specific databases. Using the questionnaire method, a study on a French population following vegetarian diets showed that there were

differences in lower levels of pesticide residues and cadmium in organic plant products compared to conventional ones, while more data are needed to establish whether the levels of nitrates, mycotoxins and some metals differ between organic and conventional products (Baudry et al., 2021). The lack of databases encompassing data on various chemical categories in organic products in the EU makes assessment through the questionnaire method rather hard.

The goal of this paper is to evaluate the relative levels of chemical substances in organic and conventional matrices consumed in the European area. More specifically, the key question we are interested in for a chemical-food matrix pair is to assess the extent to which the concentration of the chemical in the biological matrix differs from that in the conventional matrix taken as a reference.

Whatever the organic or conventional origin of the food matrix, concentrations of chemical contaminants can vary within the same farm or from one farm to another within the same year or from one year to another, for a whole range of reasons that are difficult to identify and list. The studies found in the literature therefore reflect the variability or heterogeneity of chemical levels, in addition to differences in the quantitative methods, from sample preparation to measurement units, used in the different studies. This makes it difficult to compare the levels of chemicals in food matrices from different studies. To avoid or limit these biases, which could lead to comparing what is not comparable, the comparison of chemical substance levels between organic and conventional foods was assessed using the excess ratio (ER). For a chemical-food matrix pair, the ER is calculated as the reduced difference in concentration levels in organic and conventional foods from the same study. As a result, we obtain distributions of ERs that can be used in further studies on exposure assessment to chemical from organic food consumption.

To do this, we used the POFE database (Choueiri et al., 2023) on pollutants present in organic foods and its counterpart for conventional foods.

II. Methods

Data

Data on pollutants present in organic and conventional foods were from the POFE database (Choueiri et al., 2023) and its counterpart, respectively. The POFE counterpart (cPOFE) is constructed from a subset of POFE for which data for both conventional and organic matrices were available and extracted from the same study or source. The database used for subsequent analyses consisted of chemical concentrations (in the same units) in conventional and biological matrices and number of samples (n) for each chemical-food matrix pair; each pair appearing as many times as there were different studies.

Data Analysis

The Excess Ratio (ER) for a chemical-food matrix pair was calculated as follows: $ER = [C_{org} - C_{conv}]/[C_{org} + C_{conv}]$, where C_{org} and C_{conv} are the contaminant concentrations in the organic and conventional food matrix, respectively, from the same study. As defined, ER > 0, ER = 0 or ER < 0 when the contamination level in the organic matrix is higher, equal, or lower, respectively, than that in the conventional one. ER tends to 1 in the limit when the contamination level of the organic matrix is quite larger than that in the conventional one and tends to -1 in the opposite limit when the organic matrix is almost free of contamination.

Distributions of ERs were generated as follows. Consider a chemical-food matrix pair from M different studies each of which with $n_{j,m}$ concentrations, (sample size) where "j = conv, org" for conventional (conv) and organic (org), and the study index, m = 1, 2, ..., M. First, we define for each study "m" the lower and upper bounds of concentrations as, $L_{j,m} = min[Concentration_{j,m}]$, and, $U_{j,m} = max[Concentration_{j,m}]$, respectively. Next, the distribution of ERs is given by the ensemble of values, $ER_{m,k,l} = [C_{org,m,k} - C_{conv,m,l}]/[C_{org,m,k} + C_{conv,m,l}]$, where $C_{j,m,k}$ is a concentration randomly generated from a uniform distribution in the interval, $[L_{j,m}; U_{j,m}]$ with, $k, l = 1, 2, \cdots, K \times n_{j,m}$), and K a larger number for statistics. $K = 1000$ was used for results reported in the work.

Results were described in three parts of chemicals corresponding to: (1) "contaminants" resulting from environmental contamination, (2) chemical substances "authorized in CONV"

representing phytosanitary products and additives authorized in the conventional mode of production, and (3) chemical substances “authorized in ORG” organic production.

The categorization of pesticides as contaminants or authorized in CONV farming was determined by referencing the status outlined in Regulation (EC) No 1107/2009 in 2021, at the time of data extraction. Those pesticides listed in the regulation during that period were considered authorized for use in conventional production. Conversely, pesticides not included in the regulation around year 2021 were classified as contaminants. For the additives, classification was based on the Regulation (EC) No 1333/2008 of the European Parliament and of the Council of 16 December 2008 on food additives and that of substances authorized in organic production was based on Regulation (EU) 2018/848 of the European Parliament and of the Council of 30 May 2018 on organic production and labelling of organic products.

All statistics and figures were done using Excel and R studio software.

III. Results

Database

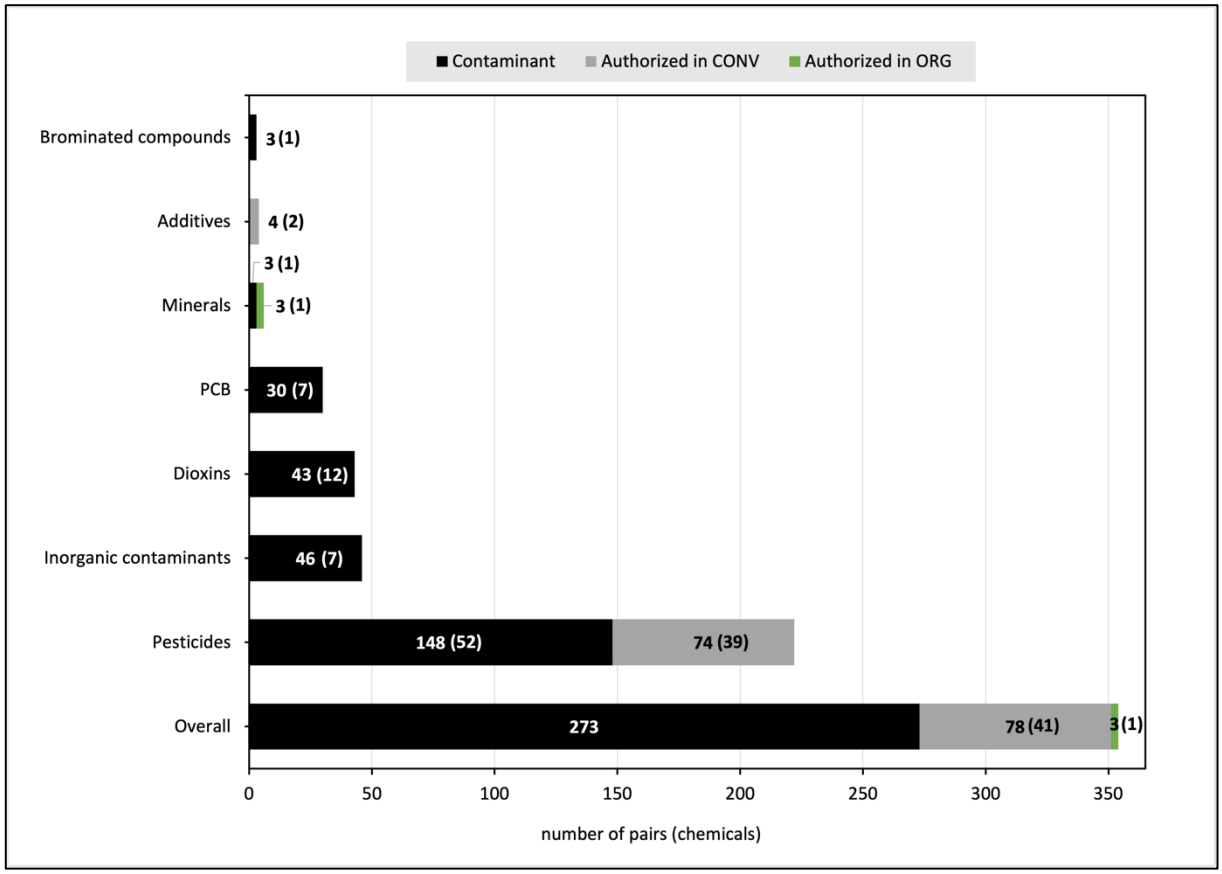


Figure 11. Summary of the descriptive statistics of the substance groups in substance group-FoodEx group pairs with medium and high scientific interest

Out of the 32 references in the POFE database, only 22 had values from both conventional and organic produce. The subset of POFE and cPOFE as described in Methods Section resulted in a database with a total of 354 unique chemical-food matrix pairs involving 122 distinct chemical substances.

Figure 11 visually illustrates the counts of pairs with involving chemical substances (between parentheses) across all chemical categories. The majority of data included were for contaminants (273 pairs) and only 3 pairs corresponded to one substance authorized in organic production (a mineral). The totality of pairs involved 7 chemical categories, with pesticides associated to most of the data (222 pairs; 148 involving contaminants and 74 involving substances authorized in CONV), and brominated compounds with the least number of data (3 pairs involving 1 chemical substance) (Figure 11).

Excess ratios (ER)

Figure 12 reports the distributions of ERs at a chemical category scale for the contaminants (Figure 12(A)) the substances authorized in CONV (Figure 12(B)) and the substances authorized in ORG (Figure 12(C)).

Overall, for the contaminants, the difference among the chemical categories for conventional versus organic was positive for PCBs (ER = 0.15) indicating higher levels in ORG, and negative for pesticides (ER = -0.22) indicating higher levels in CONV. For the substances authorized in conventional products, the overall ER for both the additives and pesticides were negative (ER = -0.06 and -0.17, respectively) indicating higher levels in CONV. And for the only substance present in the data and that is authorized in organic, the levels were higher in organic products (ER = 0.07).

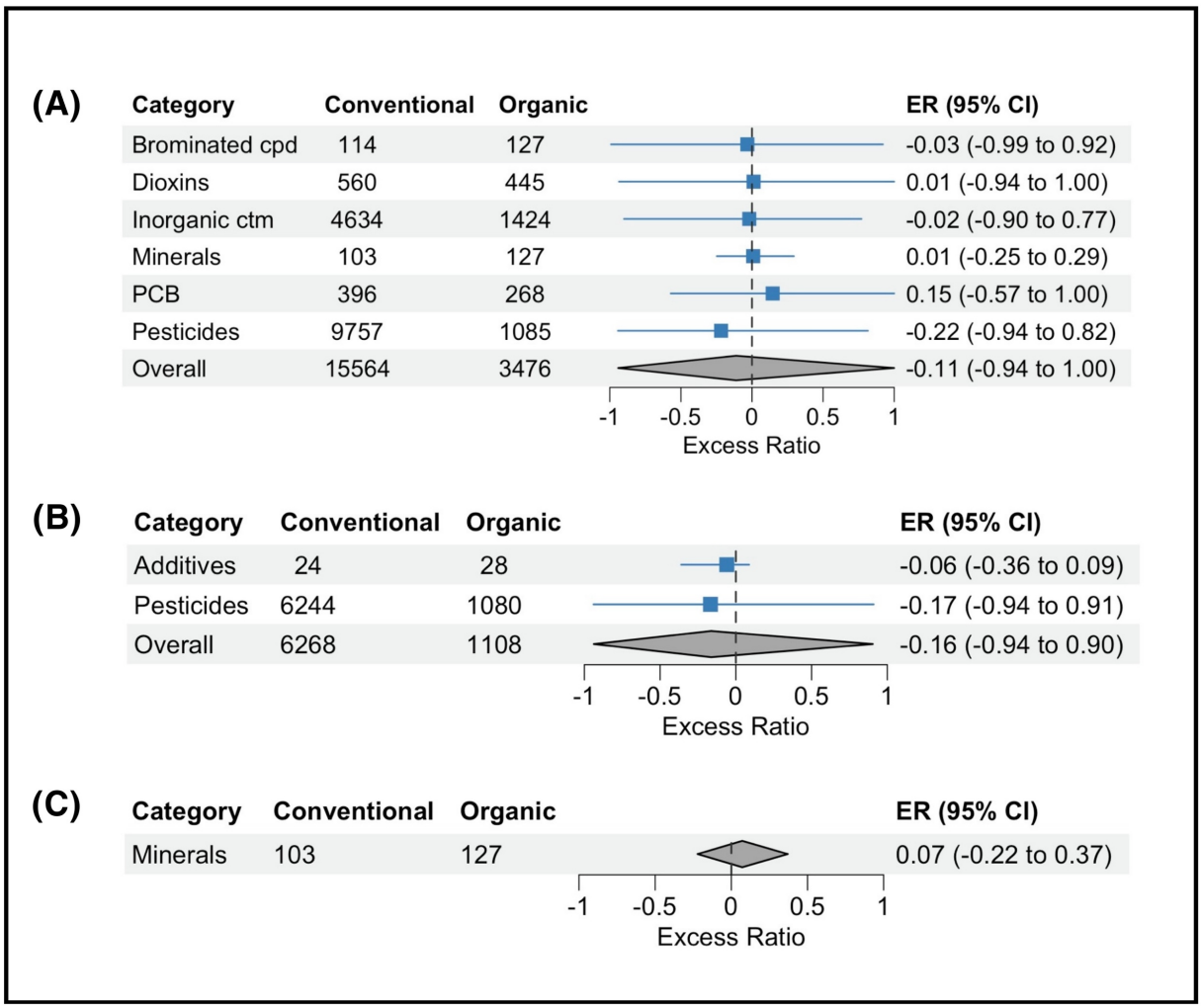


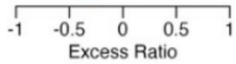
Figure 12. Distributions of Excess Ratios (ER) at chemical category scale for (A) Contaminants (B) authorized in CONV and (C) authorized in ORG

ERs for Contaminants

The distribution of ERs for pairs involving banned pesticides, is represented in Figure 13. Out of the 148 pairs, 52 (35%) have ER > 0 reflecting higher levels of contaminant pesticides in the organic produce, and 95 (64%) have ER < 0 reflecting higher levels of contaminant pesticides in conventional produce. Only a single pair (DDD o,p’ - wheat grain) had ER = 0, showing no difference in between organic and conventional produce. The pairs with the highest ER (> 0.5) corresponded to organochlorine pesticides (DDD p,p' (0.63), DDT o,p' (0.60), DDT p,p' (0.71), lindane (0.66)) with beetroots, organochlorine pesticides also (heptachlor (0.75) and heptachlor epoxide (1)) with carrots, chlorate and chard (0.82), chlorpropharm and lentils (0.82), hexachlorobenzene and poultry (0.66), chlorate and strawberries (0.62), and diphenylamine and tea (0.91) (Figure 13).

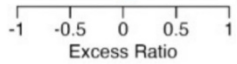
Pesticides

Product	Conventional	Organic	ER (95% CI)
∑DDTs - Cheese	54	7	-0.69 (-0.69 to -0.69)
∑OCP residues - Cheese	54	7	-0.74 (-0.74 to -0.74)
aldrin - Beetroots	5	5	0.36 (0.36 to 0.36)
aldrin - Carrots	5	5	-0.25 (-0.25 to -0.25)
aldrin - Rye grain	5	5	0.50 (0.50 to 0.50)
aldrin - Wheat grain	5	5	0.02 (0.02 to 0.02)
Aldrin_Dieldrin - Courgettes	1	2	-0.04 (-0.09 to 0.00)
Aldrin_Dieldrin - Cucumbers	13	1	-0.06 (-0.20 to 0.12)
Aldrin_Dieldrin - Pumpkins	2	3	0.24 (-0.01 to 0.43)
Amitraz - Honey	38	22	-0.66 (-0.84 to -0.11)
Anthraquinone - Tea (green, black)	485	17	-0.42 (-0.87 to 0.58)
BAC (RD) - Baby foods	1	2	-0.43 (-0.47 to -0.39)
BAC (RD) - Spinach	8	1	0.40 (0.15 to 0.72)
Biphenyl - Wheat	1	1	0.14 (0.14 to 0.14)
Carbaryl - Beans (dry)	1	1	-0.87 (-0.87 to -0.87)
Carbendazim - Guavas	22	1	0.21 (-0.15 to 0.80)
Carbendazim - Herbal infusions	6	1	-0.75 (-0.91 to -0.04)
Carbofuran - Buckwheat.pseudo-cereals	2	2	-0.23 (-0.25 to -0.21)
Chlorate - Apricot	1	1	0.41 (0.41 to 0.41)
Chlorate - Baby foods	11	8	0.20 (-0.35 to 0.89)
Chlorate - Bananas	13	1	-0.88 (-0.97 to -0.38)
Chlorate - Basil and edible flowers	42	6	-0.50 (-0.97 to 0.16)
Chlorate - Beans with pods	5	1	-0.14 (-0.48 to 0.56)
Chlorate - Broccoli	42	1	-0.71 (-0.90 to 0.06)
Chlorate - Celery	10	2	-0.28 (-0.54 to 0.17)
Chlorate - Chards	1	2	0.82 (0.80 to 0.83)
Chlorate - Chards/beet leaves	1	1	-0.15 (-0.15 to -0.15)
Chlorate - Coriander leaves	3	1	-0.39 (-0.58 to -0.10)
Chlorate - Courgettes	35	2	-0.47 (-0.95 to 0.43)
Chlorate - Cucumbers	58	4	-0.37 (-0.91 to 0.52)
Chlorate - Cultivated fungi	10	2	-0.62 (-0.86 to -0.07)
Chlorate - Ginger	3	1	-0.93 (-0.98 to -0.47)
Chlorate - Herbs and edible flowers	17	3	-0.98 (-1.00 to -0.90)
Chlorate - Horseradishes	5	2	0.25 (-0.10 to 0.67)
Chlorate - Kohlrabies	5	1	-0.35 (-0.56 to -0.00)
Chlorate - Lamb's lettuces	10	1	-0.77 (-0.92 to -0.07)
Chlorate - Land cresses	4	1	-0.82 (-0.89 to -0.69)
Chlorate - Lettuces	98	7	-0.32 (-0.95 to 0.84)



Pesticides

Product	Conventional	Organic	ER (95% CI)
Chlorate - Melons	5	1	0.36 (0.14 to 0.66)
Chlorate - Milk (sheep)	1	1	0.20 (0.20 to 0.20)
Chlorate - Milk.milk products	14	1	0.34 (0.21 to 0.49)
Chlorate - Oranges	25	3	-0.44 (-0.65 to 0.06)
Chlorate - Parsley	18	1	-0.69 (-0.88 to 0.01)
Chlorate - Peas (with pods)	84	5	-0.73 (-0.94 to -0.03)
Chlorate - Peas without pods	3	1	-0.42 (-0.54 to -0.25)
Chlorate - Plums	4	1	-0.22 (-0.41 to 0.05)
Chlorate - Potatoes	3	1	-0.88 (-0.97 to -0.22)
Chlorate - Radishes	2	1	-0.01 (-0.05 to 0.02)
Chlorate - Rice	12	2	-0.69 (-0.92 to -0.05)
Chlorate - Rucola	5	4	0.09 (-0.31 to 0.44)
Chlorate - Spinach	35	2	-0.66 (-0.93 to 0.23)
Chlorate - Strawberries	32	1	0.63 (0.39 to 0.91)
Chlorate - Sweet bell peppers	39	3	-0.65 (-0.98 to -0.03)
Chlorate - Sweet potato	3	1	0.25 (0.21 to 0.29)
Chlorate - Table grapes	6	3	-0.39 (-0.80 to 0.12)
Chlorate - Thyme	4	2	-0.50 (-0.96 to -0.01)
Chlorate - Tomato	39	4	-0.50 (-0.90 to 0.18)
Chlorate - Wheat	9	1	-0.38 (-0.53 to -0.16)
Chlorfluazuron - Tea (green, black)	2	1	-0.39 (-0.57 to -0.12)
Chlorothalonil - Potatoes	1304	38	-0.43 (-0.43 to -0.43)
Chlorothalonil - Tomato	1632	31	-0.60 (-0.60 to -0.60)
Chlorpropham - Apples	8	1	-0.66 (-0.85 to 0.00)
Chlorpropham - Lentils, dry	1	1	0.82 (0.82 to 0.82)
Chlorpropham - Potatoes	1767	43	-0.88 (-0.88 to -0.88)
Clothianidin - Maize	1	1	0.35 (0.35 to 0.35)
DDD o,p' - Beetroots	5	5	0.39 (0.39 to 0.39)
DDD o,p' - Carrots	5	5	-0.26 (-0.26 to -0.26)
DDD o,p' - Rye grain	5	5	0.14 (0.14 to 0.14)
DDD o,p' - Wheat grain	5	5	0.00 (0.00 to 0.00)
DDD p,p' - Beetroots	5	5	0.63 (0.63 to 0.63)
DDD p,p' - Carrots	5	5	-0.30 (-0.30 to -0.30)
DDD p,p' - Cheese	54	7	-0.33 (-0.33 to -0.33)
DDD p,p' - Rye grain	5	5	0.33 (0.33 to 0.33)
DDD p,p' - Wheat grain	5	5	0.16 (0.16 to 0.16)
DDE p,p' - Beetroots	5	5	0.34 (0.34 to 0.34)
DDE p,p' - Carrots	5	5	0.06 (0.06 to 0.06)



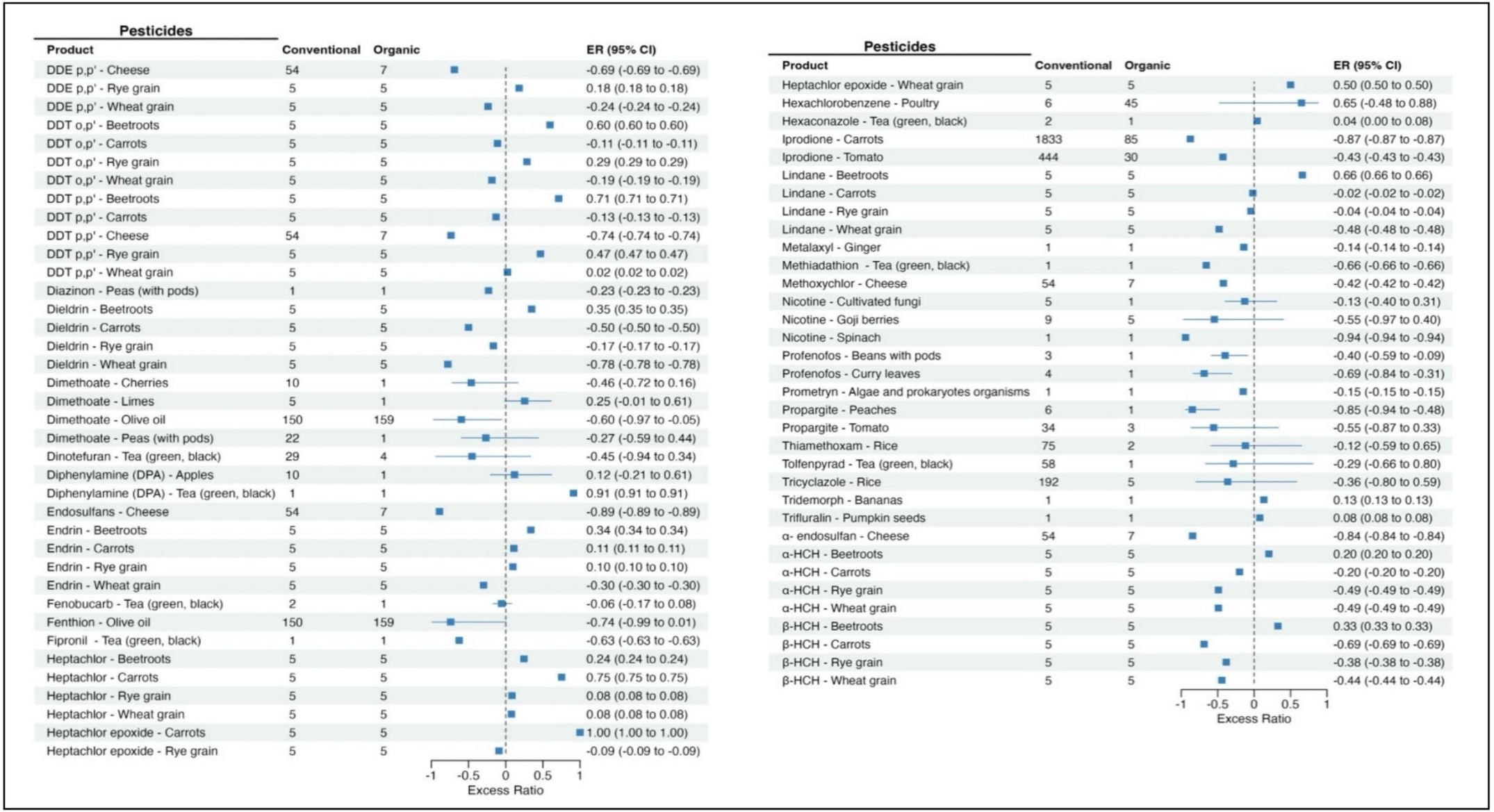


Figure 13. Distribution of Excess Ratios (ER) for pairs (chemical – food matrix) involving banned pesticides in farming since 2021 (contaminants)

As for the distribution of ER values for the pairs involving inorganic contaminants (IC), out of the 46 pairs, 16 (34.8%) reflected higher levels of IC in the organic produce (ER>0), 27 (59%) reflected higher levels of contamination in conventional produce (ER<0), and 3 pairs showed no difference between the two production types (ER= 0) (Cadmium in apples and barley and arsenic in carrots) (Figure 14). For pairs with the highest ERs for IC (>0.5), only one pair "Lead – Carrots" emerges with ER = 0.68.

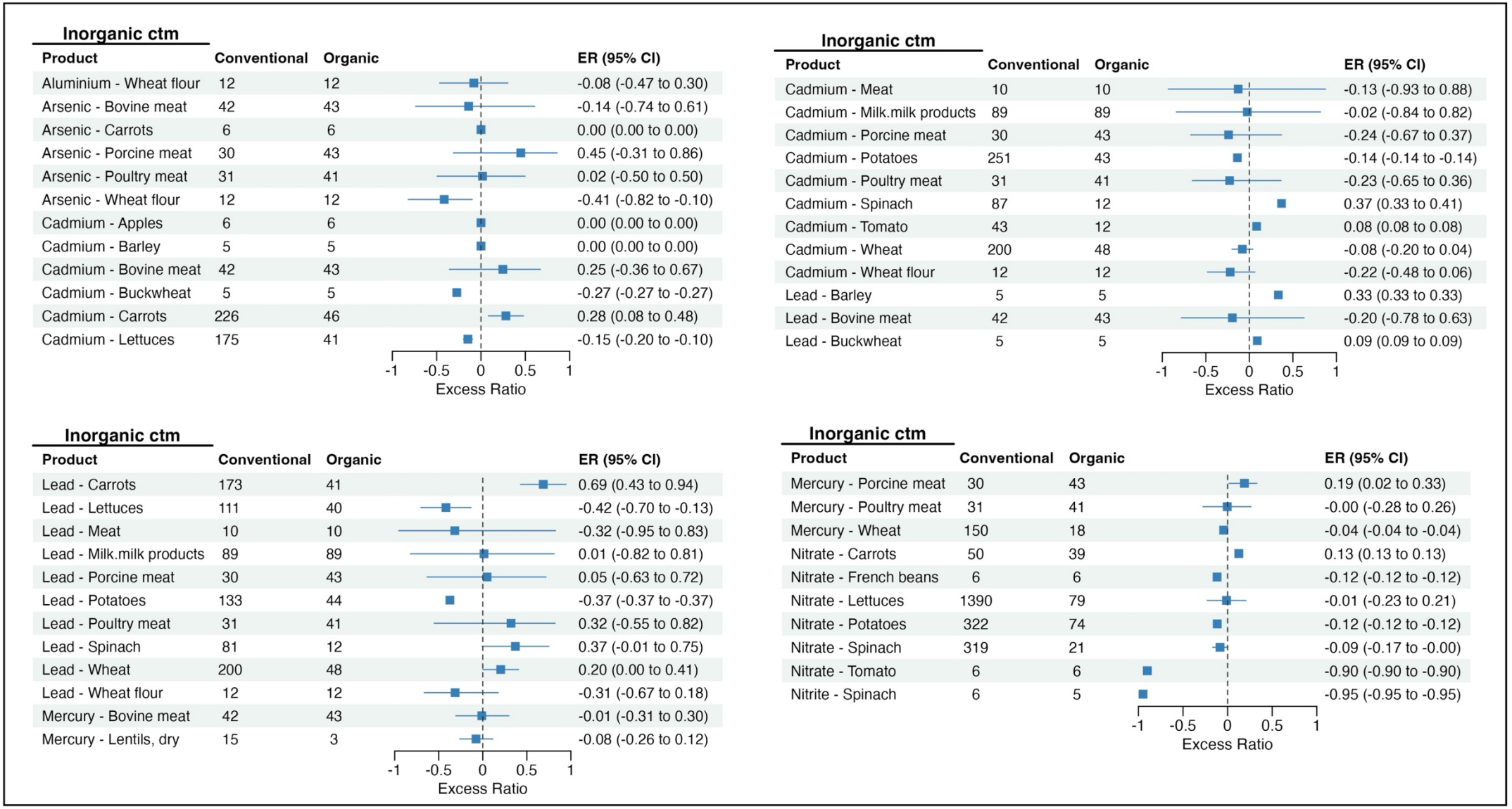


Figure 14. Distribution of Excess Ratios (ER) for pairs (chemical – food matrix) involving inorganic contaminants

For dioxins, out of the 43 pairs, 20 reflected higher levels of dioxins in the organic produce (ER > 0), 20 pairs reflected higher contamination in conventional produce (ER < 0), and 3 pairs (PCB 118-Carrots, PCB 157-Carrots and PCB 105-Wheat grain) showed no difference between the two production types (ER = 0). Pairs with highest ERs for dioxins (> 0.5) include PCB 105, 118, 126 with Beetroots, PCB77 – Carrots and PCB 77 - Wheat grain all with ER = 1, DL-PCBs - Poultry meat (ER = 0.54) and PCB 157 - Rye grain (ER = 0.6) (Figure 15).

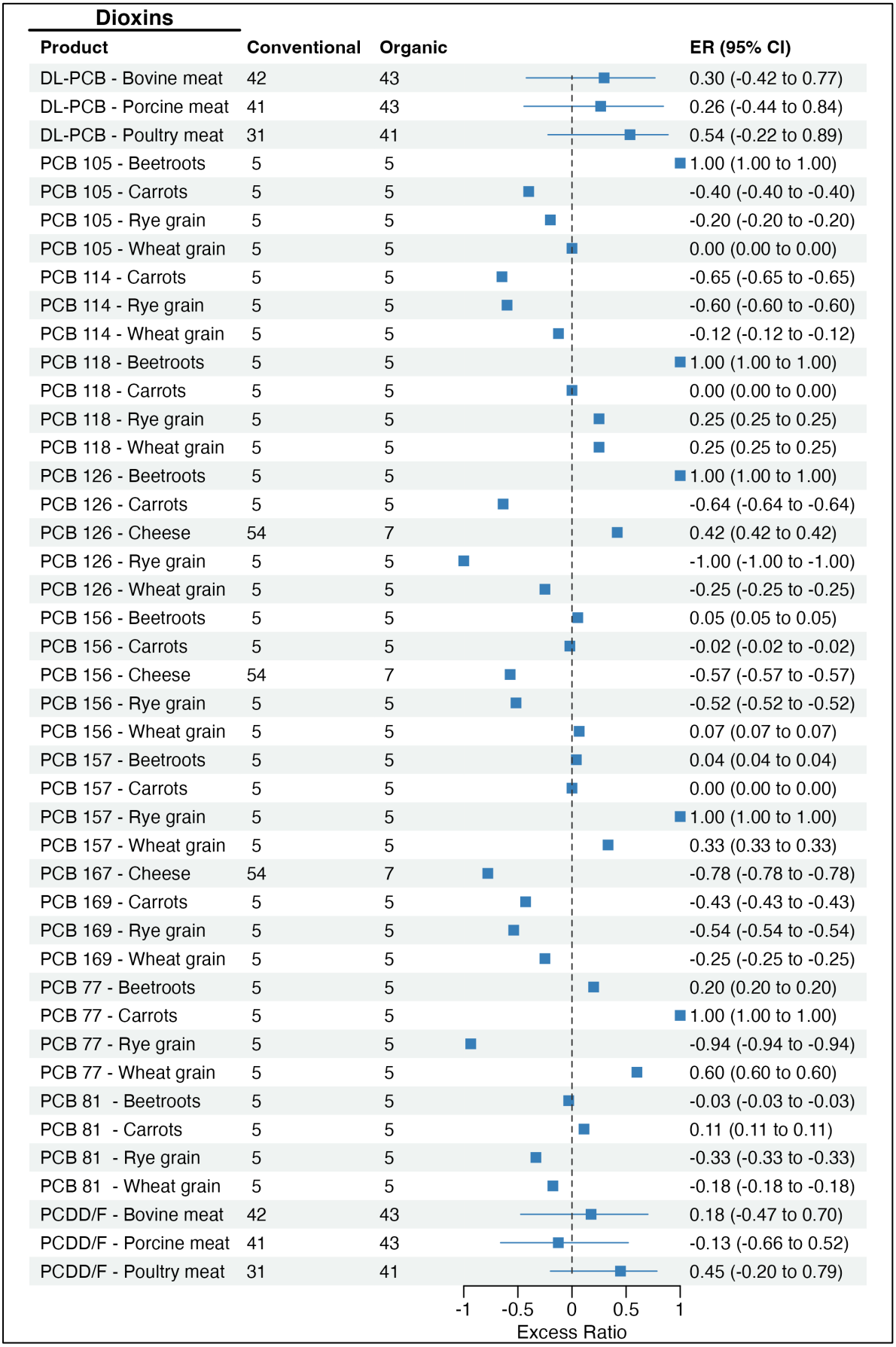


Figure 15. Distribution of Excess Ratios (ER) for pairs (chemical – food matrix) involving dioxins

Furthermore, for PCBs, out of the 30 pairs, 19 reflected higher levels of PCBs in the organic produce (ER > 0), 9 pairs reflected higher contamination in conventional produce (ER < 0), and 2 pairs (PCB 153-Carrots and PCB 138-Rye grain) showed similar contamination between the two production types (ER = 0).

Pairs with highest ERs for PCBs (> 0.5) corresponded to PCB 180 - Rye grain and PCB 180 - Wheat grain both with ER= 1 (Figure 16).

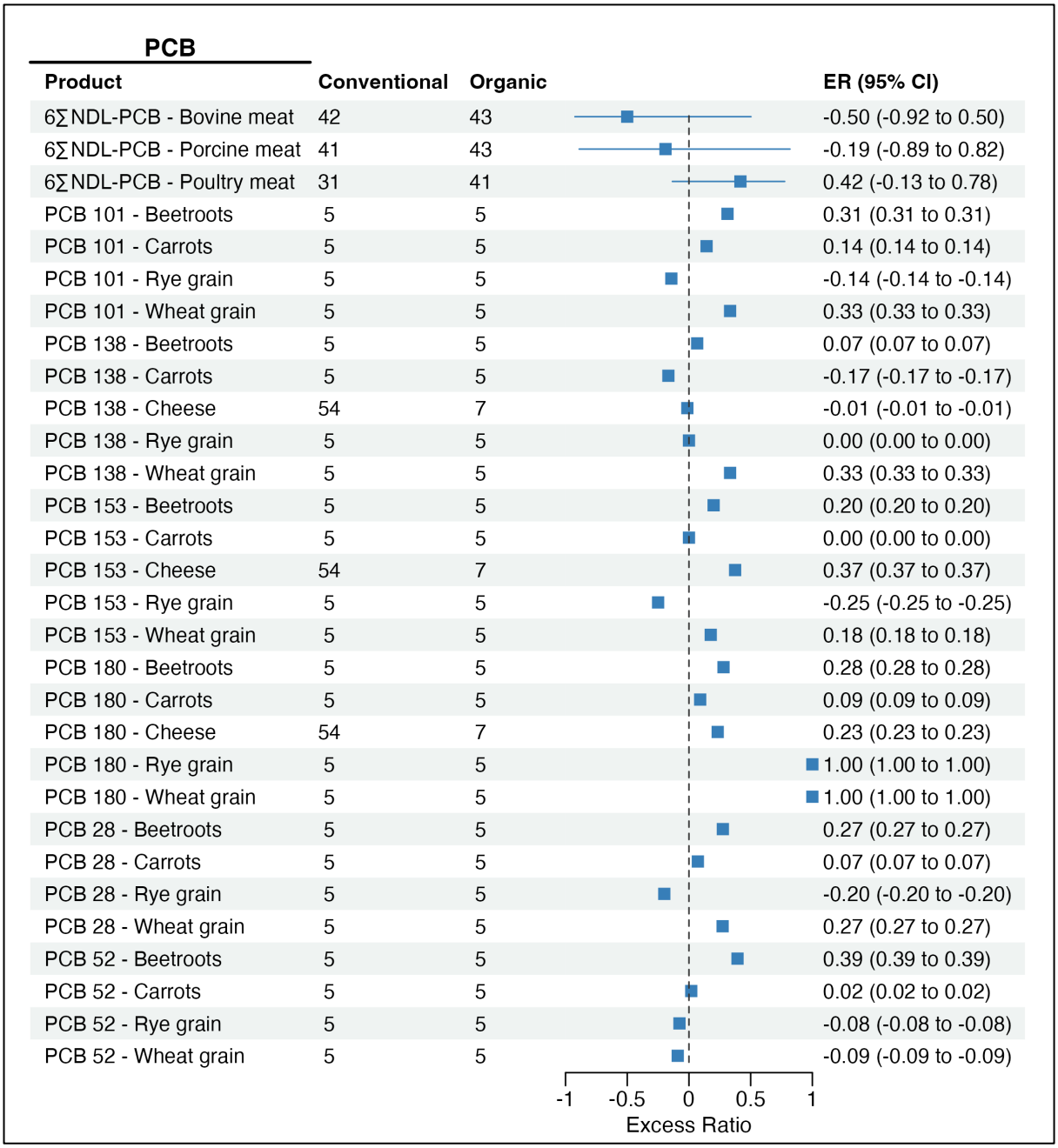


Figure 16. Distribution of Excess Ratios (ER) for pairs (chemical – food matrix) involving Polychlorinated Biphenyls

Finally, for the brominated compounds, in Bovine and poultry meat the contamination levels were observed higher in organic matrices (0.45 and 0.36, respectively) while for the mineral zinc, the difference in between tested matrices were negligeable (Figure 17).

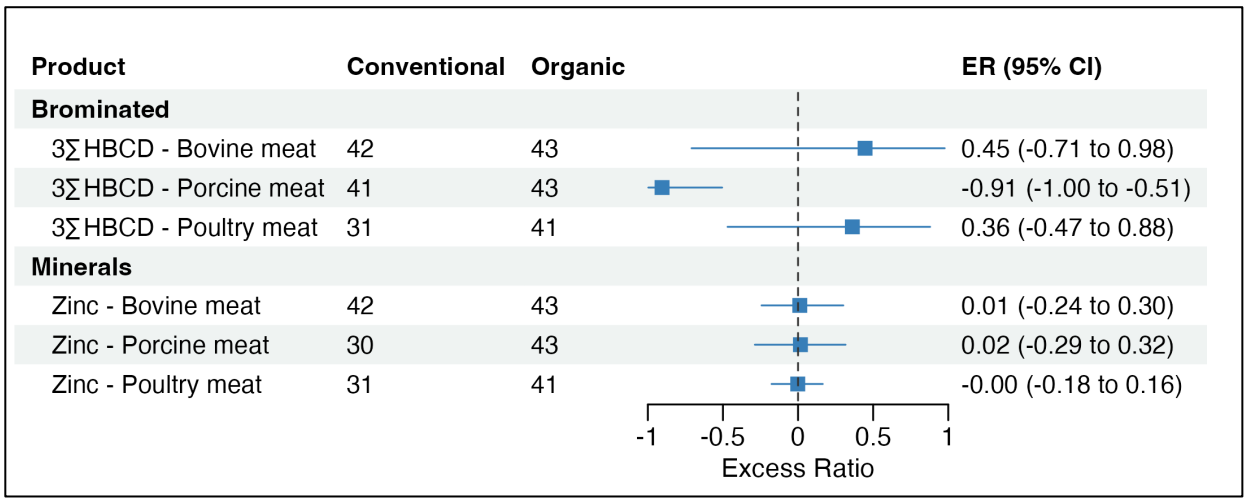


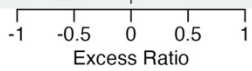
Figure 17. Distribution of Excess Ratios (ER) for pairs (chemical – food matrix) involving brominated compounds and minerals

ERs for substances authorized in conventional production

For the authorized pesticides in conventional production, ERs varied: out of the 74 pairs involving authorized substances in conventional produce, 26 (35%) had higher levels in the organic matrices (ER > 0), while 48 (65%) had higher levels in conventional matrices (ER < 0). In addition, 6 pairs had ER > 0.5 including: Thiacloprid - Honey, Chlorpyrifos - Laurel/bay leave, Thiophanate methyl - Lettuces, Chlorantraniliprole – Tea, Flubendiamide - Tea and Diflubenzuron - Wheat with ER = 1, 0.65, 0.93, 0.72, 0.87 and 0.83, respectively, (Figure 18).

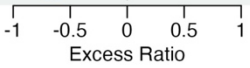
Pesticides

Product	Conventional	Organic	ER (95% CI)
2-phenylphenol - Tea (green, black)	3	4	0.09 (-0.24 to 0.63)
2-phenylphenol - Wheat	125	135	-0.27 (-0.27 to -0.27)
Acetamiprid - Tea (green, black)	143	2	-0.60 (-0.85 to 0.21)
Acetamiprid - Tomato	28	2	-0.18 (-0.73 to 0.49)
Azoxystrobin - Grape leaves and similar species	9	1	-0.82 (-0.94 to -0.24)
Boscalid - Grape leaves and similar species	9	1	-0.83 (-0.94 to -0.20)
Bromide ion - Brazil nuts	5	4	0.15 (-0.03 to 0.22)
Bromide ion - Carrots	1220	16	-0.32 (-0.82 to -0.02)
Bromide ion - Rice	5	4	0.10 (-0.05 to 0.21)
Bromide ion - Rucola	1	1	0.11 (0.11 to 0.11)
Bromide ion - Spinach	994	14	-0.48 (-0.83 to -0.30)
Bromide ion - Wheat	605	9	-0.14 (-0.54 to 0.22)
Buprofezin - Tea (green, black)	33	2	-0.44 (-0.83 to 0.09)
Carboxin - Poppy seeds	1	1	-0.68 (-0.68 to -0.68)
Chlorantraniliprole - Tea (green, black)	4	2	0.72 (0.36 to 0.92)
Chlormequate - Pears	388	13	0.17 (-0.88 to 0.54)
Chlormequate - Table grapes	1	2	-0.15 (-0.16 to -0.14)
Chlormequate - Wheat	125	135	-0.82 (-0.82 to -0.82)
Chlorpyrifos - Apples	75	1	-0.58 (-0.80 to 0.06)



Pesticides

Product	Conventional	Organic	ER (95% CI)
Chlorpyrifos - Beans (dry)	6	2	-0.12 (-0.41 to 0.24)
Chlorpyrifos - Buckwheat.pseudo-cereals	2	2	0.01 (-0.11 to 0.13)
Chlorpyrifos - Herbs and edible flowers	1	1	-0.99 (-0.99 to -0.99)
Chlorpyrifos - Laurel/bay leave	1	1	0.65 (0.65 to 0.65)
Chlorpyrifos - Lettuces	1	1	0.10 (0.10 to 0.10)
Chlorpyrifos - Olives for oil production	11	3	-0.40 (-0.78 to 0.01)
Chlorpyrifos - Parsley	3	1	-0.06 (-0.14 to 0.03)
Chlorpyrifos - Peaches	5	1	-0.36 (-0.58 to 0.04)
Chlorpyrifos - Table grapes	13	1	-0.13 (-0.45 to 0.38)
Chlorpyrifos - Tomato	43	3	-0.73 (-0.92 to 0.06)
Cypermethrin - Beans (dry)	1	1	-0.53 (-0.53 to -0.53)
Cypermethrin - Guavas	4	1	-0.19 (-0.40 to 0.14)
Cypermethrin - Pulses (dry), not specified	1	2	0.34 (0.24 to 0.42)
Cyproconazole - Coffee beans	1	1	0.35 (0.35 to 0.35)
Deltamethrin - Wheat	125	135	-0.17 (-0.17 to -0.17)
Diflubenzuron - Wheat	862	1	0.83 (0.83 to 0.83)
Dithiocarbamate - Cauliflowers	3	2	0.16 (-0.15 to 0.35)
Dithiocarbamate - Cultivated fungi	5	1	-0.61 (-0.79 to -0.17)
Dithiocarbamate - Spinach	7	1	-0.67 (-0.88 to 0.03)



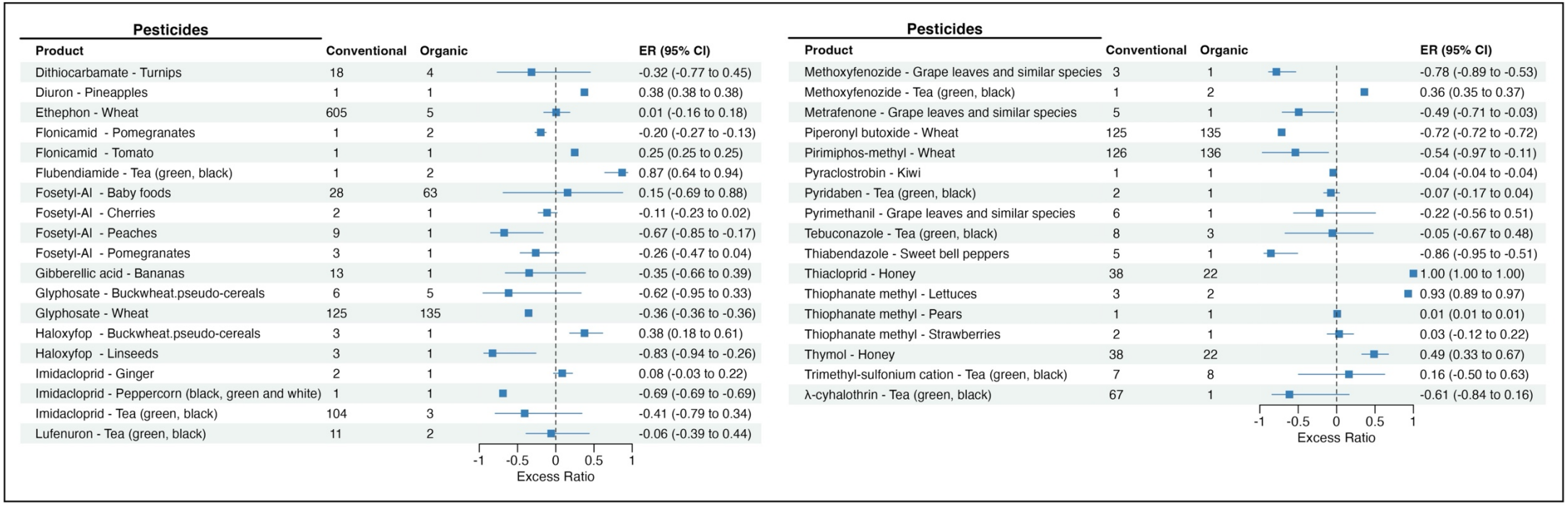


Figure 18. Distribution of Excess Ratios (ER) for pairs (chemical – food matrix) involving authorized pesticides in conventional production

As for the additives, ERs for Free and Total sulphite in Wine showed no difference between the two production types excepted for Free sulphite - White wine with ER = -0.36 (Figure 19).

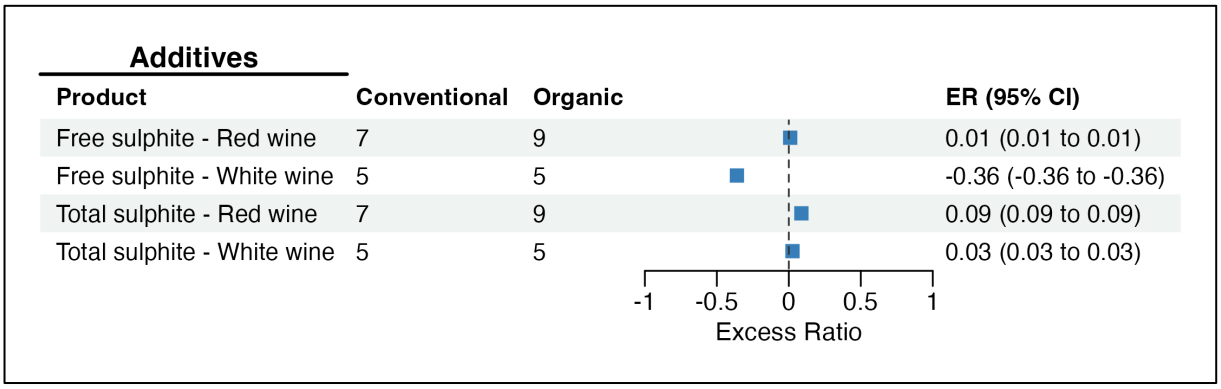


Figure 19. Distribution of Excess Ratios (ER) for pairs (chemical – food matrix) involving authorized additives in conventional production

ERs for substances authorized in organic production

For the substances authorized in organic production, only one substance resulted from the included studies — Copper, evaluated in bovine, porcine, and poultry meat. Figure 20 shows that the pair Copper- Porcine meat had ER > 0.

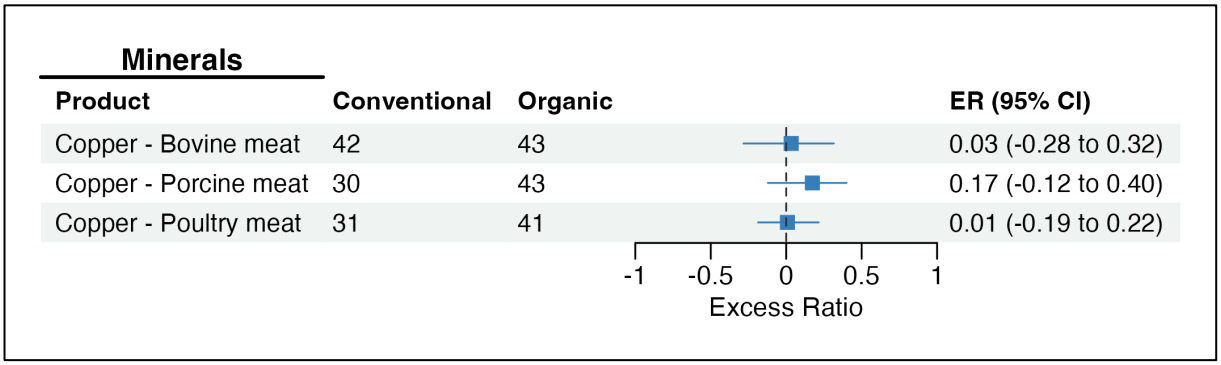


Figure 20. Distribution of Excess Ratios (ER) for pairs (chemical – food matrix) involving authorized substances in organic food production

IV. Discussion

This study aimed to address the question of whether levels of chemical in organic food in the EU are lower than in conventional food. To answer this question, data from existing literature on contamination levels in organic food and conventional food was gathered. Levels from the same articles were compared to eliminate all limitations related to quantification methods. Comparison between organic and conventional produces was done through the calculation of the Excess Ratio (ER) for pairs (chemical-food matrix) such that ER > 0, ER = 0 or ER < 0 indicating that the contamination level in the organic matrix is higher, equal, or lower than that in the conventional one, respectively. Results were described in three class of chemical substances: contaminant, authorized in conventional and authorized in organic.

In summary, for contaminants and the authorized chemical substances in conventional production, the overall ERs are -0.11and –0.16, respectively, reflecting a lesser contamination in organic food, while for the substances authorized in organic production, ER was equal to 0.07 reflecting higher levels in organic products. However, no conclusions can be drawn for substances authorized in organic production given the very low number of data.

For contaminants, main differences between organic and conventional food were observed for pesticides and PCBs. Pesticide contamination was lower in organic food (ER = -0.22), while PCB contamination was higher in organic food (ER=0.15). This confirms that difference in contamination between organic and conventional can be quite variable depending on the chemical category, or even substance considered, and on the food matrices.

For the banned pesticides, highest ER were fond for root plants with the persistent organic pollutants (POPs), organochlorine pesticides (DDD p,p', DDT o,p', DDT p,p', lindane, with beetroot and heptachlor, heptachlor epoxide with carrots) and poultry with hexachlorobenzene which is also an organochlorine pesticide. For the other pairs with ER > 0.5, no conclusion can be drawn given the organic sample number ≤ 2. As for the other contaminants, ERs > 0.5 resulted for pairs involving Beetroot (with PCB 105, 118 and 126), Carrots (with PCB77 and lead), Poultry meat (with DL-PCB), Rye grain (PCB 157 and 180) and Wheat grain (PCB 77 and 180). Moreover, for the brominated compounds, in Bovine and Poultry meat the contamination levels were found higher in organic matrices (ER = 0.45 and 0.36, respectively).

The contamination in organic samples, especially for plant-based food could be due to multiple factors, including a recent shift to organic farming in the area might have impacted the soil's

contamination levels, or the proximity to toxic waste repositories, or to a dirt road. Also, the differences in how contaminants accumulate in various plant species or varieties, especially in carrots and beets, could also be a significant factor. Carrots, containing carotene, known for its tendency to accumulate compounds, might explain why they exhibit higher levels of certain pollutants especially POPs compared to other analyzed vegetables (Witczak & Abdel-Gawad, 2012).

As for the increased levels detected in poultry meat, POPs tend to accumulate more in the fatty tissues of animals as they age. Given that organic farming practices often involve raising animals for a longer duration, the higher concentration of these substances in organic poultry can be attributed to such practice (Dervilly-Pinel et al., 2017).

For the pesticides authorized in conventional production, 6 pairs had ER > 0.5 including Thiacloprid - Honey (ER=1). For this pair, the result was from a study from Slovenia where Thiacloprid is authorized for use on oilseed rape, apples, pears, and ornamentals, the very plants from which bees gather pollen. Despite the honey being produced organically, the presence of thiacloprid in the honey indicates that bees collected residues of thiacloprid from their environment and transferred it to their hive during the honey-making process (Baša Česnik et al., 2019). A conclusion cannot be made for this pair based on a single study as well as for the remaining 5 pairs given the low number of tested samples ($n \leq 2$).

For the sulfite additives, although ER variations was mostly different from zero for Free sulphite in White wine (ER = -0.36) implicating lower levels in organic white wine, however, the presence of these chemicals in organic wine can be due to the natural occurrence in wine without being added as additives (EFSA, Younes, et al., 2022).

Finally, for the substances authorized in organic production, only copper resulted with bovine, porcine and poultry meat with ER > 0 for all three reflecting higher levels in organic. These results were retrieved from one study (Dervilly-Pinel et al., 2017). More studies are needed to be able to make conclusions on these substances, especially that many of the substances authorized in organic production, on a certain level, can have negative impacts on human health (Mossa et al., 2018).

It is worthy to note that for the substances authorized in organic production based on the regulation (EU) 2018/848 of the European Parliament, no specific MRLs are established. Thus, the same MRLs apply equally to organic and to conventional food (EFSA, 2018a).

No comparable studies were found in the existing literature, emphasizing the unique nature of this work. The absence of similar studies underscores the originality of our work and its contribution in addressing a notable gap in understanding the potential contaminations in organic food. Our study presents a novel insights, by evaluating the level of chemical substances in organic food and emphasizing the need for further quantitative and evaluation studies, especially that in the literature, there is little evidence on organic food consumption on human health.

Limitations of this study include, the low number of some samples analyzed, especially when there is a big difference between the number of conventional and organic samples. Another limitation is the heterogeneity of data where some chemical categories were not studied enough (e.g., minerals, brominated compounds). These two limitations hindered drawing definitive conclusions regarding the chemical levels in the matrices.

Further research is necessary, specifically requiring more quantitative studies comparing organic versus conventional produce. Additionally, there is a necessity for in-depth investigations into chemical categories, especially those demonstrating higher contamination in organic food. The standardization of quantification methods, sample preparation techniques, and reporting practices, including uniform units used for analysis, is crucial for facilitating easier data analysis from available literature.

To conclude, this study emphasizes that contamination levels in organic food significantly vary based on the specific chemical substance, its characteristics, and the matrix being studied. More comprehensive research is essential to understand the reasons underlying these variations and to identify other contributing factors that could influence contamination levels. The deeper understanding is important for the effective management and reduction of these contaminant levels to safeguard human health.

Chapter 5 - Food habits of university students in Grenoble, France: adherence to the National Nutritional Program and association with food insecurity

This thesis aims to examine the exposure profile of a specific population, namely the young adult population of university students. This group is of particular interest due to their increased susceptibility to developing Food Insecurity (FI) as a result of factors such as the transition to adulthood, moving out of the family home and living on a budget. Given the current rate of inflation, the likelihood of developing FI is expected to increase. Investigating the exposure of this population through what they eat could yield interesting results. The target population for this study is the students of the University of Grenoble Alpes (UGA), who are part of a larger, more comprehensive study assessing various aspects of their wellbeing in relation to their FI status. The diet quality of the students and the association with FI was carried out by this thesis as part of the work.

The first step to estimate the exposure levels of a population to chemical substances using a questionnaire-based method, is to assess their food intake. In this chapter written in the form of an article, the eating habits of the students were assessed and evaluated based on their compliance with the French Health and Nutrition Programme (PNNS), through a score developed for this purpose. A regression test was then performed to determine the association between adherence to the PNNS and the FI status of these students.

Food habits of university students in Grenoble, France: adherence to the National Nutritional Program and association with food insecurity

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Abstract

Purpose: This study aims to evaluate the eating habits of students at the University of Grenoble Alpes (UGA) using a score based on the French National Nutrition and Health Program (Programme National Nutrition et Santé (PNNS)) to assess their adherence to the dietary guidelines and explore the impact of food insecurity (FI) on their compliance with these recommendations. **Methods**: The dietary intake was assessed through an online diffused questionnaire, then the eating habits were evaluated through a developed PNNS score of 12 criteria. The FI status was previously assessed among these students in a study conducted by members of our consortium (Isoard-Gautheur et al., 2023). **Results**: Overall and across all diets, the rate of compliance with PNNS guidelines was 62%. As for the FI students, while their prevalence was relatively low (9%), this study showed that they had lower adherence to the fruits and vegetables ($p = 0.004$), the alcohol ($p = 0.005$), and milk and dairy products ($p=0.159$). Conversely, they exhibited higher adherence to the meat, poultry, and eggs criterion ($p = 0.021$) and the whole grain cereals criterion ($p = 0.01$). Additionally, FI students tended to experience more weight variations during the academic year. **Conclusion**: This study shows the compliance of UGA students to the different PNNS criteria highlighting the impact of FI on this conformity. This study can inspire universities to reflect on how to improve the well-being of students and provide guidance for policies aimed at improving the nutritional status of students.

Key words: Food insecurity, students, PNNS score, eating patterns, university, France

I. Introduction

The nutritional choices of university students draw significant attention globally, particularly because this population undergoes a critical phase of transition from adolescent dependence on parental support to young adulthood debuting their professional careers (Sprake et al., 2018). Assessing eating habits of university students is important at this phase, given that this transition can lead to the development of unfavorable eating habits (Papadaki et al., 2007) and a significant shift towards deteriorated, globalized behaviors (Chourdakis et al., 2010; Steptoe et al., 2002). Unhealthy dietary behaviors observed during the university years have been linked to increased weight gain (Anderson et al., 2003), which may persist throughout an individual's life (Winkleby & Cubbin, 2004).

Many studies have assessed the eating habits of students and have found that in general, university students are more likely to eat fatty, sugary and ready prepared meals, as they also tend to eat less fruits and vegetables (Sogari et al., 2018; Tanton et al., 2015; Yun et al., 2018).

In France, data on university students' eating behaviors is limited. A study assessed the consumption of six food groups (fruits and vegetables, nuts, legumes cereals, dairy and fish) for 3508 university students, showed that cereals were the only food which had more than 50% of compliance with the dietary recommendations (L. Miller et al., 2022). This raises questions about whether French university students follow healthy eating patterns.

One way to evaluate these eating patterns is through comparing them to official nutritional guidelines. In France, the National Health and Nutrition Program (PNNS: Programme National Nutrition Santé), initially developed in 2001 and coordinated by the French Ministry of health, provides guidelines serving as a valuable framework for promoting healthy eating habits and preventing nutrition-related diseases among the French population (Ministère des solidarités et de la santé, 2019). In general, these guidelines provide evidence-based recommendations for various aspects of dietary intake, including the consumption of fruits and vegetables, whole grains, dairy products, proteins, and the avoidance of excessive sugar, salt, and saturated fats. Until now four PNNSs have been established in 2001 (PNNS-1), in 2006 (PNNS-2), in 2011 (PNNS-3) and in 2017 (PNNS-4) (Manger Bouger, 2022).

Based on these guidelines, two PNNS scores have been developed to date: PNNS-Guidelines Score (PNNS-GS) (Estaquio et al., 2009) and PNNS-GS2 (Chaltiel, Adjibade, et al., 2019), as tools that measures adherence to the French dietary guidelines. Several studies have

used the PNNS score to assess the association between adherence to dietary guidelines and health outcomes (Chaltiel, Julia, et al., 2019; Estaquio et al., 2008; USDA, 2014).

PNNS scores has been applied to many populations including vulnerable groups such as pregnant women (Kadawathagedara et al., 2017), yet this score has not been widely applied for university students in France. An online cross-sectional study was performed in May 2021 among Rouen (France) university students to evaluate their eating patterns before and during COVID. The PNNS-G2 was used as a scoring tool, however the evaluation was only done for a limited number of food groups (6) (L. Miller et al., 2022).

The eating behaviors of university students can be determined by many factors most importantly the financial status of these individuals, as financial burdens often result in limited resources for students to meet their basic nutritional needs (Maroto et al., 2015; Von Ah et al., 2004). This results in food insecurity, a condition characterized by the Food and Agriculture Organization (FAO) as the state in which individuals lack reliable access to enough safe and nutritious food for regular growth, development, and the maintenance of an active and healthy lifestyle. This insecurity arises from factors such as the unavailability of food, limited purchasing power, improper distribution, or inadequate utilization of food at the household level. Typically, food insecurity manifests at four levels: (1) food secure, (2) mildly food insecure, (3) moderately food insecure, and (4) severely food insecure (FAO, 2023a). Numerous studies suggest that university students experience FI at greater rates than the general population (Abbey et al., 2022; Davitt et al., 2021; DeBate et al., 2021; Nikolaus et al., 2019).

In France, the general inspection of social affairs reported that almost 20% of university students lived below the poverty threshold, further exacerbating the odds of unhealthy eating (Marie E et al., 2015). Moreover, the economic difficulties faced by students have been exacerbated by the COVID-19 pandemic, which particularly affected the following years (2020-2021), leading to an increased level of precariousness, as evidenced by the long lines at charity organizations (e.g. “Restos du Cœur” and the Federation of General Student Associations (FAGE) (Jacquemart, 2021)) and food banks (e.g. University of Grenoble Alpes Foundation (Fondation Université Grenoble Alpes, 2021)).

Consequently in this study, first, the objective was to assess the eating habits of university students at the University of Grenoble Alpes (UGA), particularly because in this population, information on nutrition is not actively seeked or intentionally researched, with the word of mouth being an important source of food-related information (Paganelli & Clavier, 2023).

These habits were evaluated using a developed score based on the PNNS recommendations for the consumption of certain food categories (fruits and vegetables, organic food consumption, nuts, legumes, cereals, whole grains, milk and milk products, meat, poultry and eggs, red meat, cold cuts, fish and fish products and alcohol). Furthermore, a second objective was to examine the association between the students' food insecurity status and the compliance to the different PNNS criteria.

II. Methods

Participants and procedure

This study is a subset of a broader research initiative called PEANUTS (Précarité Etudiant·es Alimentation Nutrition UniversiTé Santé), which aims to investigate the well-being of UGA students and various aspects related to their level of FI. Initially, the entire UGA students population was targeted (59000 students); however, only 4012 students completed the food insecurity assessment part (Isoard-Gautheur et al., 2023). Only these students were therefore contacted to participate in the food habits survey. To assess their eating habits, a questionnaire was designed using a “Le Sphinx”, an internal survey software at Université Grenoble Alpes. This questionnaire was structured into four parts: Part 1 encompassed general inquiries such as type of residence, kitchen accessibility, and the availability of kitchen appliances and equipment; Part 2 focused on questions related to budget and spending habits concerning food; Part 3 concerned participants' self-assessment of their eating habits; and Part 4 concerned the type of diet, food consumption habits and a qualitative and quantitative assessment of food consumption. the questionnaire was sent by email to the participants. The email reemphasized the confidentiality of the study and the voluntary participation. Also, to encourage participation, the email announced a draw that would be done among the participants to win two free meals and a surprise package of sustainable food accessories. Informed consents were obtained from all participants in the beginning of every questionnaire. This study followed ethical principles outlined by the American Psychological Association (APA) (Ethics Committee of the American Psychological Association, 1996). Moreover, institutional approvals were obtained before conducting the research project from the data protection delegate of the targeted University.

Food consumption

Part 4 of the questionnaire assessed the consumption of 22 food categories in a way that students had to precise the number of portions for every food group along with the frequency of consumption (per day, week, or month). The assessed food categories were the following: meat, fish/shellfish, eggs, legumes, cereals (including bread), vegetables (including soups), potatoes, fruits, dairy products, soy-based deserts, vegetable oils, butter, nuts, snacks, pre-packed/pre-cooked meals, canned food, ready-to-eat sauces, confectionery, water, fruit/vegetables juices, wine/beer, and tea/coffee. A reference containing illustrations of estimated portion sizes relative to hand size for each food type has been provided to help students specify the portion size for each food consumed. The reference is available in the appendix A5 (Figure A5-1).

Diet type

Participants were also asked to specify whether they had a special diet (i.e. vegan, lactose-free, vegetarian, etc.).. These declarations were crosschecked with the food consumption of participants to assess their accuracy, particularly for those who declared special diets. A reclassification was done in cases where the type of diet declared did not correspond to food consumption data. Definitions of the most prevalent special diets ($n \geq 3$) are found in the supplemental material (Table A5-1).

French National Nutrition and Health Program adherence

To evaluate this French students' diet, the French National Nutrition and Health Program (PNNS) model was chosen as a reference for a healthy diet. Launched in 2001, the PNNS aims to improve the health of the entire population by acting on one of its major determinants: nutrition. A score was constructed to describe the adequacy of these students to the PNNS recommendations. To develop this score, a match was made with the dietary questionnaire, resulting in 12 PNNS criteria to be tested. These criteria evaluated the consumption of fruits and vegetables, of legumes, of cereals and cereal-based products, of whole grains cereals, of organic consumption (of fruits and vegetables, legumes, and cereals), of nuts, of milk and dairy products, of cold cuts (deli meat), of red meat, of meat, poultry and eggs, of fish and seafood, and of alcohol (Table 6).

Recommendations were quantified directly from the PNNS for all criteria, except for whole grain consumption, where some indirect conversion was made. In fact, the PNNS recommend

a daily intake of at least one portion of cereals in the form of whole grains, emphasizing their natural richness in fibers. This recommendation is linked to the consumption of cereal products. This recommendation couldn't be directly applied to the corresponding question of our questionnaire, because the questionnaire asks about the frequency of whole grain consumption, with a maximum frequency of 3x/week, so the score was translated as follows: the highest score was given to students who consumed whole grain products more than 3 times a week, while a score of 0 was given to those who consumed no whole grain products or those who consumed whole grain products never or less than once a month. Those who fell between these extremes were credited proportionally. All PNNS recommendations and scores for the criteria tested are detailed in the Table 6. Each criterion was scored out on an integer scale from 0 to 3 indicating 0 to 100% compliance with the recommendation, respectively. The 0 compliance with that recommendation meant either excessive consumption as in the case for alcohol, cold cuts, and red meat, or 0 consumption of that food group as in the case for all other groups. In some cases, the criterion score was either upgraded or downgraded, for example, in the case of a person who consumes fruits and vegetables with more than one portion corresponding to juice, one point was removed from the score for this criterion. Also, for the fish consumption criterion, for individuals with a consumption of fatty fish (Salmon) of 50% of their overall fish consumption, one point was added to the score for the fish criterion. Note that, these rules are in accordance with the PNNS guidelines where whole fruits are preferred over fruit juices due to their higher nutritious value, and fatty fish is recommended while also considering the potential exposure to linked pollutants.

Table 6. Score PNNS: Recommendations and Scoring. Quantities are defined according to the PNNS criteria

Group	PNNS		Consumption		Score
	Quantity	Frequency			
Fruits and vegetables	≥ 5 portions	daily	0 consumption		0
			(0, 2.5) portions/day		1
			[2.5, 5) portions/day		2
			≥ 5 portions/ day		3
			> 1 portion of jus		-1
Legumes	≥ 2 portions	weekly	0 consumption		0
			(0,1) portion/ week		1
			[1,2) portions/week		2
			≥ 2 portions/ week		3
Bread, cereals (B&C)	≥ 3 portions	daily	0 consumption		0
			(0,1.5) portions/day		1
			[1.5, 3) portions/day		2
			≥3 portions/day		3
Whole-grain cereals	≥ 1/3 cereal portions	daily	consumption of B&C=0		0
			consumption of B&C≠0	Never, < once / month	0
				once or twice/ month	1
				Once or twice/ week	2
				More than three times /week	3
Organic consumption	≥ 20% /week for 3 criteria: (1) F &V (≥7 ORG portions) (2) Legumes (≥0.4 ORG portions) (3) Cereals (≥4.2 ORG portions)	weekly	0 criteria fulfilled		0
			1 criterion fulfilled		1
			2 criteria fulfilled		2
			3 criteria fulfilled		3
Nuts	1 portion	daily	0 consumption		0
			(0, 0.5) portion/ day		1
			[05,1) portion/day		2
			≥1 portion/ day		3
Milk and dairy products	2 portions	daily	0 consumption		0
			(0,1) portion/ day		1
			[1,2) portions/day		2

			≥ 2 portions/ day	3
Cold cuts	≥ 150 g	weekly	[0-150] g / week	3
			>150 g/ week	0
Red meat	< 500g	weekly	<500g	3
			≥500g	0
Meat, poultry and eggs	1-2 portions	daily	0 consumption	0
			(0,0.5) portion/day	1
			[0.5,1) portion/day	2
			[1,2] portions/day	3
			≥2 portions/day	0
Fish and seafood	2 portions	weekly	0 consumption	0
			(0,1] portion/ week	1
			(1,2] portions/ week	2
			50% of consumed fish correspond to F&S salmon	+1
Alcohol	≤ 2 cups	daily	≤2 cups/ day	3
			>2 cups/day	0

Body mass Index

The Body Mass Index (BMI) was calculated as weight (kg)/height (m2). In terms of classification, individuals with BMIs below 18.5 were categorized as underweight, those with BMIs between 18.5 and 24.9 were considered normal, individuals with BMIs ranging from 25.0 to 29.9 were labeled as overweight, and those with a BMI of 30.0 or higher were classified as obese (Weir & Jan, 2023).

Food Insecurity

The student population surveyed in this work (4012 participants) is common with a previous study done to assess the relationships between food insecurity (FI) and physical activity (PA), with detachment from studies on one hand, and students well-being, on the other hand (Isoard-Gautheur et al., 2023).

A detailed description including the FI assessment method, which is a French version of the Food Insecurity Experience Scale (FIES), an eight key question scale developed by the Food and Agriculture Organization (FAO) (Cafiero et al., 2018), is available in Isoard-Gautheur et al.. Although this scale allows a classification based on four FI levels (secure, mild, moderate, severe) (FAO, 2023a), an additional grouping was performed in this work to homogenize the size of the groups given the low number of students in the severe FI group. This was achieved by grouping secure and low FI as the "non-FI" group, and moderate and severe FI as the "FI" group.

Statistical analysis

For the description of the students, continuous data were expressed as median values with the range from the minimum to the maximum, and categorical data were presented as frequencies along with their respective percentages.

Moreover, to comprehensively assess the overall adherence to the PNNS guidelines, the percentage of PNNS compliance for all 12 criteria was calculated for both the entire student population and the distinct diet type groups. The calculation of the percentage PNNS compliance involved the formula: $100 * \frac{\sum_{i=1}^{N} Wi * Ci}{3 \sum_{i=1}^{N} Wi}$

with *i* being the PNNS criterion, *N* being the total number of PNNS criteria (N=12), *Wi* being the proportion of compliant students and *Ci* being the score (0 to 3). This formula accounts for

the weighted sum of the products of the proportion of students (Wi) and their respective scores (Ci), normalized by the sum of the proportions of students. This approach ensures a comprehensive assessment, taking into consideration the diverse dietary patterns and varying degrees of adherence to each criterion. Following this, we further refined our analysis by computing the frequency of compliance, using the formula: $\frac{\sum_{i=1}^{N} \%\text{PNNS compliance i}}{12}$ where *i* is the PNNS criterion.

This approach allowed the evaluation of overall compliance of the entire student population and meaningful comparisons of compliance levels among different diet groups. Data analysis and figures were prepared using Excel and R Studio. In addition, a logistic regression was used to study the relationship association between students' food insecurity status and their compliance with the PNNS criteria assessed in the study. Results were considered significant when $p < 0.05$.

III. Results

Of the 4012 students contacted, 3707 to participate in this study. Subsequently, 384 responded to our questionnaire, with 308 responses meeting the criteria for completeness of response and validity of anonymity numbers. The participation pool was further refined to 257 individuals who provided comprehensive information regarding their dietary habits. Figure 21 provides a detailed outline of the selection process.

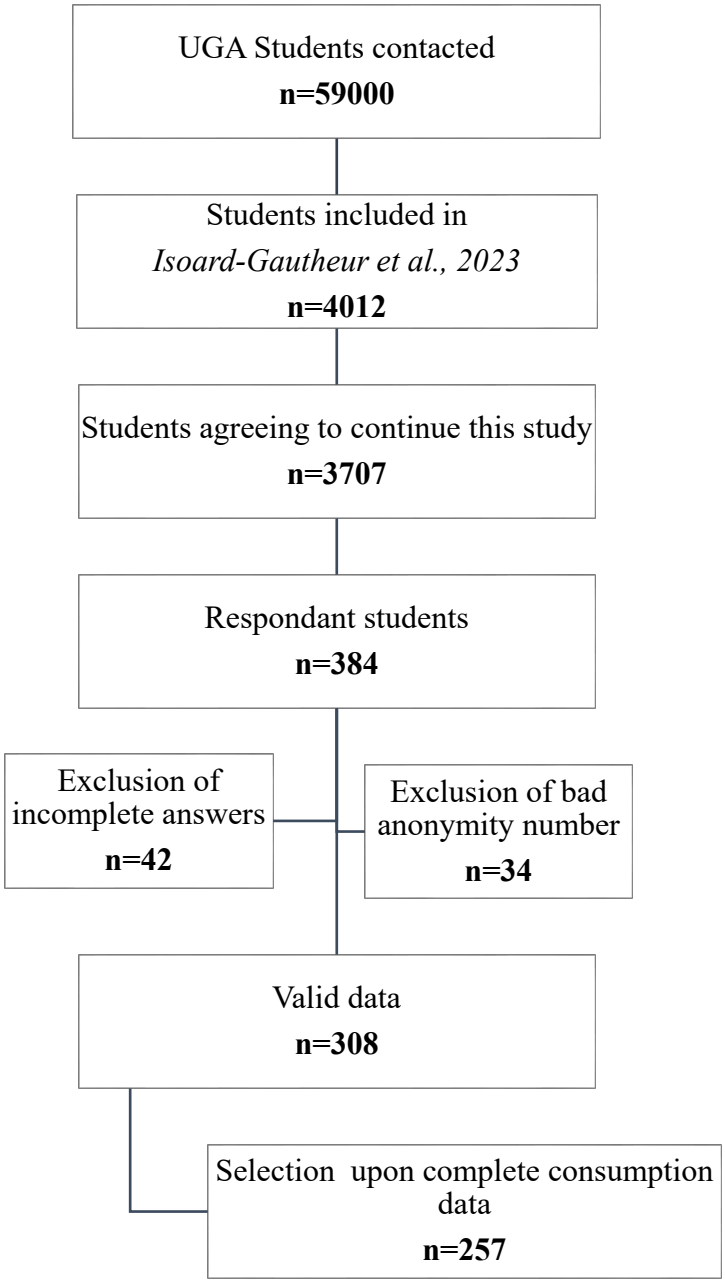


Figure 21. Participants selection criteria based on accepted participations, validity of the anonymity number, and completion of answers to the dietary questionnaire

As a result, students included in the study had a median age of 21 years, with the majority being females (72%). Table 7 provides a summary of the characteristics of the population. Most students had normal BMIs (74%), although 50% of students had experienced weight changes since the start of the academic year. Only 19% of students still reside with their families, while the majority (66%) allocate most of their funds to housing expenses, followed by 16% of students prioritizing their spending on food. As for the sources of income, 38% of students rely primarily on financial support from their families, while 18% depend on social aids. It is important to highlight that 23% of students sustain themselves through employment, whether it is a full-time job or a student/summer job opportunity.

Table 7. Student's characteristics (N=257)

Age in years (median (min; max))	21 (17; 50)
Gender (women/ men /other) (n, %)	186 (72%)/ 66 (26%)/ 5 (2%)
BMI (normal/ underweight/ overweight/obese) (n, %) (n=256)	190 (74%)/ 30 (12%)/ 25 (10%)/ 11 (4%)
Weight variation (yes/ no/ unknown) (n, %)	128 (50%)/ 96 (38%)/ 32 (13%)
Living in rental/ in shared flat/ with family/ in university residence/ other (n, %)	102 (40%)/ 64 (25%)/ 50 (19%)/ 35 (14%)/ 6 (2%)
Primary spending factor (housing/ food/ entertainment/ transportation/ school registration fees/ unknown/ phone and internet / other) (n, %)	169 (66%)/ 40 (16%)/ 13 (5%)/ 11 (4%)/ 10 (4%)/ 10 (4%) /2 (1%)/ 2 (1%)
Primary source of income (family/ social aids/ scholarship/ job/ student job or summer job/ other/ pension/ student loan) (n, %) (n=256)	97 (38%)/ 46 (18%)/ 41 (16%)/ 39 (15%)/ 21 (8%)/ 6 (2%)/ 4 (2%)/ 2 (1%)

Diet types within the population

Among the 257 students surveyed, the majority (190) did not adhere to any specific dietary regimen. However, a notable portion of students opted for various diet types, with ovo-lacto (16), ovo-lacto pescitarian (13), flexitarian (13), lactose-free (6), and vegan (6) being the most prevalent choices. For students following fewer common diets with a representation of $n \leq 3$, they were still considered in the study but were grouped together in an "all diet" category with

the other groups, to provide a comprehensive overview of dietary compliance within the entire student population.

Food insecurity among students

The students' population was categorized into two groups based on their FI status: 24 students (9%) identified as FI and 233 students (91%) identified as not FI. Figure 22 shows the distribution of students by FI status and diet type. Notably, FI students tended to follow fewer diet types. The lactose-free category had the highest proportion of FI students (50%), while there were no FI students in the vegan, flexitarian or ovo-lacto categories.

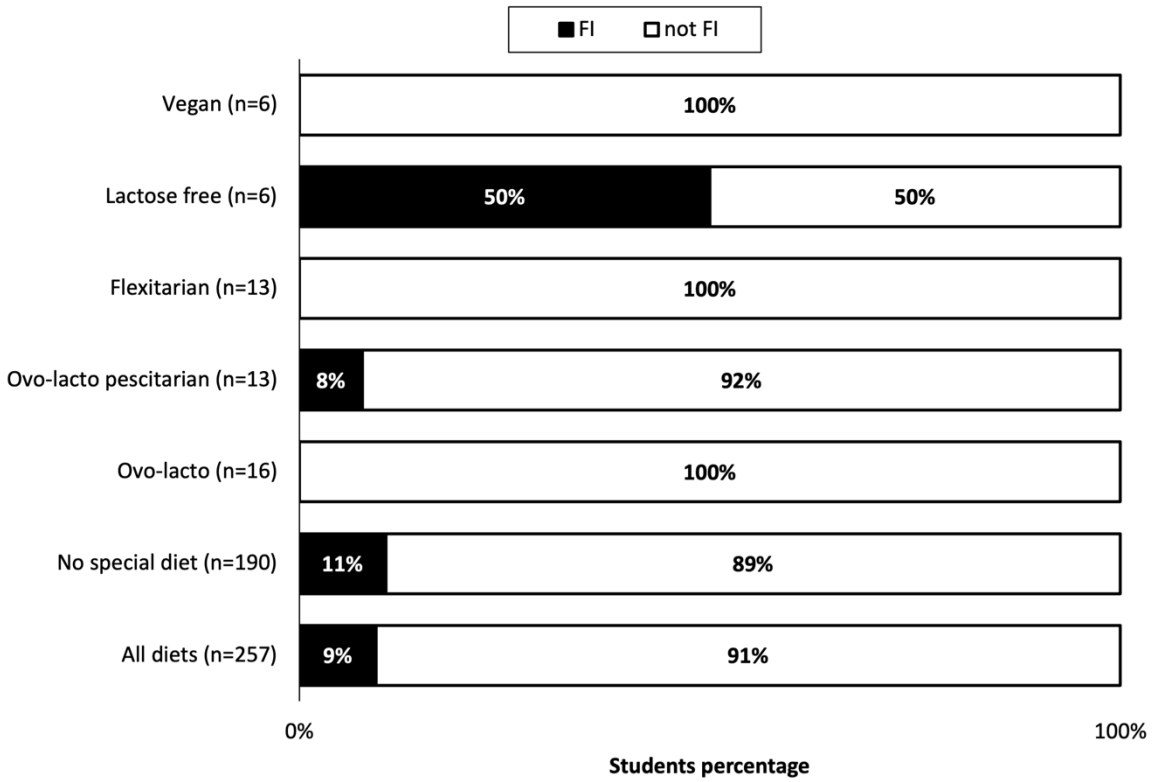


Figure 22. Percentage distribution of the total population and of the different diet type groups according to their food insecurity status

Body Mass Index and weight variation

Figure 23 (A) illustrates the percentage distribution of students based on their BMI status, highlighting the percentage of FI students in each category. The overweight class had a higher proportion of FI students (20%) compared to the other classes (7% in underweight, 8% in normal, and 9% in the obesity class). In addition, Figure 23 (B) shows the percentage

distribution of students based on their weight changes, revealing that a higher proportion of FI students (16%) witnessed weight changes since the start of the academic year compared to proportions of FI students who did not have any variations (2%) or those who have not noticed any variations. No valid statistical tests available to prove significance given the low number of FI students.

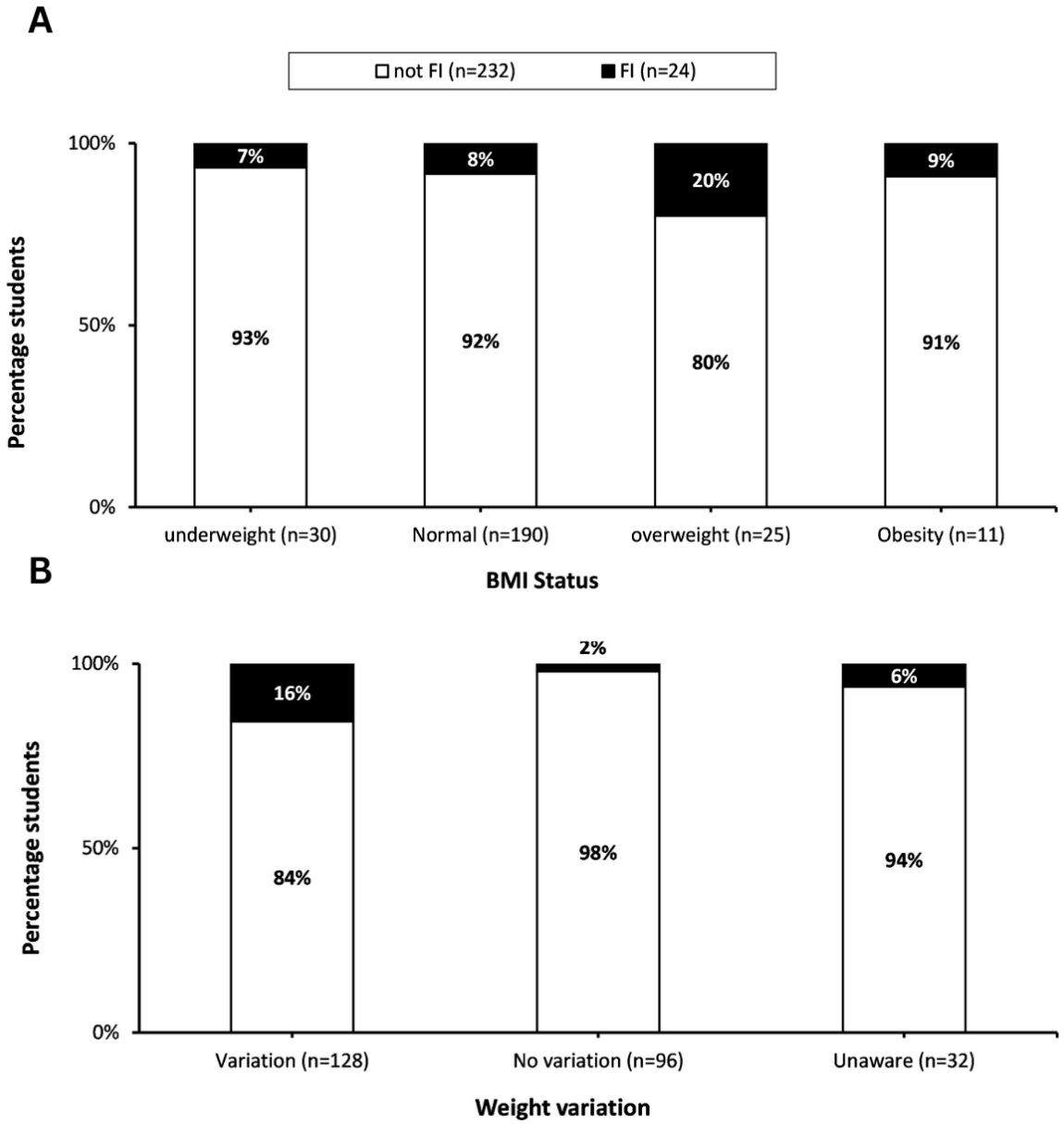


Figure 23. (A) Percentage distribution of students by BMI status, with prevalence of food insecurity status highlighted; (B) Percentage distribution of students by reported weight changes since start of academic year, with prevalence of food insecurity status highlighted

PNNS scores distribution

As detailed in the methodology section, a PNNS score was developed based on 12 specific criteria, with individual scores assigned to each criterion rather than calculating an overall score. Figure 24 shows a comprehensive breakdown of the students' scores for each criterion, encompassing the entire student population and different diet types of groups while considering the students' food FI status. To explain more the figure, the percentage without brackets correspond to the percentage of students in the total population and that in brackets represents the percentages from the FI group; we take the example of fruits and vegetables: 12% of the population with a score of 3 are not FI and make up 13% of the non-FI population. And 1% of the population with a score of 3 is FI and represent 8% of the FI population.

To begin, the scores for the entire population are described irrespective of their FI status: for the fruits and vegetables criterion and for all diet types, only 13% of the population were fully compliant to this recommendation (had a score of 3). The ovo-lacto (OL) group had the highest compliance rate (31%) while in the lactose free (LF) group and the flexitarian (F) group no score 3 were observed.

As for the legumes criterion, 51% of the population was compliant with the recommendation especially the V group, were all vegans consumed the required amount of legumes per week (≥2 portions).

Moreover, for the cereals and cereal based products group, only 23% of the population were compliant, with compliance being more evident in OLP (38%), F (38%), V (33%) and OL (31%), rather in the NSD (19%) group or LF group where no compliance was observed. Regarding whole grain cereals consumption, 15% of the population were completely compliant, with the OLP group having the highest number of score 3 (38%), and the LF group showing zero compliance to this criterion.

For the organic consumption of fruits and vegetables, legumes and cereals, 30% of the total population achieved full compliance. Higher compliance rates were observed in the vegetarian groups (F (69%), OL (44%), OLP (46%), and V (83%)) compared to the LF (17%) and non-specific diet (NSD) (25%) groups.

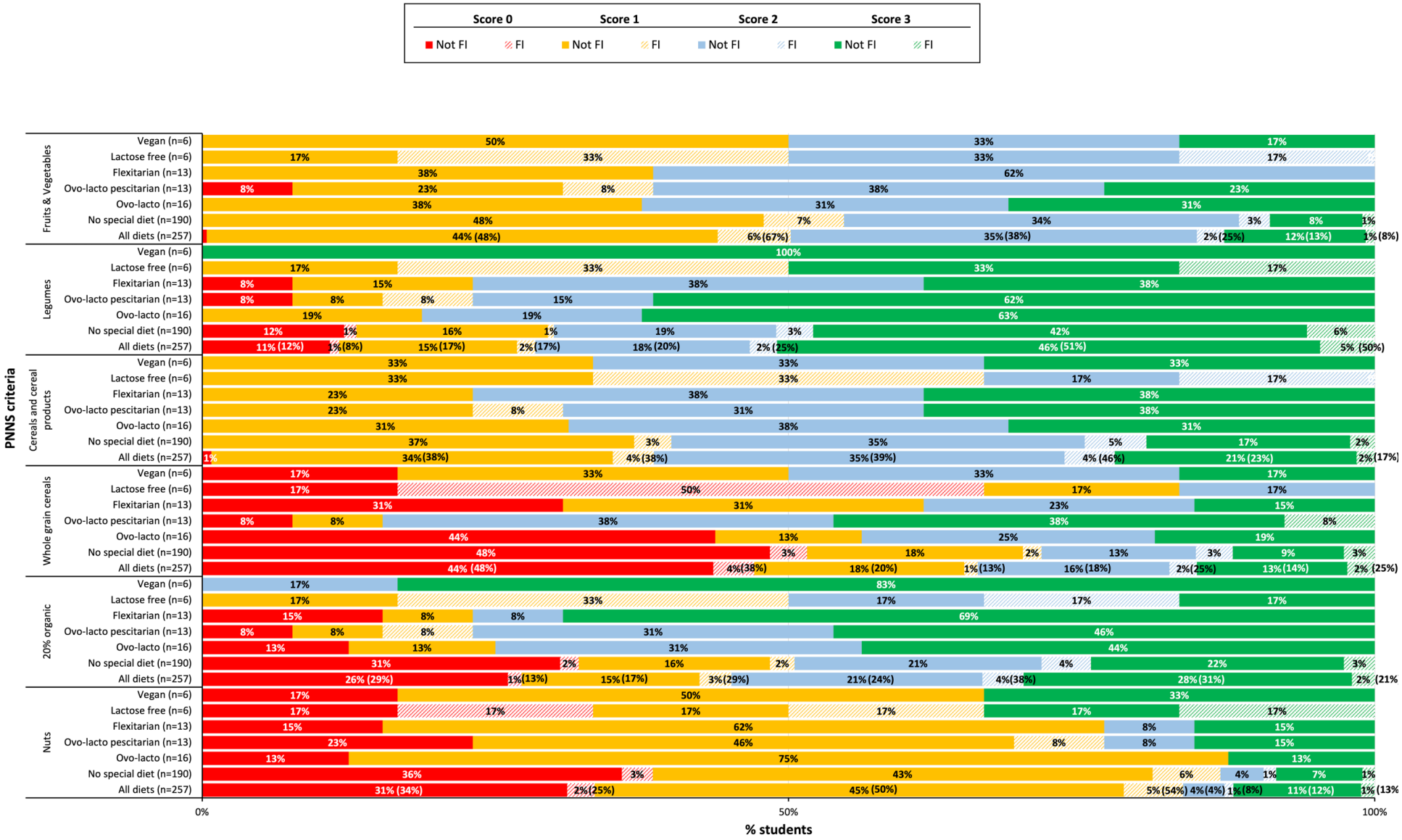
Moving for the nuts criterion, only 12% of the total population had a score of 3, with the groups V and LF being the most compliant (33% and 34% respectively).

In terms of milk and dairy products, 41% of the population adhered to this criterion with a high compliance observed in the OL (50%) and OLP (62%) groups. Zero compliance was observed for the V group, which abstains from consuming animal products.

In terms of meat, poultry, and eggs consumption, 38% of the total population was completely compliant to the criterion. As predicted, the non-vegetarian groups showed the highest compliance rates (NSD (44%) and LF (50%)), while the vegetarian groups (except for V) showed some compliance, primarily owing to their egg consumption.

Only 4% compliance was observed among the students in the fish and fish products criterion, with little compliance observed only in the NSD group (3%). Lastly, for the criteria related to the consumption of cold cuts, red meat and alcohol, the majority of the population was compliant (75%, 90%, 98% with scores of 3 respectively). Interestingly, none of the students were fully compliant with all 12 criteria. Yet only two students were able to meet all of the criteria with the exception of fish consumption criterion.

For an overall evaluation of compliance considering all scores (0, 1, 2, and 3), Figure A5-2(A) in the supplementary material illustrates the distribution of % PNNS compliance, calculated as explained in the methods, across the 12 PNNS criteria for the entire population and the different diet type groups. Additionally, Figure A5-2(B) represents the frequency of PNNS compliance for the entire population and individual diet type groups. Overall, this study found that the population exhibits a 62% compliance rate with the PNNS recommendations, with the OLP group displaying the highest probability of compliance at 74%. The frequency of compliance for the remaining groups ranged from 61% to 68% (Figure A5-2(B)).


Score 0
Score 1
Score 2
Score 3
Not FI
FI
PNNS criteria
Fruits & Vegetables
Legumes
Cereals and cereal products
Whole grain cereals
20% organic
Nuts
Vegan (n=6)
Lactose free (n=6)
Flexitarian (n=13)
Ovo-lacto pescitarian (n=13)
Ovo-lacto (n=16)
No special diet (n=190)
All diets (n=257)
0%
50%
100%
% students

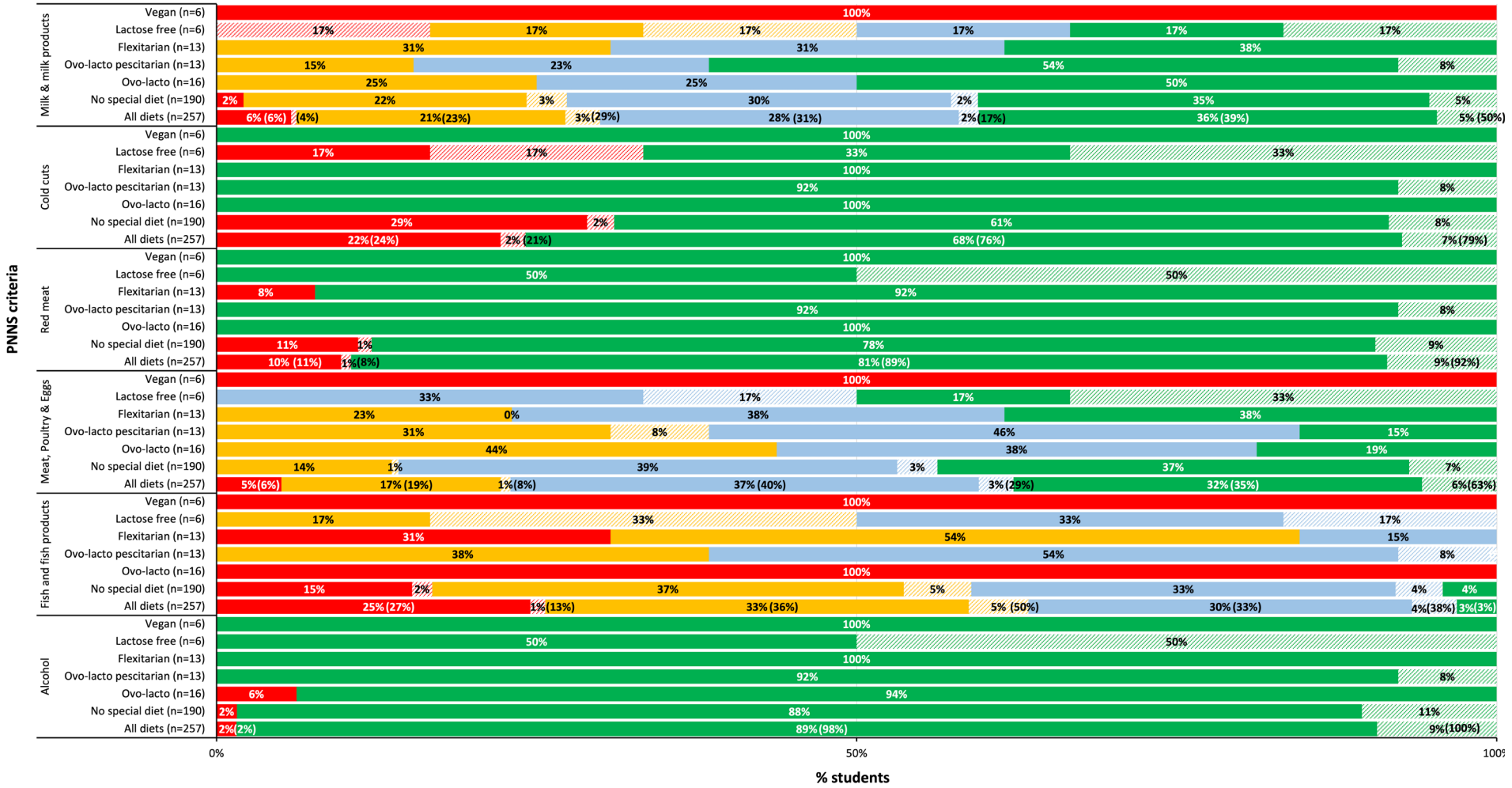


Figure 24. Distribution of students by PNNS scores for 12 criteria, stratified by diet type and food insecurity status: Percentage distribution of students by diet type (percentage students by food insecurity status) based on their compliance scores; score 3 (green) indicates 100% compliance, score 2 (blue) and 1 (yellow) indicate varying levels of partial compliance, and score 0 (red) indicate 0 compliance to the criterion

Food Insecurity and PNNS compliance

Concerning FI and adherence to the PNNS criteria, Figure 24 also illustrates the distribution of students according to their PNNS Scores, with the different diet types and the food insecurity status of students being highlighted.

As the percentage distribution of students by diet types is shown on the bars, the percentage of students by food insecurity status has been added in parentheses to enable a comparison between the "FI" and "not FI" groups. The comparison was conducted at the "all diets" level, without considering the diet types, especially because FI students were not present in all diet categories. Based on the 100% compliance (score 3), higher compliance was observed in the "not FI" group for the fruits and vegetables (13% versus 8%), the legumes (51% versus 50%), the cereals and cereal products (23% versus 17%), and the 20% organic food consumption (31% versus 21%) criteria. For the fish and fish products criterion, only 3% compliance was found in the "not FI" group, while the FI group showed zero compliance.

On the other hand, the FI group exhibited a higher compliance for criteria including whole grain cereals consumption (25% versus 14%), nuts (13% vs. 12%), milk & dairy products (50% versus 39%), cold cuts (79% versus 76%), meat, poultry, and eggs (63% versus 35%), red meat (92% versus 89%), and alcohol (100% versus 98%).

Moreover, a logistic regression was performed to investigate whether these differences were significant across all scores (0,1, 2 and 3, rather than just 100% compliance (score 3)) and to characterize the FI profiles in terms of PNNS compliance. Results revealed that, out of the 12 criteria tested, only five exhibited significant differences between the FI and not FI groups. These criteria are as follows: the fruits and vegetables, the whole grain cereals, the milk and dairy products, the meat, poultry and eggs, and the alcohol. Table 8 shows the detailed results for the five groups in focus.

Table 8. Results of the logistic regression for the association between FI status and the five significant food groups (groups with p-value < 0.16)

Variable	Odds Ratio [IC95%]	*p*-value
Fruits & Vegetables	0.32 [0.14, 0.65]	0.004
Whole grain	1.71 [1.14, 2.58]	0.010
Milk & dairy products	0.71 [0.44, 1.14]	0.159
Meat, poultry & eggs	1.92 [1.12, 3.43]	0.021
Alcohol	0.52 [0.33, 0.82]	0.005

As reported in Table 8, there is evidence to suggest that FI students exhibit different consumption patterns compared to not FI students. Specifically, FI students show a lower tendency (OR < 1) to consume fruits and vegetables as well as milk and milk products, and alcohol. Conversely, they are more likely to adhere (OR > 1) to the criteria for whole grain cereals and for meat, poultry, and eggs.

IV. Discussion

This study aimed to assess the dietary habits of students from the University of Grenoble Alpes by employing a PNNS score, which was constructed based on 12 PNNS guidelines. Additionally, the study aimed to investigate the influence of food insecurity on these habits.

The study involved 257 UGA students, all of whom had previously undergone food insecurity assessments as part of a previous study (Isoard-Gautheur et al., 2023). The average age of the participants was 21, with women constituting 72% of the sample. Most participants had a normal BMI (74%), and approximately half of the studied population had experienced noticeable fluctuations in their body weight since the beginning of the academic year. The majority didn't follow any specific diet type (190/274), while the rest opted various diet types. Only diet types that were followed by more than three people were studied individually (OL (16), OLP (13), F (13), LF (6) and V (6)) for the PNNS compliance while the others were only included with "all diets" group.

For the prevalence of FI, after regrouping the moderate and severe groups for a more meaningful comparison, 9% of the population was classified as having FI. Yet only 1.2% of our population experienced severe FI. Research on the prevalence of food insecurity in Europe, particularly among university students, remains limited. In a study done on Greek university

students, the rate of severely FI students was 38 times higher than that observed in this study (45.3%). One possible explanation for this disparity is the economic crisis that affected Greece. According to the Food and Agriculture Organization, approximately 17.2 ± 2.7% of the Greek population experienced moderate to severe food insecurity in 2014 (Cafiero et al., 2016). Additionally, the two studies used different assessment methods to measure food insecurity (Theodoridis et al., 2018). Furthermore, when compared to studies conducted in Western societies, the prevalence of food insecurity in this study is the lowest, as other studies have reported rates ranging from 14.8% to 58.8% (Berg & Raubenheimer, 2015; Bruening et al., 2016; Chaparro et al., 2009; Freudenberg et al., 2009; Gaines et al., 2014; Hughes et al., 2011; Micevski et al., 2013; Morris et al., 2016; Patton-López et al., 2014; Payne-Sturges et al., 2018).

Moreover, although relatively low, FI prevalence still revealed interesting characteristics of this group. FI individuals showed a higher percentage of weight variations compared to those who did not notice any changes. This proves that individuals experiencing food insecurity may be more susceptible to weight fluctuations, potentially due to the challenges they face in maintaining a stable and balanced diet.

Also, FI individuals demonstrated fewer variations and followed only two special diet types (LF and OLP). This finding implies that FI individuals may have limited access to a variety of food options or may be less likely to prioritize following a specific dietary pattern. This aligns with previous research that has associated food insecurity with a lack of dietary variation (Seligman et al., 2010). However, further research is necessary to better understand the underlying factors driving to the diet type choices.

As for the compliance with the PNNS, in general, regardless of the FI status, the overall compliance with the PNNS criteria across the entire population stood at 62%, which can be considered satisfactory. However, this level of compliance varied among different groups, with the OLP group exhibiting the highest compliance at 74%, while the NSD and the LF groups showed the lowest compliance at 61%. The evaluation of individual criteria explains the disparities between these groups, particularly as compliance levels differed across criteria and within various dietary categories.

Starting with the criteria to which the majority was compliant. All the groups were 100% adherent by majority (more than 50%) for the cold cuts, red meat, and alcohol criteria (75%, 90%, 98% with scores of 3 respectively for the total population).

As for the other criteria, the compliances varied significantly. For fruits and vegetables only 13% of the population were 100% compliant reflecting insufficient adherence within all group types although rates varied. This agrees with numerous studies suggesting that most college students consume inadequate amounts of fruits and vegetables (Althubaiti, 2022; Henley et al., 2023). For the legumes, half of the population were 100% compliant with 100% vegans being adhering to the criteria as legumes are an integral part in many diets including vegans as a main source of fibers and proteins (Polak et al., 2015). For cereal products, only 23% of the students adhered 100% to this criterion, with the population of NSD having the least compliance (19%). This can be explained by the fact that NSD individuals do not restrict themselves from animal proteins which provides satiety in people (Morell & Fiszman, 2017), while the other groups have to consume more carbs to feel full. For the 20% organic consumption, 30% of the total population were compliant with higher compliance rates in vegetarian groups (F, OL, OLP and V). This complies with multiple studies which have shown that consumers who have a strong preference for organic food tend to consume less meat or animal products than other consumers (Kilian & Hamm, 2021; Vigar et al., 2019). For nuts only 12% compliance in total population with higher compliance in LF and V groups. The consumption of plant-based milk is a common practice among LF and V groups, as they use it as a substitute for cow's milk. These milk alternatives are also called 'non-dairy alternatives' and are mainly composed of nut-based milk (Aydar et al., 2020; Sethi et al., 2016; Vanga & Raghavan, 2018). Also higher consumption of nuts among Vs, can be due to their use as a protein source (Mariotti & Gardner, 2019). For milk and dairy products also, 41% of total population adhered with vegetarian groups where milk is consumed (OL and OLP) having highest adherences. This can be due to the fact that these groups rely on milk and dairy products as a source of proteins (Vorvick, 2022). For meat, poultry, and eggs 38% of the population was compliant due the vegetarian groups that did not consume any animal products. For no special diet and lactose free groups, 44% and 50% had scores 3 respectively. And finally, only 4% of the population had score 3 for this criterion especially that the third point was added if the person considered 50% of their fish intake from fatty fish. Without considering the fatty fish criteria, compliance increased to 34% for the total population and it is highest for the OLP group which is predicted given that it is a main component of their diet. The insufficient knowledge among college students on the benefits and the risks of fish and fish product consumption can be one reason for the low compliance to the criteria. In a study done on college students from New Jersey, results suggested that risk managers must target young populations or information about the risks and benefits of consuming fish (Burger, 2005).

Research on the dietary habits of French college students is relatively scarce; however, one study conducted in 2021 in Rouen examined university students to assess changes in their diet quality before and during the COVID-19 period. This study evaluated the consumption of six components: fruits and vegetables, nuts, legumes, cereals, dairy, and fish, employing the PNNS-GS2 score. While a different scoring system was used, the same criteria were adapted for assessing fruits and vegetables, nuts, legumes, and dairy.

When comparing the prevalence data from this study to the prevalence data from the pre-COVID period, it was noted that this study showed greater compliance with the criteria for nuts, legumes, and dairy (12%, 51%, and 41% compliance, respectively) compared to the pre-COVID figures (10%, 21%, and 31%, respectively) (L. Miller et al., 2022). Additional research is necessary to gain a deeper understanding of the variations within the French student population.

Now regarding to the effect of FI on the adherence to the PNNS criteria, significant differences were found in four criteria between FI and not students. FI students demonstrated lower adherence to the fruits and vegetables ($p=0.004$), milk and milk products ($p=0.159$), as well as the alcohol ($p=0.005$) criteria. On the other hand, they showed higher adherence to the meat, poultry, and eggs ($p=0.01$), as well as the whole grain cereals ($p=0.02$) criteria. These results can be explained by the high cost of alcohol (Morrell et al., 2021; Xu & Chaloupka, 2011), fruits and vegetables. A survey conducted by IPSOS (Public Opinion Polling Institute Sector) in January 2021 revealed a decrease in fruit and vegetable consumption among the general French population, with one in two French individuals perceiving them as too expensive (IPSOS, 2022). These findings are also consistent with those of a study that examined the concurrent relationships between food insecurity and many health-related outcomes in a University of California student population, where FI was associated with fewer daily servings of fruits and vegetables , which in turn was associated with poor health (Martinez et al., 2019).

As for the meat, poultry, and eggs criterion, the FI students showed higher compliance which can be related to the fact that FI groups were not prevalent in diet types with meat consumption restrictions. Also, compliance was mainly due to egg consumption in FI individuals' group. Eggs are the least expensive among animal proteins. According to the CSA survey (Consumer Science & Analytics) to the CNPO (Organization of the National Committee for Egg Promotion), 71% of French people believe that eggs are a good economical alternative

to animal protein in a context of inflation and reduced meat and fish consumption (CNPO, 2021).

The relationship between diet quality and food insecurity has been well established in the literature. A study done in Greece showed that severe FI was negatively correlated with adherence to the Mediterranean diet (MD) among a university student sample, suggesting that individuals experiencing higher levels of food insecurity tended to opt for a less expensive but lower quality diet (Theodoridis et al., 2018).

To date, research regarding FI and students in the EU is still limited. And while this study demonstrated relatively low prevalence of FI among the student population studied, it is crucial to conduct further studies assessing food insecurity among university students due to their vulnerability and susceptibility to developing unhealthy eating habits during this transitional phase of life. These studies can offer valuable insights into the factors influencing eating habits in college students and help shape national policies on student financial assistance. Additionally, conducting larger-scale studies in France is necessary to gain a comprehensive understanding of the prevalence and impact of food insecurity on eating habits among university students.

The strength of this study lies in its novelty, as to our knowledge, no study has examined the relationship between food insecurity and the adherence to the PNNS guidelines among university students in France. However, limitations include a relatively small sample size and the use of non-probability sampling, which limits the representativeness of the findings to the broader French university student population. In addition, specific questions can be added to the dietary assessment questionnaire to assess compliance with other important PNNS criteria, such as those related to added fats, sweetened foods and beverages, salt, and physical activity. Not to forget the limitation related to the fact that this study was carried out during the pandemic period, a particular period that could have influenced the results: either exacerbating the food insecurity of students who couldn't work or reducing it for students who were able to benefit from the family environment.

In conclusion, this study gives an insight into the eating habits of the studied population of the UGA students vis à vis the French National Nutrition Program and characterizes the eating profiles of food insecure students versus students with no food insecurity. Findings can be useful to implement plans to improve the dietary habits and promote the overall well-being of college students. Further research is needed to broaden the tested food groups and better

understand the reasons behind food insecurity as well as other factors that might influence a healthy eating pattern in students.

Chapter 6 - Student's exposure profile and effect of organic food consumption on the exposure

The chapter is divided in two parts:

- The **first part** will include results on the evaluation of exposure of students. As a reminder, the questionnaire method was used in this work to assess the eating habits of the students then the exposure levels were calculated using contamination values from the French total diet study (ANSES, 2011).
- The **second part** consists of studying the potential impact of organic food on the exposure levels. A calculation of the relative exposure based on the proportion of organic matrices consumed was expressed for different pairs (chemical-food matrix). This showed how the exposure to different chemicals varied with increasing consumption of specific organic matrices. This approach was developed using the excess ratios (ER) calculated in chapter 4.

As such, this chapter is dedicated to achieving two main objectives. First, it aims to characterize the dietary exposure of UGA students to food contaminants. Second, it aims to assess the impact of specific organic food on exposure to chemical substances.

I. Methods

Questionnaire

The eating habits were collecting using a questionnaire. The participation procedure and questionnaire description were described in the previous chapter (Chapter 5). In summary, the questionnaire was sent by email to the UGA students and it assessed the consumption of 150 food items grouped into 22 food groups: meat, fish/shellfish, eggs, legumes, cereals (including bread), vegetables (including soups), potatoes, fruits, dairy products, soy-based deserts, vegetable oils, butter, nuts, snacks, pre-packed/pre-cooked meals, canned food, ready-to-eat sauces, confectionery, water, fruit/vegetables juices, wine/beer, and tea/coffee. The questionnaire was accompanied with a guide on portion sizes inspired by principle of the "Hand control portion" making it possible to estimate the weight of a portion using the hands (Gibson et al., 2016).

Every time a questionnaire was filled, the data were transferred from the questionnaire software (Sphinx) to an Excel spreadsheet where consumption was first expressed in g/week. For the food groups encompassing many elements (e.g., food group: Fruits, elements: cherries, strawberries, kiwis, etc.), the consumption was calculated using equation 1.

Equation 1: *Total consumption for food groups with different elements in g/week*

$$\text{Total consumption of food group X (g/week)} = \sum_{n=1}^{N} (\#\ portions_n * weight\_pp_n * frequency_n)$$

Where N is the total number of elements within the food group X, "n" is the index of the element, $\#\ portions_n$ *is the number of portions of the consumed element,* $weight\_pp_n$ *is the weight in grams per portion of the element, and* $frequency_n$ *is the frequency of consumption of the element.*

For fruits and vegetables, when the element was a seasonal product, the consumption of the element was divided by 12 if consumed 1 month per year, by 4 if consumed 3 months per year and by 2 if consumed 6 months per year. For example, if a student indicated having consumed 3 portions of cherries per week for one month/year, the consumption of cherries is

calculated by multiplying the number of portions with the weight_pp with the frequency which is 3/week then divided by 12 to correspond to one month of consumption.

Selection of chemical substances

A selection of 54 pollutants was made including 11 pesticides, 7 inorganic contaminants, 17 Dioxins and Furans (PCDD/F), 12 PCBs Dioxin-Like (PCB-DL), 6 Non-Dioxin Like PCBs (NDL-PCB) and 1 additive. These were selected according to 3 criteria: the pollutant is either classified as high-risk substances in the French TDS study (EAT2), or it has quantified values in the Rhone Alpes region (Region 6) of the database for the most consumed food products by the student's population, or it belong to substances authorized in organic production.

Calculation of exposure levels

Dietary exposure to each contaminant of interest was calculated for every student in the studied population, according to equation 2.

Equation 2: *Calculation of the dietary exposure of student to a substance S in µg/kg BW/day*

$$\text{Dietary exposure level to S (} \mu g/kg\ BW/day) = \sum_{n=1}^{N} \frac{C_{s,n} * I_{i,n}}{7 * BW_i}$$

Where N is the number of food elements included in the consumption, $C_{s,n}$ is the concentration (in µg/g) of chemical substance S in the food element "n", $I_{i,n}$ is the student consumption (in g/week) of the food element "n" by student "i", and BW_i (in kg) is the body weight of student "i". The number "7" account for the number of days in a week.

Contamination levels were retrieved from: ANSES EAT2 study (Etude Alimentation Totale). The contamination data are provided for each foodstuff, based on sampling carried out between 2007 and 2009. Data are provided at national or regional level. In this thesis, values from the most recent sampling were favored (2009), along with values corresponding to the study region, the Rhône-Alpes region (Region 6 in EAT), or, in the absence of such data, the national values were used (Region 99 in EAT).

In EAT2, food was purchased and prepared "as consumed", i.e., washed, peeled and cooked, then grouped into samples. For each analyzed substance, the concentration in the sample is provided. However, in some cases, substances were not quantified for two reasons:

- First, the chemical was not detected in the sample. In this case, two hypotheses are made: a lower bound (LB) hypothesis, where the contamination value is expected to be null (0) and an upper bound (UB) hypothesis, where the contamination value is considered the detection limit (LOD).
- Second, the chemical was detected but couldn't be quantified. In this case, the LOD was assigned as the LB hypothesis and the limit of quantification (LOQ) was assigned as the UB hypothesis.
- When the chemical was quantified for most of the foods, a middleboud (MB) hypothesis was set based on the quantified levels. And finally, when the measurement was not done, no data was reported.

Calculation of dioxins exposure

For dioxins (17 PCDD/F and 12 DL-PCBs), the contamination level of food is expressed as a unit of Toxic Equivalency (TEQ). For each food, a TEQ is calculated using WHO 2005 Toxic Equivalent factors (TEFs) corresponding to dioxin congener according to equation 3.

Equation 3: *Calculation of the Toxic Equivalency unit (TEQ) for every food element*

$$For\ every\ food:$$

$$TEQ = \sum_i (PCDDj * TEFj) + \sum_i (PCDFj * TEFj) + \sum_i PCBj * TEFj)$$

TEQ is the level of contamination of the food in TCDD equivalent expressed in pg WHO2005 TEQ /g fresh weight (FW) of food, while $PCDD_j$ represents the concentration (in pg/g FW) of the congener $PCDD_j$, and TEF_j is the weighing coefficient assigned to congener j.

The dietary exposure is then calculated according to equation 2 using the TEQ values as the contamination level to dioxins ($C_{s,n}$ in Equation 2).

Calculation of methylmercury exposure

In assessing dietary mercury exposure, EAT2 presented contamination values for total mercury (Hg). However, it is crucial to differentiate between exposure to methylmercury (organic mercury) and inorganic mercury, as they entail distinct toxicological effects, each governed by a unique TRV (JECFA, 2010). Consequently, given that almost all dietary exposure to methylmercury comes from seafood, exposure to methylmercury was estimated based on mercury exposure through fish, mollusks, and crustaceans, as recommended by JECFA. Moreover, the Calipso study confirms the assumption that 100% of mercury is in the form of methylmercury in seafood (Sirot et al., 2008). Furthermore, the exposure to inorganic mercury was estimated based on exposure to mercury from all other food groups, excluding seafood. However, in the results we only focused on the exposure to methylmercury as it has been classified as a high-risk substance in EAT2.

Calculation of inorganic arsenic exposure

As for the exposure to arsenic, only total arsenic was expressed in the EAT2 study. Yet, the TRV defined for total arsenic is no longer considered in risk assessments conducted at the international level, as the risk assessment should focus on inorganic arsenic due to its potential toxic effects (ANSES, 2011b). Hence, the calculation of exposure to inorganic arsenic involved deriving it from the total arsenic exposure. This was achieved by applying hypothesis regarding the speciation of inorganic arsenic. The specific proportion of inorganic arsenic was determined using data obtained from the CALIPSO and Yost et al study (Sirot et al., 2009; Yost et al. 2004).

Risk assessment

The risk assessment was conducted in the following manner:

- For substances with threshold effects: The health risk for the population was evaluated by comparing exposure values to TRVs. The TRVs selected for our study were those used in EAT2.
- In cases where some substances had more recent TRV values, assessments were also conducted relative to these updated values.
- For substances without threshold effects: Margin of exposures (MOEs) for both the mean exposure and the 95th percentile value were calculated using Equation 4.

Equation 4: *Calculation of the Margin of Exposure (MOE)*

$$MOE = \frac{BMDLx}{exposure\ measurement}$$

The $BMDL_x$ (Benchmark Dose Lower Confidence Limit) represents the dose level at which the observed change in response is expected to be less than x%, with the term "likely" determined by the statistically credible level, typically set at the 95% confidence level (EFSA, More, et al., 2022). X is either 1, 5 or 10.

Based on the $BMDL_{10}$, EFSA suggested a method to evaluate health risks through the MOE: “an MOE of 10,000 or higher is considered of low concern from a public health perspective and may be deemed a low priority for risk management actions and vice versa” (EFSA, 2005b). However, EFSA's guidelines lack clarity regarding the appropriate MOE when $BMDL_{01}$ or $BMDL_{05}$ values from animal studies or BMDLs from human studies are available. In this study, $BMDL_{01}$ values were determined for lead and arsenic. For the evaluation, the same methods applied in the EFSA reports for lead and arsenic in the scientific opinion reports, were applied in this chapter as well. These were also the method applied in EAT2 (ANSES, 2011b). For lead, an MOE greater than 10 is sufficient to ensure that there is no risk of prevalence of cardiovascular or nephrotic effects (EFSA Journal, 2010). As for arsenic, no specific reference point was identified for MOEs; instead, exposure levels were compared to the $BMDL_{01}$ range (0.3-8 µg/kg bw/day). If exposure values fell within this range, indicating a small to no MOE, this implied that the possibility of excluding the risk was unlikely (EFSA Journal, 2009).

The impact of organic food consumption on the dietary exposure

The consumption of organic food was assessed in the questionnaire through 15 questions. Respondents had to specify the proportion in percentage of organic food consumed for 15 food categories (e.g., how much of your fruits consumption is dedicated to organic fruit). Consequently, the organic food consumption profile of students was characterized by food groups rather than individual food elements.

In the subsequent analysis, ER (excess ratio) values generated in Chapter 4 were used to calculate the relative exposure, R, as a function of the proportion of organic food consumed, ranging from 0% to 100% organic, in order to determine the impact of organic food consumption on exposure to food contaminants. As a reminder, ER values were generated for pairs (chemical-food matrix) having contamination values from both conventional and organic productions.

The calculation of R was achieved by a formula that was developed according to the following reasoning.

First, the dietary exposure to a chemical contaminant was expressed as follows:

$$E = C * C_{conv} + B * C_{org}$$

Where E is the dietary exposure to a chemical, C and B are the intakes of conventional and organic food, respectively, and C_{conv} and C_{org} are the concentration of the chemical under consideration in the conventional and the organic food, respectively.

Taking φ as the proportion of organic in from the total consumption, T, the B and C can be expressed as follows:

$$B = \varphi T \; and \; C = (1 - \varphi)T$$

Moreover, by defining q as the ratio of conventional to organic concentration, then q can be expressed in function of ER as:

$$q = \frac{C_{Org}}{C_{Conv}} = \frac{1 + ER}{1 - ER}$$

As a result, E can be expressed as follow:

$$E = C * C_{conv} + B * C_{org} = T * C_{conv} * [q\varphi + (1 - \varphi)]$$

where $E_{conv} = T*C_{conv}$, represents the exposure corresponding to consumption of conventional foods only.

Finally, the relative exposure compared to the conventional exposure was expressed according to equation 5.

Equation 5: *Relative exposure in function of the percentage of organic matrix consumption*

$$R = 100 * \left(\frac{E - E_{conv}}{E_{conv}}\right) = 100 * \left\{\left[\frac{1 + (2\varphi - 1)ER}{1 - ER}\right] - 1\right\}$$

This formula portrays the relative exposure linearly based on the proportion of organic matrices consumed non-linearly based on the ER for a given pair (chemical-food matrix)

Data Analysis

Concerning statistical analysis, consumption, and exposure calculations, along with the generation of graphical representations were carried out using Excel and R software.

II. Results

1. Food intake profile

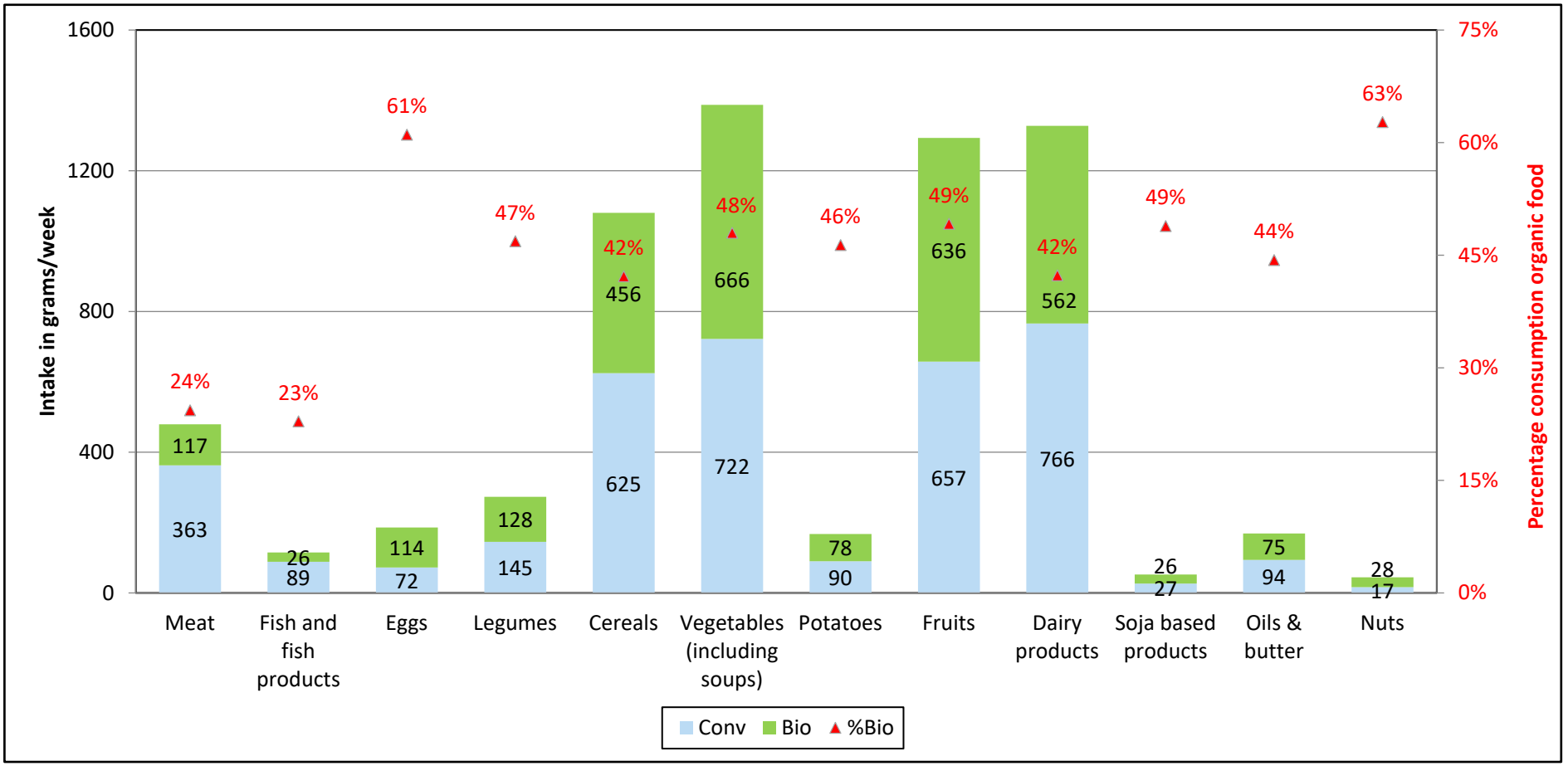


Figure 25. Mean weekly student consumption of main food groups with emphasis on the proportion of organic alternatives, excluding beverages

Figure 25 shows the average consumption of students for 12 main food groups in g/week, highlighting the share of organic products consumed for every food group, excluding beverages. The food groups that were mostly consumed by students were vegetables (including soups) (1388g/week), dairy products (1328g/week), fruits (1293g/week), and cereal products (1080 g/week).

The mean consumption of organic products for all food groups was 45% (data not shown). Organic food consumption was mostly prevalent for nuts (63%) and eggs (61%).

2. Dietary exposure levels

Table 9. Results of exposure to the selected chemicals

Chemicals	TRV (µg/kg bw /day)	TRV Source	LB hypothesis					UB hypothesis				
			UGA STUDENTS					UGA STUDENTS				
			Mean	p95	%> TRV	[IC95%]	Main contributor	Mean	p95	%>TRV	[IC95%]	Main contributor
Dimethoate	1	(EFSA, 2006)	0,0	0,1	0%	-	Fruits (99%) cherries ++& vegetables (1%)	0,8	1,5	**27%**	[22; 33]	Tea/ coffee (31%), Vegetables (18%) & Fruits (14%), wine & beer (7%)
Deltamethrin	10	(EFSA, 2009c)	0,0	0,0	0%	-	-	0,2	0,4	0%	-	Vegetables (29%), tea/coffee (16%)
Pyrethrins	40	(EFSA, 2013a)	0,0	0,0	0%	-	-	0,0	0,0	0%	-	Cereals avec Bread (82%) including Pasta/semolina (43%), vegetables (16%)
Chlorothalonil	15	(EFSA, 2018)	0,0	0,0	0%	-	Vegetables (100%)	0,1	0,2	0%	-	Vegetables (23%) Tea/ coffee (21%)

Chemicals	TRV (µg /kg bw /day)	TRV Source	LB hypothesis					UB hypothesis				
			UGA STUDENTS					UGA STUDENTS				
			Mean	p95	% >TRV	[IC95%]	Main contributor	Mean	p95	% >TRV	[IC95%]	Main contributor
Hexachlorobenzene	0,8	(Arnold et al., 1985)	0,0	0,0	0%	-	Meat (100%)	0,1	0,1	0%	-	Tea/coffee (25%), vegetables (14%)
	0,07	**(ATSDR, 2015)**			0%	-				0%	-	
Chlorpyriphos-ethyl	10	(European Comission, 2005a)	0,0	0,0	0%	-	Fruits (99,9%)	0,1	0,2	0%	-	Vegetables (39%), fruits (18%)
	1	**(EFSA, 2014a)**			0%	-				0%	-	
Chlorpyriphos-methyl	10	(European Comission, 2005b)	0,0	0,0	0%	-	Cereals (70%) with Bread (100%), fruits (30,4%)	0,1	0,2	0%	-	Vegetables (41%) including Tomatoes et carrots (33% et 28%) & Tea/coffee (16%)

Chemicals	TRV or BMDL (µg/kg bw/day)	TRV Source	LB hypothesis					UB hypothesis				
			UGA STUDENTS					UGA STUDENTS				
			Mean	p95	%>TRV or MOE= p95-Moy	[IC95%]	Main contributor	Mean	p95	%>TRV or MOE= p95-Moy	[IC95%]	Main contributor
Ethion	2	(WHO-JMPR, 1990)	0,0	0,0	0%	-	Meat (100%) Cold cuts	0,1	0,1	0%	-	Tea/coffee (23%) Cereals (14,4%) Vegetables (14,2%)
Pirimiphos-methyl	4	(EFSA, 2005a)	0,1	0,1	0%	-	Cereals (74%) including Breads (56%)	0,2	0,3	0%	-	Cereals (35%) & Vegetables (24%)
Bifenthrin	15	(EFSA, 2011)	0,0	0,0	0%	-	Fruits (100%) including apples (86%)	0,1	0,3	0%	-	Vegetables (37%) Tea/Coffe (14%)
λ-cyhalothrin	5	(European Comission, 2001)	0,0	0,0	0%	-	Fruits (100%) apples (75%)	0,1	0,3	0%	-	Vegetables (18%) Tea/Coffee (13%), Fruits (13%)
	2,5	**(EFSA, 2014b)**			0%	-				0%	-	
Inorganic Arsenic	0,3 - 8	(EFSA Journal, 2009)	0,1	0,5	**MOE0,3= 0,6 - 2,1 MOE8= 16 - 57**		Water (34%) & dairy products (29%)	0,5	1,0	**MOE0,3= 0,3- 0,6 MOE8= 8,3 -16,5**	-	Water (39%)

Chemicals	TRV (µg/kg bw/day)	TRV Source	LB hypothesis					UB hypothesis				
			UGA STUDENTS					UGA STUDENTS				
			Mean	p95	%>TRV	[IC95%]	Main contributor	Mean	p95	%>TRV	[IC95%]	Main contributor
Copper	**150**	**(EFSA et al., 2018)**	24,5	46,9	0%	-	Cereales (17.08%) et Tea/ coffee (21%)	24,5	46,9	0%	-	Cereales (17%) et Tea/coffee (21%)
Methylmercury	0,23	(JECFA, 2004)	0,0	0,1	0%	-	Fish and fish products (100%)	0,0	0,1	0%	-	Fish and fish products (100%)
Sulfites	700	(SCF, 1996)	0,0	0,1	0%	-	Wine/beer (82%) fisha nd fish products (18%)	0,0	0,1	0%	-	Wine/beer (75%), Fish and fish products (16%) meat (2%)

Chemicals	TRV or BMDL	TRV Source	Middlebound (MB) hypothesis				
			UGA STUDENTS				
			Mean	P95	%>TRV or MOE =p95- Mean	[IC95%]	Main contributor
Cadmium	2,5 µg/kg bw/day	(EFSA, 2009a)	0,1	0,2	0%	-	Vegetables (24%), cereal products (12%), potatoes (9%)
	0,357 µg/kg bw/day	**(ANSES, 2019)**			**1,2%**	[0,2; 2,6]	
Lead	0,5 µg/kg bw/day (Neurotoxic)	(EFSA Journal, 2010)	0,2	0,4	-	-	Vegetables (24%), water (14%)
	0,63 µg/kg bw/day (Nephrotoxic)				**MOE1,5=3,5 - 6,2**		
	1,5 µg/kg bw/day (Cardiovascular)				**MOE0,63= 1,5 - 2,6**		
Nickel	22 µg/kg bw/day	(WHO, 2005)	3,1	5,4	0%	-	Water (36%)
	13 µg/kg bw/day	**(EFSA, 2020)**					
Aluminum	143 µg/kg bw/day	(JECFA, 2006)	43,9	73,6	0%	-	Water (24,4%), vegetables (13,8%), cereals (11%)
PCDD/F & PCB-DL	2,33 pg TEQ WHO_{98} / kg bw/day	(JECFA, 2001)	0,4	0,9	0%	-	Fish and fish products (33%) Dairy products (27%), butter (11%)
	0,29 pg TEQ WHO_{05}/kg bw/day	**(EFSA CONTAM Panel et al., 2018)**			**71%**	[65; 77]	
NDL-PCB	10 ng/kg bw/day	(Afssa, 2007)	2,4	5,9	0%	-	Fish and fish products (56%)

***TRVs that are in bold represent the most recent TRV values for the substance**
For lead, an MOE greater than 10 → no risk (EFSA Journal, 2010)
For arsenic, no specific reference point for MOEs; exposure levels were compared to the $BMDL_{01}$ range, if within range -> risk (EFSA Journal, 2009)

The average dietary exposure as well as the 95 percentile (p95) and the percentage exceedance of the TRV were calculated for each substance studied. As mentioned earlier, the risk assessment was initially based on the TRV values used in the EAT to facilitate comparison of results for the two populations (General French population versus the students' population). Furthermore, for substances with more recent TRV values, a different assessment was made in respect to these values. Results are detailed in Table 9.

Substances reflecting a level of concern based on the TRVs used in the EAT study were dimethoate, inorganic arsenic, and lead.

For **dimethoate**, 27% (%IC_{95}=[22; 33]) exceedance of the TRV (1 µg/kg bw/day (EFSA, 2006)) was observed only for the UB hypothesis. The major food groups contributors for the UB hypothesis are tea and coffee (31%), vegetables (18%), fruits (14%) and wine & beer (7%).

For **Inorganic arsenic**, based on EFSA's $BMDL_{01}$ of 0.3 µg/kg bw/day (EFSA Journal, 2009), for UB and LB hypothesis, MOEs was 0.6 and 2.1 for the average exposure levels respectively and 0.3 and 0.6 for the 95th percentile respectively (Table 9). As for the $BMDL_{01}$ of 8 µg/kg bw/d (EFSA Journal, 2009), MOEs for UB and LB hypothesis were 8.3 and 16 for the average exposure and 16.5 and 57 for the p95.

Given that some estimated exposure levels are within the $BMDL_{01}$ range (0.3 to 8 µg/kg b.w./ day), implying little or no MOE, the possibility of a risk from Asi cannot be excluded.

The major contributors based on the LB hypothesis are water (34%) and dairy products (29%), while for the UB the major contributors are water (39%) and canned food (11%).

Furthermore, for **lead**, the calculated MOEs based on the $BMDL_{10}$=0.63 µg/kg bw/day (EFSA Journal, 2010) was 3.5 and 6.2 for the p95 and the average exposure respectively, as for those calculated based on $BMDL_{01}$=1.5 µg/kg bw/day (EFSA Journal, 2010), MOEs were respectively 1.5 and 2.6. However, given the magnitude of the MOEs (<10), the possibility of developing health effects due to exposure to lead should not be excluded. The major food groups contributors were water (14%) and vegetables (24%).

On another hand, although **cadmium**, and **dioxins** did not exceed the threshold values used in the EAT study, reevaluation based on more recent TRVs (0.357 µg/kg bw/day (ANSES, 2019) and 0,29 pg TEQ/kg bw/day (EFSA CONTAM Panel et al., 2018)) revealed exceedance of 1,2% (%IC_{95} =[0,2 ; 2,6]) and 71% (%IC_{95}= [65; 77]) with respect to the TRVs respectively.

The major food group contributor for cadmium were vegetables (26%), cereal products (12%) and potatoes (9%), while for dioxins, the major food groups contributing the exposure are fish and fish products (33%) dairy products (27%), butter (11%).

3. Impact of organic food consumption

The calculation of relative exposure according to the proportion of organic consumption was carried out for all substances that were selected previously when they had an ER with a matrix we assessed in the questionnaire (Chapter 4). As a result, in total, the relative exposure was calculated for 53 pairs (chemical-food matrix).

In this chapter, only chemicals that reflected some levels of concern from the previous exposure part (dimethoate, Cd, As_i, Pb, and dioxins) and substances authorized in organic production for which we had ER values (Cu), will be discussed (33 pairs). The results are plotted in figures 26 to 35. More figures for the rest of chemicals can be found in the appendix A6.

In the figures, relative exposure is expressed in percentage (left-axis) and represents the variation of the exposure to the substance based on the proportion of the organic matrix consumed. The histograms in the figures (corresponding the right-axis) show the distribution of students according to the percentage of organic food group consumed. Furthermore, indicated in the figures are the percentage contribution of each food matrix, and in brackets the percentage contribution of the food group to which the matrix belongs) to the exposure to the studied chemical, as well as the number of samples from the available data in the literature for conventional and organic matrices (n conventional/n organic).

For **dimethoate**, the consumption organic cherries, peas, and olive oil would lead to a reduction in exposure (red axis), with a 100% organic consumption of these matrices reducing the exposure to dimethoate by 71%, 52%, and 77%, respectively (Figure 26).

Concerning **arsenic**, a consumption of 100% organic bovine meat and wheat flour would reduce exposure to this substance by 31% and 56%, respectively. However, 100% organic poultry meat would increase exposure by 5%, and organic carrots would have an no effect on exposure compared to conventional carrot consumption (Figure 27).

As for **cadmium**, organic consumption of apples and dairy products, showed no impact on the exposure level to this substance compared to conventional produce, while for carrots,

tomatoes, spinach and bovine meat, a consumption of 100% of these matrices in organic will increase the exposure to cadmium by 78%, 18%, 117%, and 83%, respectively. Meanwhile, a 100% organic consumption of lettuce, potatoes, wheat, wheat flour, and poultry would reduce exposure to cadmium by 26%, 24%, 15%, 34%, and 43%, respectively (Figures 28,29,30).

Moreover, for **lead**, for bovine meat, lettuce, potatoes and wheat flour, a consumption of 100% organic of these matrices would reduce the exposure to lead by 37%, 59%, 54% and 50% respectively, while a consumption of 100% of poultry meat, carrots, spinach, and wheat would increase the exposure to lead by 132%, 439% (5.4 times), 118% and 51% respectively. For dairy products, organic consumption has no effect on the exposure to lead compared to consumption of conventional dairy products (Figures 31-32-33).

For **dioxins**, data in organic meat indicated that for both bovine meat and poultry, a consumption of these organic matrices will increase the exposure to dioxins (by 108% and 300% respectively for DL-PCB and by 49% and 203% for poultry) (Figure 34).

Similarly, for **copper**, available data indicates an increase in exposure to Cu when 100% organic consumption is applied to bovine and poultry meat (10% and 1% increase, respectively) (Figure 35).

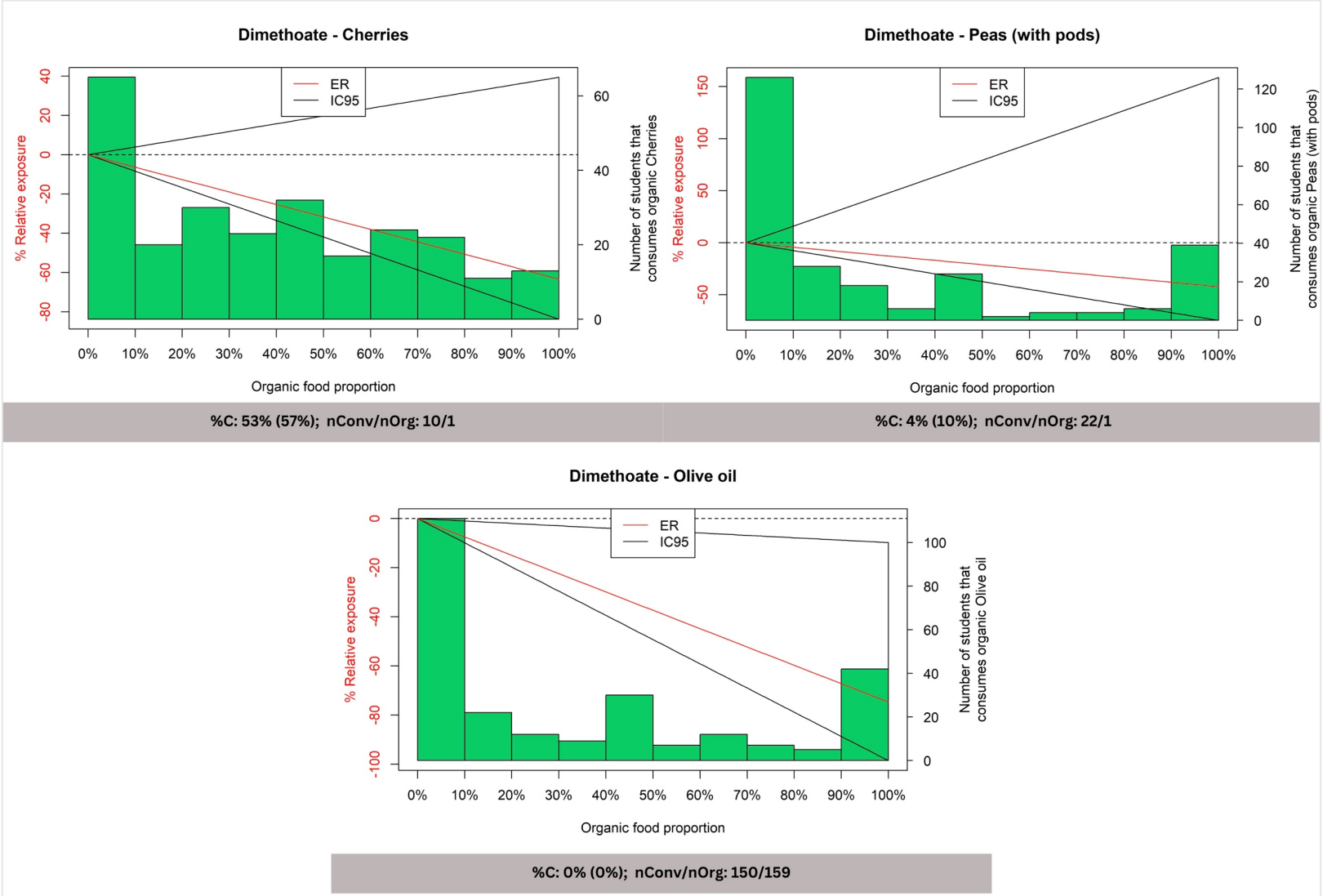


Figure 26. Relative exposure to dimethoate according to the proportion of organic food consumed for different foods; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the mean percentage contribution of the specific matrix to the exposure of the chemical substance (mean percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

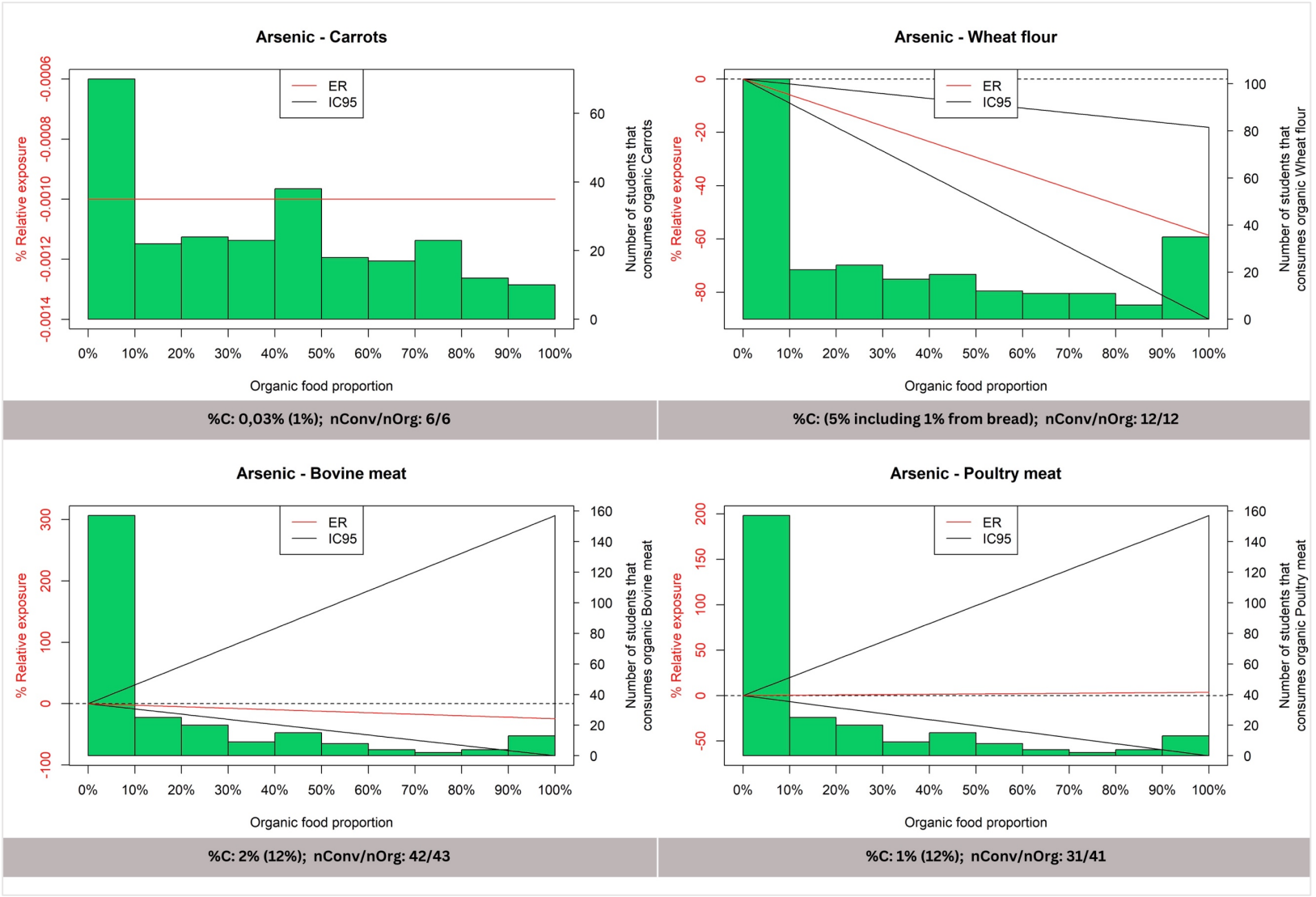


Figure 27. Relative exposure to arsenic according to the proportion of organic food consumed for different foods; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y- axis) is represented by the histograms; %C: the mean percentage contribution of the specific matrix to the exposure of the chemical substance (mean percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

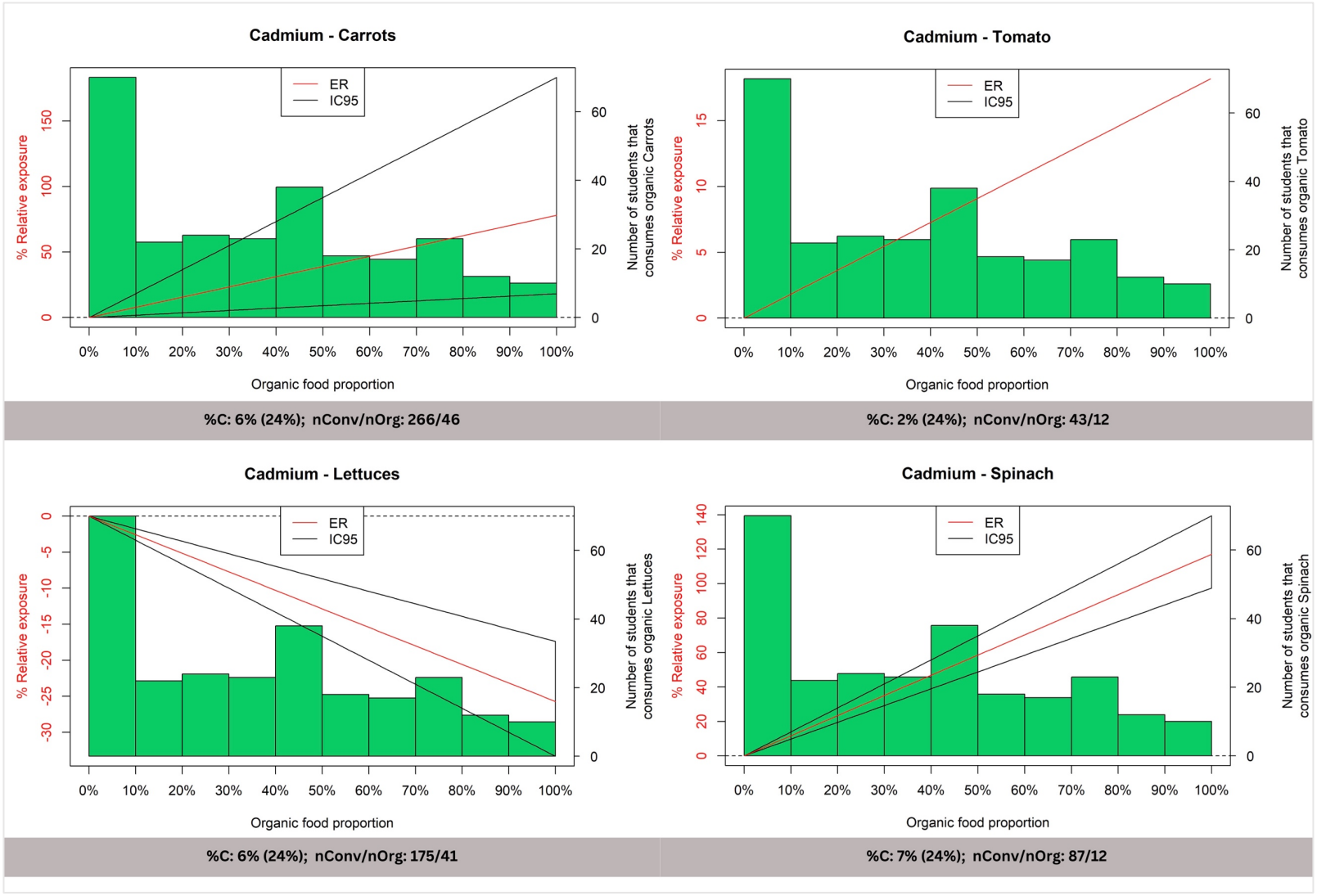


Figure 28. Relative exposure to cadmium according to the proportion of organic food consumed for different vegetable matrices; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the mean percentage contribution of the specific matrix to the exposure of the chemical substance (mean percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

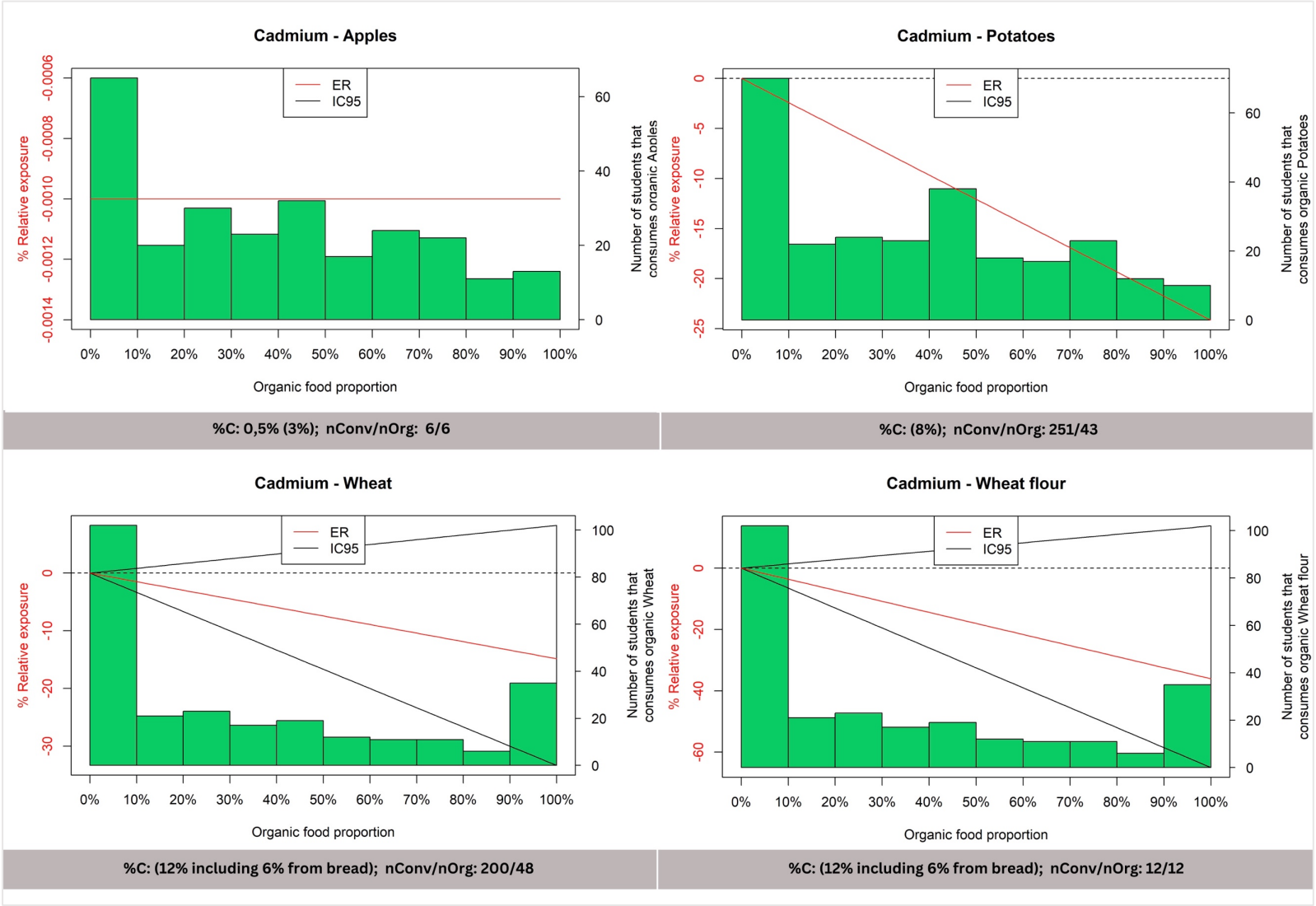


Figure 29. Relative exposure to cadmium according to the proportion of organic food consumed for different matrices; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

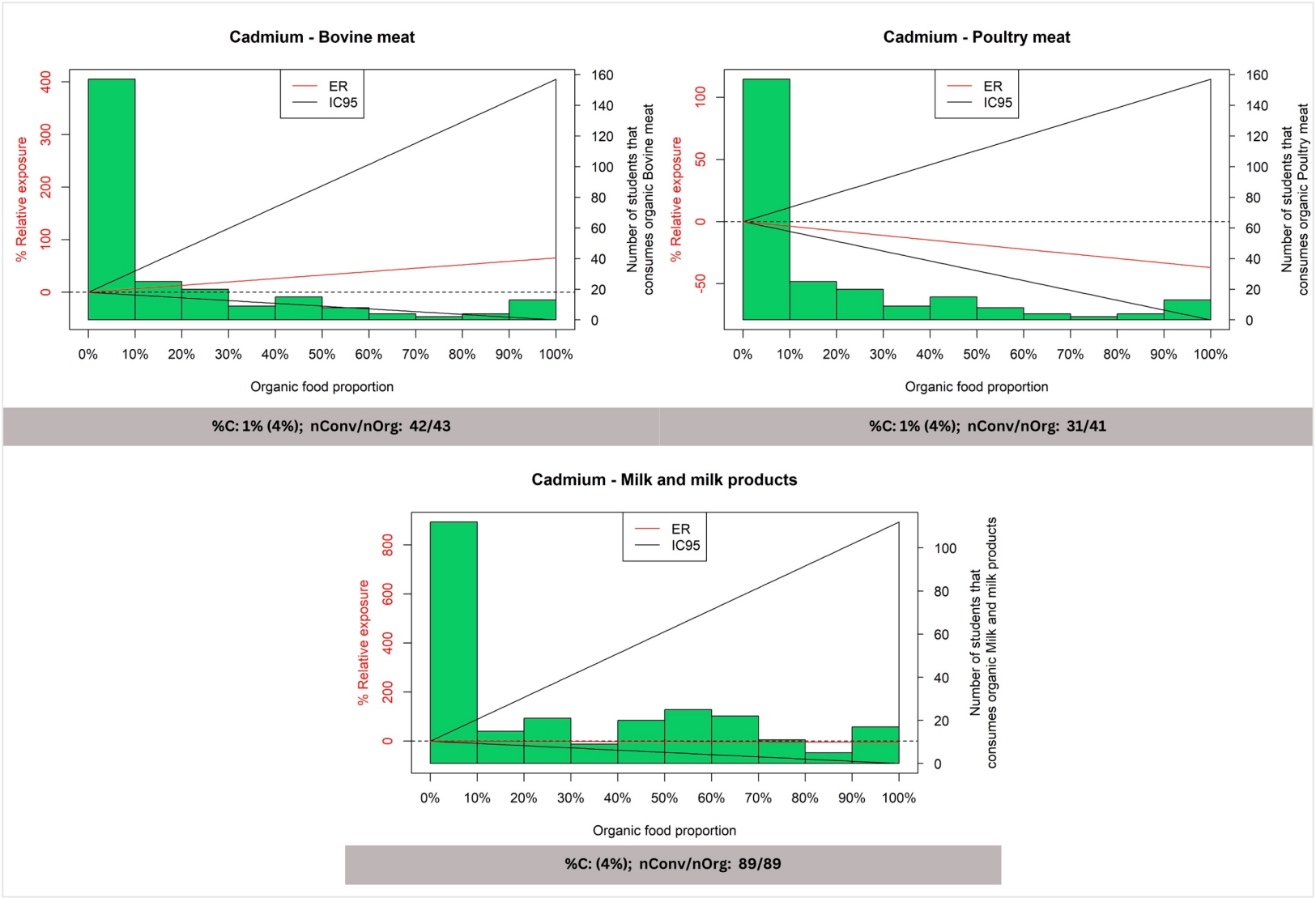


Figure 30. Relative exposure to cadmium according to the proportion of organic food consumed for meat and dairy products; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

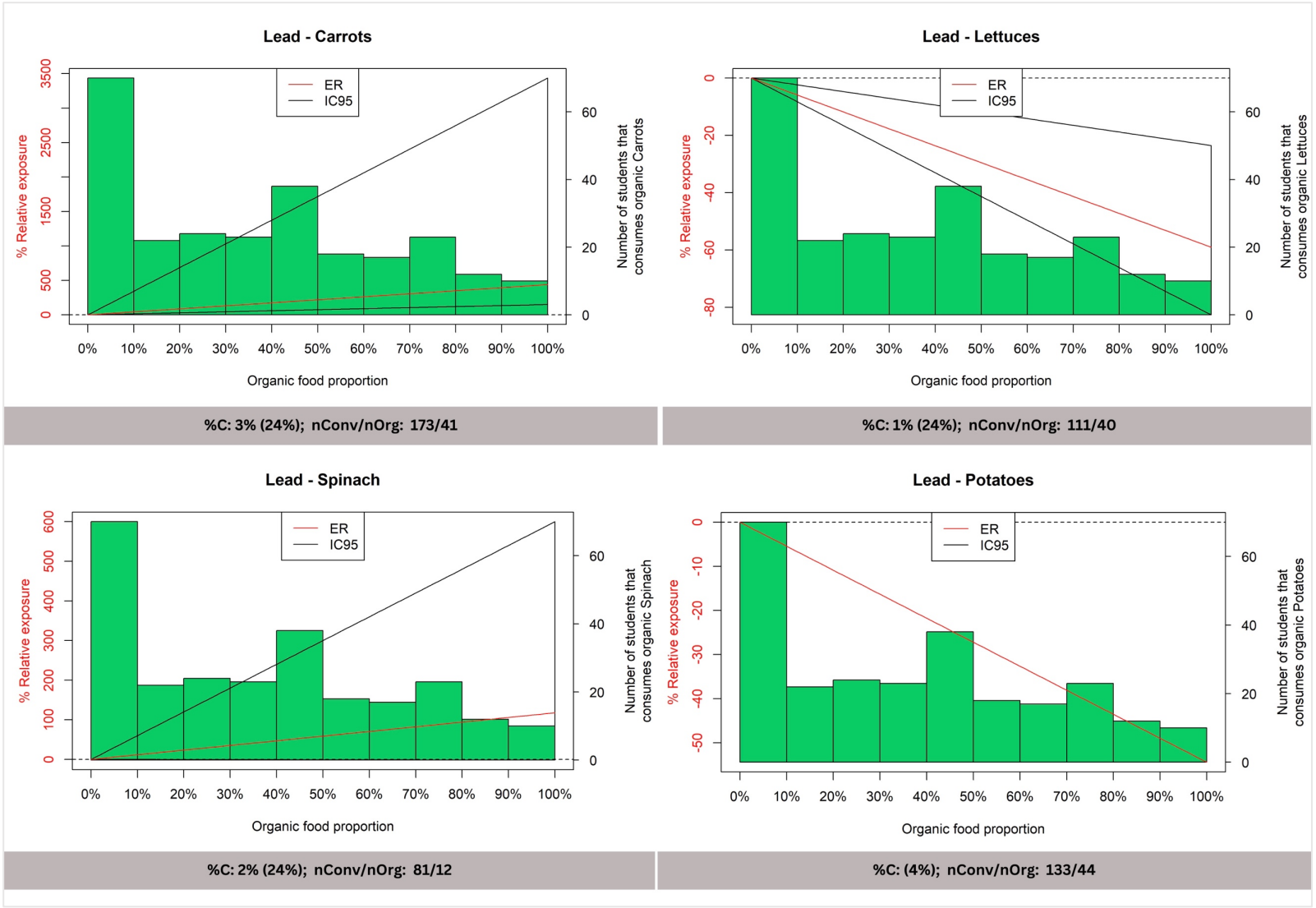


Figure 31. Relative exposure to lead according to the proportion of organic food consumed for vegetables and potatoes; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

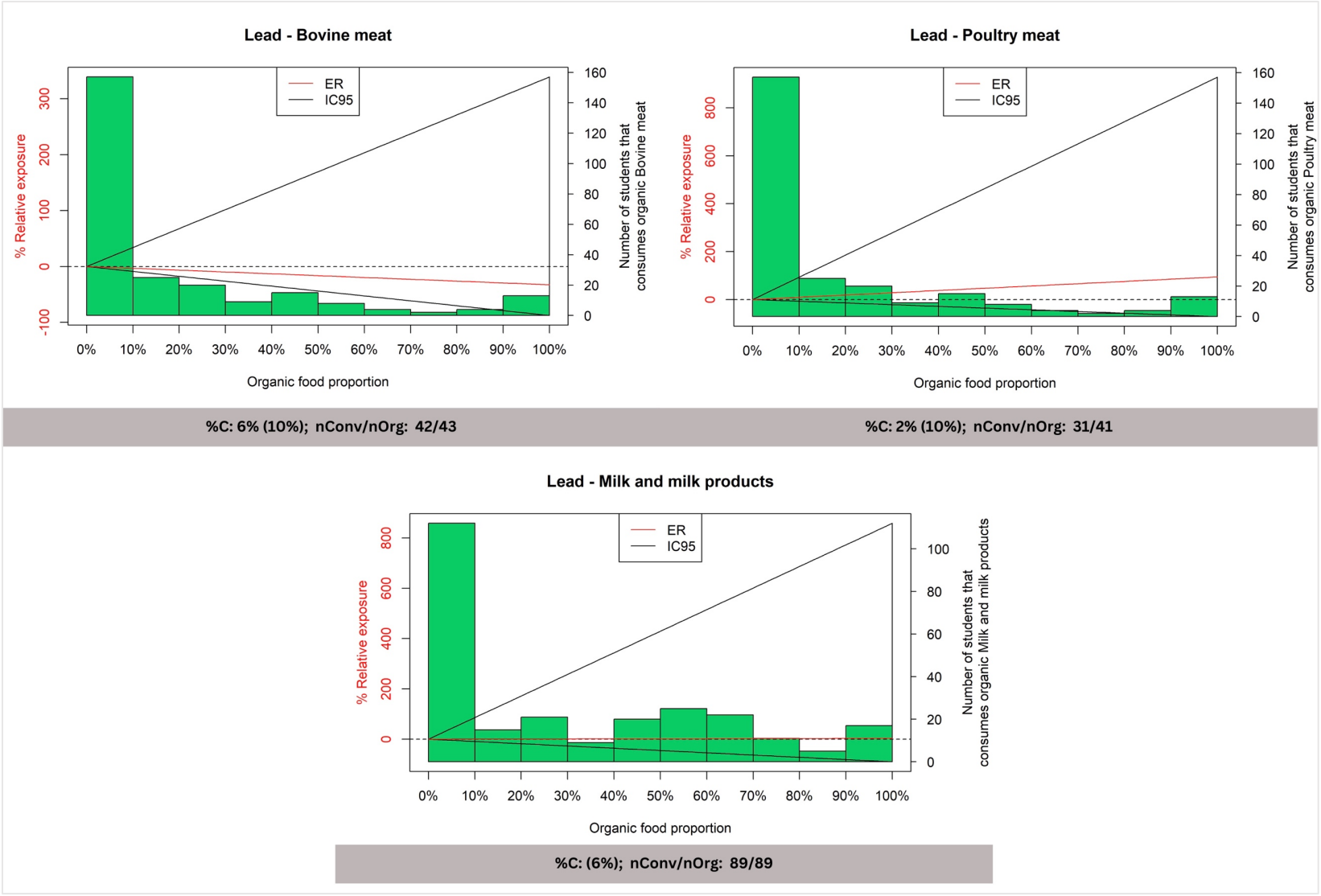


Figure 32. Relative exposure to lead according to the proportion of organic food consumed for meat and milk and dairy products; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

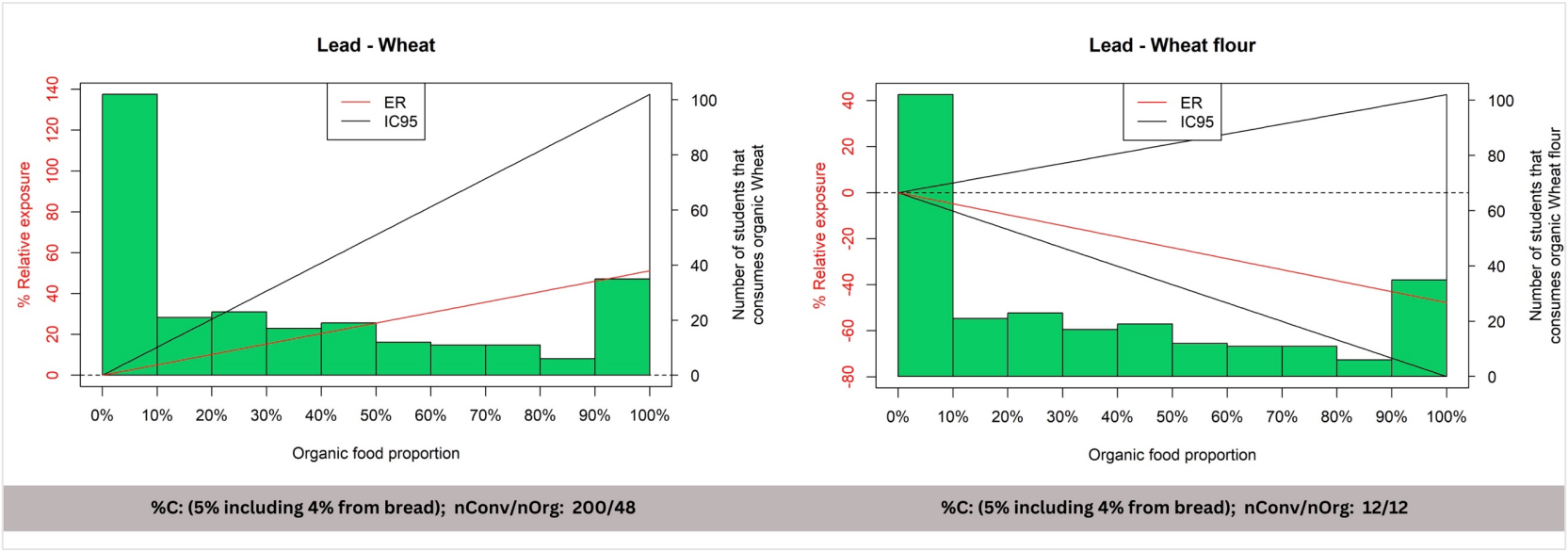


Figure 33. Relative exposure to lead according to the proportion of organic food consumed for cereals; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

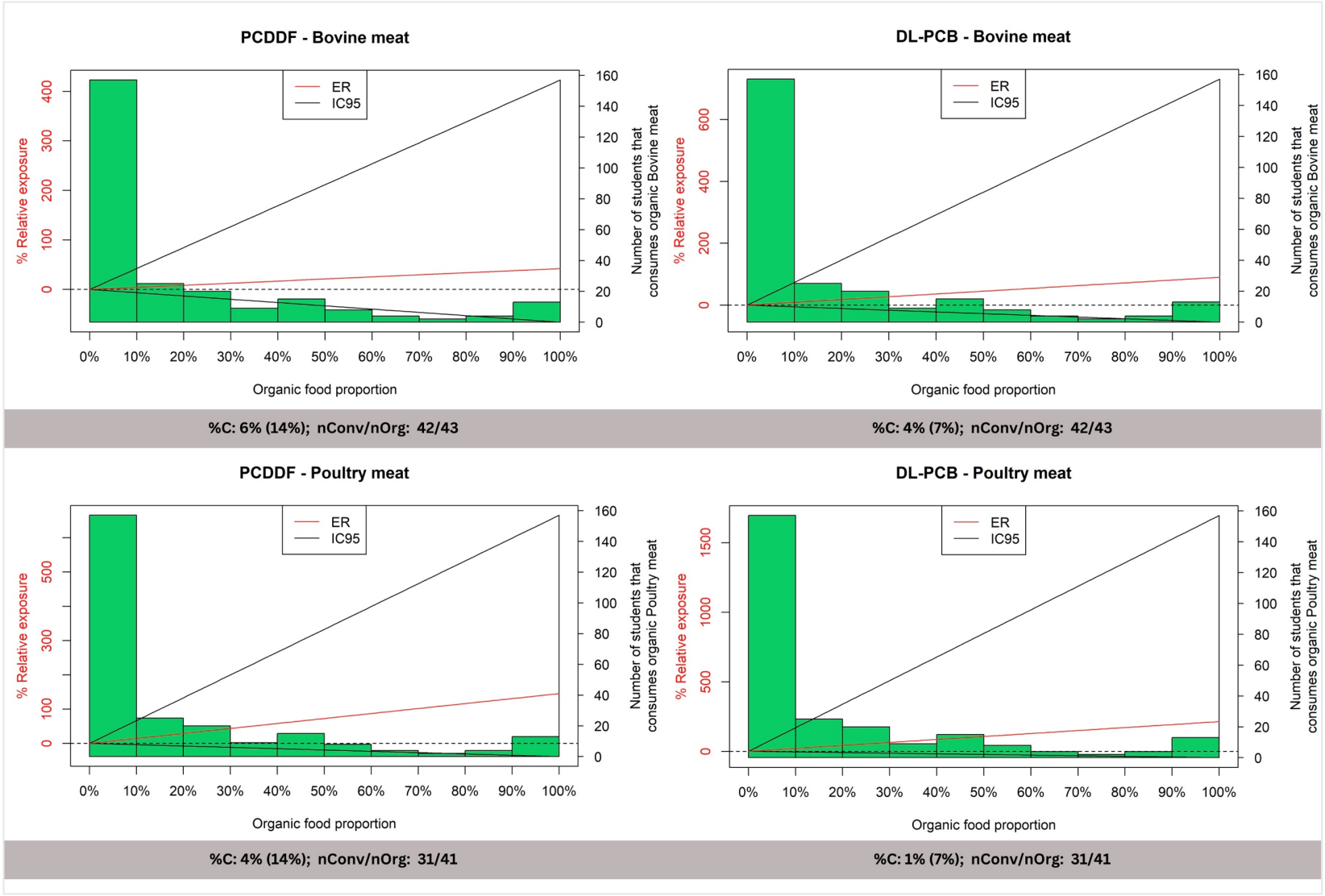


Figure 34. Relative exposure to dioxins according to the proportion of organic food consumed for meat; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms. %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

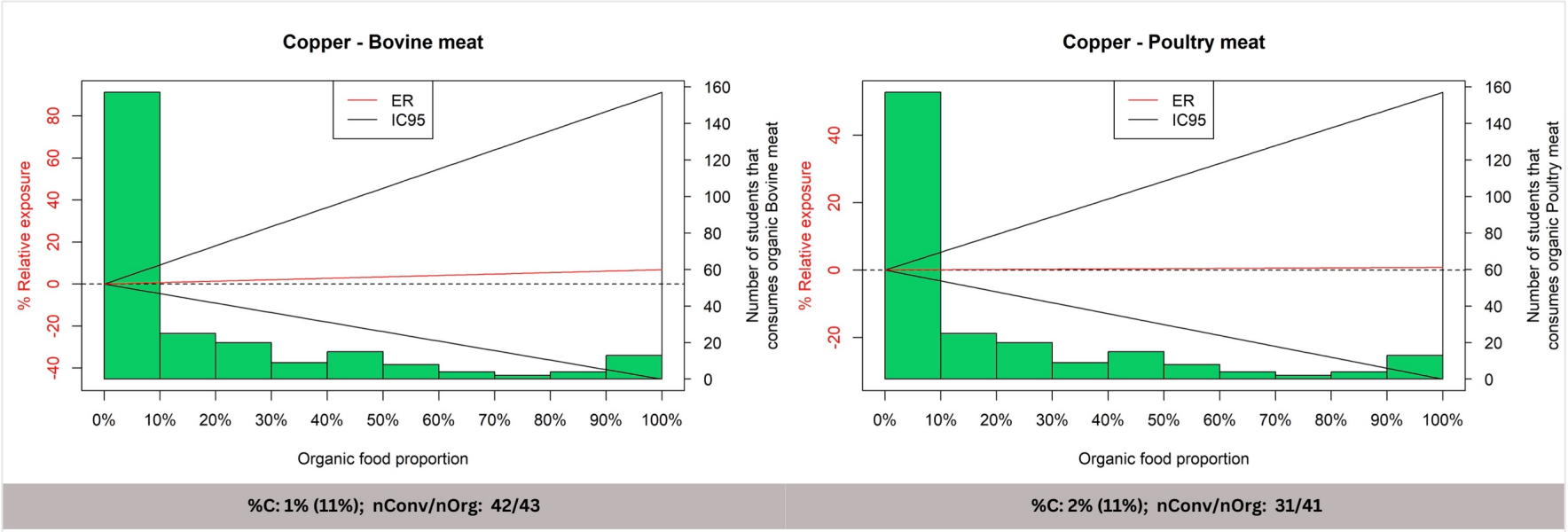


Figure 35. Relative exposure to copper according to the proportion of organic food consumed for meat; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value.

III. Discussion

This chapter is devoted to accomplishing two primary goals. First, it aims to characterize the dietary exposure of UGA students to food contaminants. Second, it aims to test the impact of organic food on the exposure to chemicals through a percentage relative exposure for chemicals of concern and specific matrices via the ERs calculated in Chapter 4.

1. Dietary habits

To characterize the exposure level of the studied population, first step was to assess the food consumption of the population and results were expressed for major food groups in g/week (Figure 25). Like the results from the INCA2 study reflecting the consumption of the French population, the food groups mostly consumed were also vegetables, dairy products fruits, and cereal products, yet with a different rate of consumption. The general French population consumed in average 38% and 5% more cereals and dairy products respectively than the students' population (1740 and 1400 respectively versus 1080 and 1328 g/week) (Afssa, 2009), while the students consumed in average 29% and 24% more vegetables and fruits (1388 and 1293 respectively versus 980 g/week for both).

Also compared to the INCA3 study, results were the same as fruits, vegetables (including soups), dairy products, and cereals (breads) constituted the top four contributors to the daily food intake in adults.

Regarding organic food consumption, the INCA2 study did not assess the proportion of consumers opting for organic food, rendering direct comparisons unfeasible. In contrast, INCA3 results demonstrated that the most frequently consumed organic items included fruits, vegetables, dairy, and eggs (ANSES, 2017a). Conversely, for the students' population, the primary organic food choices comprised nuts, eggs, fruits, and soy-based products (figure 25).

Moreover, a Belgian food consumption survey conducted in 2014-2015 showed that the most frequently purchased organic products were vegetables, fruit, dairy products, meat and bread (Bel S et al., 2015). Additional research is required to comprehensively understand the factors influencing the students' choices for organic products, specifically to unravel the reasons behind the varying preferences for certain substances being purchased in organic forms over others.

2. Dietary exposure

Based on the findings of this chapter, the following substances were found to be of concern for the studied population: dimethoate, cadmium, lead, inorganic arsenic and dioxins.

Starting with **dimethoate**, it was observed that the exposure levels in the students' population were lower compared to the EAT2 results. Also, the %TRV exceedance was 0.4% under the LB hypothesis and 59.1% under the UB hypothesis in the EAT2 study, while it's only 27% for the students' population under the UB hypothesis.
As per EAT2, the high analytical limits (LOD) for certain food groups, such as beverages and vegetables, which are major contributors to dimethoate, may have led to an overestimation of the substance and exposure levels under the UB hypothesis. Therefore, it is challenging to draw a conclusion on the risk with exposure levels surpassing TRVs under the UB hypothesis in this scenario, yet the exposure to dimethoate shouldn't be ignored.

Dimethoate, formerly used as an insecticide for treating vineyards, fruit, and vegetable crops, has not been authorized since 2019 under Regulation (EU) 2019/1090. However, during the EAT study measurements, its usage was still permitted. This implies that more recent measurements might reveal lower levels, hence, further research is needed to assess the evolving trends in dimethoate levels and associated exposure risks.

In addition, the ESTEBAN study aimed to measure organophosphorus pesticide levels in the adult French population through urine analysis, including dimethoate, the main metabolites of which are dimethylphosphate (DMP), dimethylthiophosphate (DMTP), and dimethyldithiophosphate (DMDTP). Methodological differences make direct comparisons between our results and those of the ESTEBAN. However, the notably high quantification rate of DMTP in urine samples from the general population (82.5% of adults) suggests potential persistent exposure to organophosphorus pesticides, although the metabolite does not specifically identify these pesticides (SPF, 2021b). This emphasizes the importance of not ignoring dimethoate exposure.

As for the exposure to **cadmium**, the student population in this study demonstrated a lower mean exposure level (0.1µg/kg bw/day) compared to EAT2 results (0.2 µg/kg bw/day) yet compared to a 7 times lower more recent TRV than that used in EAT2 (0,36 versus 2,5 µg/kg bw/day), 1.2% of the population exceeded the TRV (%IC_{95} = [0.2; 2.6]). Both studies identified

the same food groups as the primary contributors to cadmium exposure, but the percentages varied due to differences in consumption rates.

It is important to note that these results align with those found by EFSA in the scientific opinion report on cadmium in food, where the two highest contributors to the dietary exposure were cereal products and vegetables. The occurrence of cadmium in these food could be explained by the cadmium impurities in phosphate fertilizers applied during production. There is currently no EU legislation limiting the maximum level of cadmium in fertilizers but some countries have permanent exceptions to use national guidelines (EFSA, 2009a). Yet, following EFSA's TRV reduction in 2011, a review of maximum cadmium levels in foodstuffs at the European level was initiated (Commission Regulation (EU) 2023/915), suggesting that recent measurements might reveal lower contaminations, emphasizing the need for updated quantitative studies on cadmium in food.

Moreover, the ESTEBAN study indicated an increased cadmium impregnation level in the French population compared to previous biomonitoring studies, with higher cadmium levels in biological samples of the French population observed compared to other European and North American countries (Oleko et al., 2021).

All of these results underscore the critical exposure to cadmium and the necessity for further studies to identify the reasons to identify the reasons for the observed increases.

For **inorganic arsenic** exposure, the students' population exhibited lower average exposure levels under the LB hypothesis compared to EAT2 results (0.1, p95=0.5 µg/kg bw/day versus 0.2, p95=0.5 µg/kg bw/day). Conversely, under the UB hypothesis, they showed higher exposure levels (0.5, P95=1 µg/kg bw/day) in contrast to EAT2 (0.3, p95=0.5 µg/kg bw/day). Despite these variations, in both studies, some estimated exposure levels are within the $BMDL_{01}$ range (0.3 to 8 µg/kg b.w. per day), hence the possibility of a risk from Asi cannot be excluded for certain consumers. More initiatives to reduce dietary intake of inorganic arsenic are needed.

The major contributors to inorganic arsenic exposure differed between the two populations, influenced by variations in food consumption. For students under the LB hypothesis, water (34%) and dairy products (including milk) (29%) were significant contributors, contrasting with EAT2 where water (27%) and coffee (16%) dominated. Under the UB hypothesis, the students' major contributors were water (39%) and canned food (11%),

while EAT2 showed water (24%) and coffee (16%). Notably, canned food stands out as a source of Asi for students, underscoring the students' significant consumption of these items.

The results for students align with results of the EFSA report on Arsenic in food where the inorganic arsenic exposures from food and water across 19 European countries, using LB and UB concentrations, have been estimated to be 0.1 (p95=0.4) µg/kg bw/day under the LB hypothesis and 0.6 (p95=1) µg/kg bw/day under the UB hypothesis. Moreover, among the common food contributors the dietary Asi were drinking water. In areas where natural arsenic levels are high, the main source of inorganic arsenic in the diet can be drinking water. Notably, beverages like "fruit and vegetable juices, soft drinks, and alcoholic beverages" also contribute significantly to inorganic arsenic exposure due to the substantial water used in their preparation (EFSA Journal, 2009).

Considering the well-established health implications associated with arsenic, it is advisable to pursue further efforts to minimize the exposure to this substance especially from high food contributors (i.e., water, milk, and canned food). Analytical methods to quantify the various forms of arsenic speciation, are needed to fine-tune exposure.

Now for **lead,** the levels of dietary exposure were equal in EAT2 and for the students (0.2 (p95=0.4) µg/kg bw/day) and were below both BMDLs (nephrotic=0.63 µg/kg bw/day and cardiovascular= 1.5 µg/kg bw/day), yet given the small MOEs (<10), the possibility of developing health effects due to exposure to lead was not excluded in both studies.

For the students, the major contributor were water (14%) and vegetables (24%) versus alcoholic beverages (14%), cereals (13%), water (11%) in EAT2, reflecting the lower consumption alcoholic beverages and cereal products in the students.

Compared to the EFSA report on lead in food, higher values were observed for the average dietary exposure for adult consumers in 19 European countries (ranging from 0.4 (p95=0.7) to 1.2 (p95=2.4) µg/kg bw/ day). However, similarly to EAT2, overall, cereals, vegetables and tap water were the most important contributors to lead exposure in the general European population (EFSA Journal, 2010).

EAT2 and ESTEBAN reported a reduction in the levels of lead exposure in the French population (ANSES, 2011b; Oleko A et al., 2020), yet there are persistent and growing concerns about lead exposure, hence efforts to reduce dietary intakes of lead should be pursued.

And finally for **dioxins**, the mean dietary exposures for students were consistent with those from EAT2 (0.4 (p95=0.9) and 0.4 (p95=0.8) pg TEQ WHO_{05}/kg bw/day respectively). However, while the EAT2 population exhibited a slight exceedance rate for the TRV based on WHO_{98} TEQ (2.33 pg TEQ WHO_{98}/kg bw/day), students showed no exceedance. Conversely, the %TRV exceedance for students from the TRV based on WHO_{05} TEQ was considerable at 71% (%IC95= [65; 77]). The evaluation of the EAT2 population based on the new TRV was not made. The increase in %TRV exceedance is primarily attributed to an 8-fold decrease in the TRV value. This underscores the potential impact of different TRV evaluations, emphasizing the necessity for new studies to accurately assess the risk to the general French population.

And although students in general didn't consume enough fish as recommended by the PNNS, the main food contributor to this exposure were first fish and fish products (33%), dairy products (27%), and butter (11%). This highlights the high level of dioxins and DL-PCBs in fish and sea food.

Compared to other studies on dioxins in food, in an EFSA report, higher levels of exposure to dioxins were observed as levels ranged from 0.3 (p95=0.8) to 1.5 (p95=4.3) pg TEQ WHO_{05}/kg bw/day, suggesting a notable exceedance compared to the TRV (EFSA CONTAM Panel et al., 2018). On another hand, ESTEBAN's study on biomarkers indicated a lower impregnation of PCBs and PCDD/Fs in the general adult population compared to previous French studies (Balestier A. et al., 2021). Given the variation in results and given the substance's chemical stability, ecosystem persistence, and continued emissions, measures to reduce exposure and safeguard public health are necessary.

3. The impact of organic food consumption

To evaluate how consuming organic food affects exposure levels compared to conventional food, it was necessary to establish a metric, which was referred to as the percentage relative exposure. It was not possible to calculate this relative exposure using the contamination levels from the EAT2, given that the proportion of organic food integrated in the analyzed samples of EAT2 is unknown. It is indicated that within the analyzed samples, an organic proportion of the food is mixed, reflecting the proportion of organic food consumption in the INCA 2 study, yet these proportions were not specified, disabling the possibility to compare or assess the exposure to the chemicals from organic food using the EAT values.

Hence, based on the ER values from Chapter 4, a percentage relative exposure formula was developed enabling the visualization of the variation in relative exposure according to the proportion of organic food consumed. This gave an idea of the impact of organic food consumption on the exposure level to certain substances when replacing one matrix from conventional food with the same matrix from organic production.

This part was bound with the limited availability of data on the contamination of organic products in the literature, as the evaluation of the relative exposure was restricted to few matrices to which the evaluated substances had contamination values. Hence, an evaluation based on a 100% organic diet could not be done, as the data did not cover all the foods consumed.

Results of this part revealed a variation in the relative exposures for both the matrices and chemicals.

For the food matrices examined for various substances of concern (wheat flour, bovine, and poultry meat), outcomes were diverse and mixed. For a consumption of 100% organic wheat flour, results showed that the exposure to As, Cd, and Pb decreased (by 56%, 37% and 50% respectively), as hypothesized. Yet, more data is needed to draw an overall conclusion on the impact of consumption of organic wheat flour vis à vis the total exposure including other chemicals.

As for bovine meat, an increase in the consumption of the matrix in an organic version led to the decrease of the exposure to As and Pb, as it also led to the increase of the exposure to Cd, Cu, DL-PCBs and dioxins and furans.

Same for poultry meat, an increase in the consumption of organic lead to an increase of the exposure to some substances (As, Pb, DL-PCBs, PCDD/F) and a decrease for others (Cd).

In this case, no decision can be done regarding the sanitary effect of these organic matrices in respect to the exposure. These variations can be explained by the fact that the organic matrices used for this interpretation, though belonging to the same category, are distinct samples obtained from different sources (articles). Consequently, variations in soil composition, agricultural practices, and geographic locations can significantly influence the occurrence and the level of chemical contaminants.

As for the chemical substances, the results were also heterogeneous. For dimethoate, no controversial results were observed as all percentage relative exposures decreased with the increase of organic matrices consumption, which is logical based on the organic food production rules. Results are interesting for dimethoate especially with cherries given that cherries contributed to an average of 53% of the exposure to dimethoate. Yet, overall, only three matrices were evaluated, more data is needed to be able to conclude the effect of organic consumption on the exposure to dimethoate.

For the other substances, including arsenic, cadmium, our findings did not support our hypothesis that the consumption of organic food would lead to a reduction in exposure to chemicals. In fact, for some chemical substances, organic consumption decreased the exposures (As in bovine, and wheat flour; Cd in lettuce, wheat, wheat flour, and poultry; and Pb in bovine meat, lettuce, potatoes, and wheat flour), while for others, it contributed to a significant increase even sometimes to an increase of fivefold (Pb in carrots +439%).

Also, in some cases, the consumption of some organic matrices didn’t have any difference on the exposure compared to conventional matrices (As in carrots; Cd in apples; Cd and Pb in dairy products).

In our methodology, the relative exposure was directly related to the concentration of the chemical in the organic matrices versus the conventional matrices and did not depend on the individual's consumption data. Cadmium's increased presence in certain organic foods can be attributed to its incorporation into mineral fertilizers, particularly those derived from natural rocks. These fertilizers, commonly utilized in organic farming to provide crops with essential phosphorus and nitrogen nutrients, significantly contribute to the deposition of cadmium in cultivated soils. According to ANSES, approximately 50% of cadmium found in soils

originates from mineral phosphate fertilizers, while an additional 25% can be traced back to animal manure used as fertilizer in organic farming. Furthermore, the presence of cadmium in animal feed can result in the contamination of food products derived from animals (bovine meat) (ANSES, 2019a).

Moreover, organic farming practices often involve raising animals for longer periods. This can explain the higher levels of As and Pb in organic poultry meat, especially since when exposed to feed contaminated with toxic elements, long-lived animals can accumulate more metal in their blood, bones, and meat. Similarly, the higher concentration of POPs (DL-PCBs and PCDD-F) in organic poultry can be attributed to their accumulation in the fatty tissues of animals as they age. Hence, this practice accounts for the higher levels of these substances in organic poultry (Dervilly-Pinel et al., 2017). Regarding the higher lead content found in plant-based foods (carrots, spinach, and wheat), no clear explanation was found for that. It was hypothesized that this difference is probably linked to sporadic contamination, as lead contamination through airborne origin can be non-negligible, rather than to the organic production itself (Harcz et al., 2007; Malmauret et al., 2002).

Also, for copper, higher relative exposure was shown with increasing consumption of organic bovine and poultry meat. Copper is authorized in organic production and given that no MRLs are defined to authorized substances in organic production in accordance with Regulation (EC) No 2018/848 (Gómez-Ramos et al., 2020), we hypothesis that some might apply these substances without any limits which would lead to high levels in the feed of animals, and similarly to the other toxic elements, causing higher contamination in organic animal-based food as they are raised for longer periods.

In summary, among the different chemical substances examined in this study, the students' population exhibited notable exposure concerns to 5 chemicals: dimethoate, cadmium, inorganic arsenic, lead and dioxins.

As for the consumption of organic food, the analysis of relative exposure data did not reveal a consistent trend regarding the decrease of these exposures. The lack of comprehensive quantitative data on chemicals in organic food, coupled with non-standardized protocols from sample choices to preparation to data reporting, and gaps in the understanding of certain organic food contamination origins, contributed to the challenge of making decisive conclusions about the impact of organic food on reducing chemical exposure. Moreover, recent quantification studies must be done especially that, many of the authorized substances at the time of the quantification period (EAT2), are now non-authorized, hence their levels and occurrence might have changed.

This study represents a pioneering effort to examine how the consumption of organic food affects the exposure of a population to chemicals from different groups, that are of concern to this population. Despite inherent limitations, the developed method can be adapted as more data becomes available. A notable strength of the method lies in its ability to overcome the heterogeneity of reported data, which often hinders inter-study comparisons. This aspect was crucial for studying the potential impact of organic consumption on the exposure profile of the population, especially with the limited data.

Further research considering all the previously stated limitations is needed. Ideally, establishing a database containing information on the chemical content of organic food at the national level in France, as that available in EAT2, would be highly beneficial.

Summary of the chapter

The aim of this chapter was twofold: first, to assess the dietary exposure of UGA students to food contaminants, and second, to evaluate the impact of consuming organic food on exposure to chemical substances of concern.

To accomplish the **first objective**, the exposure levels of the student population were calculated using data from the EAT2 study. The results showed that the students had notable concerns regarding exposure to 5 chemicals: dimethoate, cadmium, inorganic arsenic, lead and dioxins.

For the **second objective**, data for the chemicals of concern were sought in Chapter 4, and an equation was developed to calculate the percentage relative exposure. This allowed for the visualization of the variation in relative exposure according to the proportion of organic food consumed. However, the evaluation of the relative exposure was limited to a few matrices due to the limited availability of data on the contamination of organic products in the literature.

The examination of relative exposure data did not disclose a consistent pattern with respect to the decrease of exposures. For the same chemical substance, certain organic matrices displayed a reduction in exposure, while others exhibited an increase.

Additionally, some noteworthy findings were observed, such as a substantial decrease in exposure to dimethoate through the ingestion of organic cherries, which is significant contributor under the LB hypothesis, or the impact of consuming organic wheat flour in decreasing exposure to various metals. However, the available data were insufficient to draw definitive conclusions regarding the influence of organic food consumption on exposure levels.

Further research focusing on organic food analysis, standardized quantification protocols, and investigating the underlying causes of contaminations in organic food is necessary to determine the effect of organic food consumption on the exposure profiles of a population.

Discussion & Perspectives

The major challenges surrounding nutrition include ensuring an optimal diet on both quantitative and qualitative levels, ensuring a good environmental impact of the food system, and ensuring food safety for the consumers including that from exposure to chemical substances especially that food is a significant source of exposure to environmental pollutants in the general population. This thesis comprehensively addressed various facets of these challenges.

First, various modes of agricultural production have emerged as responses to the environmental impact challenge, with organic farming playing a prominent role in addressing sustainability concerns. Specifically in Europe, a steady increase in organic farming was witnessed with 9.9% of total agricultural areas corresponding to organic farming in 2021, with France holding the largest organic production area in the EU (Eurostat, 2023). This method of production has been strongly encouraged and has been growing due its sustainable impact on the environment (Reganold & Wachter, 2016; Shubha et al., 2021), as well as the higher nutritional value for organic produced food compared to conventional ones as shown by different studies (Głodowska & Krawczyk, 2019; Lou et al., 2020; Zheng et al., 2019). Moreover, the French National Nutrition and health program (PNNS) recommends 20% of total consumptions of fruits and vegetables, cereals, and legumes from organic productions (Ministère des solidarités et de la santé, 2019). Nevertheless, the expansion of organic production gives rise to several inquiries. Existing literature highlights that, despite the prohibition of chemically synthesized molecules, which mitigates exposure to certain pesticides, specific issues associated with this production method persist (Dervilly-Pinel et al., 2017). That and the fact that the environmental contaminations can still occur in organic products as supported by the literature (Almeida-González et al., 2012; Baša Česnik et al., 2019; EFSA, 2018a; Witczak & Abdel-Gawad, 2012).

To date, risk assessments in France have either not considered the consumption of organic food or have been limited to specific chemical classes. Despite three total diet studies (EAT) conducted by ANSES on the French population in 2005, 2011 and 2016, with a specific focus on children (EATi), none of these studies differentiated contamination levels according to the food production method (ANSES, 2011b, 2011c, 2016; Leblanc et al., 2005). In addition, a study using BioNutriNet data only assessed chemical exposures from organic foods, focusing on pesticides (Baudry et al., 2021).

Currently, the assessment of exposure through the consumption of organic products by the means of the assessment approach, suggested by the WHO, FAO, and EFSA (EFSA et al., 2011b), which consists of quantifying exposure levels by incorporating data from both contamination and consumption sources is limited due to the lack of comprehensive databases for chemical substances in organic food In Europe. Therefore, the first objective of this thesis was (1) **to develop and implement a methodology to assess the impact of organic food consumption versus conventional food consumption on consumer exposure to environmental contaminants in Europe**, and thus understanding the impact of this environmentally friendly production mode on the chemical food safety of the investigated population.

Moreover, the second part of the thesis focused on the facet of evaluating nutritional quality. The population of university students at the University of Grenoble Alpes, who were already part of a larger initiative study PEANUTS, whose main objective is to assess all the variables that ensure the well-being of students, were selected to assess their dietary habits. This choice of population was made because numerous studies suggest that university students experience food insecurity at a higher rate than the general population (Abbey et al., 2022; Davitt et al., 2021; DeBate et al., 2021; Nikolaus et al., 2019). This vulnerability among students often leads to limited resources for students to meet their basic dietary needs (Maroto et al., 2015). Therefore, the second objective of this thesis was (2) **to assess the dietary quality of the young adult population using a PNNS score and examine the impact of food insecurity on the adherence to the national dietary recommendations**.

To achieve the first objective, the first step was to create a comprehensive database by collecting data on chemicals found in organic food from the available literature. This database was intended to combine data for different categories of chemicals in organic foods. The originality of this work lies in the fact that other databases, although containing some data on organic food, are not comprehensive. More specifically, these databases focus mainly on data related to pesticides in organic food (EFSA, 2013b, 2014c, 2016, 2017, 2018d, 2019, 2020, 2021c, 2022) or include other chemicals but with less emphasis in comparison to pesticides (CVUA Stuttgart, 2019). The importance of establishing an exhaustive database lies in its ability to facilitate a more comprehensive analysis, providing insights into the diverse range of compounds present in organic foods. In addition, this database serves as a valuable tool for understanding the coverage of research in the literature on organic foods, shedding light on both what have been studied and the areas that need to be further explored.

As a result of this database, we now know that data on chemical contaminants in organic food are poorly documented in the literature. This is also confirmed by the low number of reported data in the EFSA reports for pesticides in organic food compared to conventional food, where organic food represented only 6.5% of all samples tested (EFSA, 2022). In addition, this database showed a high heterogeneity in terms of the variables studied, with matrices, substances and couples being studied unevenly. The food groups "Cereals and primary derivatives of cereals" and "Fruit used as fruit" (nomenclature based on the FoodEx classification) were the most studied food groups in the existing literature, as they were studied for 83 and 65 different chemical substances, respectively (corresponding to 40% and 31% of the total chemical substances included in the database). With regard to the consumption of our study population, the average percentage consumption of these two food groups in organic form is 43% and 49%, respectively. However, the highest percentage of organic consumption was for eggs and nuts (61% and 64% of total consumption of food group respectively), which were only analyzed for 4 and 18 substances (Chapter 3). Therefore, more data on the matrices they consume mainly in organic form are needed to better assess the impact of organic consumption on this specific population studied.

Regarding the chemical substances from the database, 95% of the substances studied were not authorized in organic farming, with pesticides being the most studied (80% of the total chemical substances), specifically organophosphates, followed by inorganic contaminants (Chapter 3). This underlines the lack in the study of authorized substances in organic food, which highlights the need to assess these substances, especially as the study by Baudry et al. showed that organic food consumption contributes to a higher exposure to authorized substances (spinosad, azadirachtin and pyrethrins) when consuming organic food (Baudry et al., 2021).

As for the substances that showed a level of concern for the university students, namely dimethoate, cadmium, inorganic arsenic, lead and dioxins, these substances were not sufficiently studied, especially for the high-contributing foods. Starting with dimethoate, which was classified in the category of low scientific interest (with < 10 values in the database), there was only one value for cherries, which is the fruit that constituted 98% of the exposure to dimethoate under the LB hypothesis. Moreover, for cadmium and lead, although classified as substances of high scientific interest (having ≥ 20 values in the database, with 33 and 30 values, respectively), only 8 and 2 values, respectively, corresponded to vegetables, which for both is the main food group contributing to their exposures. Similarly, for dioxins, the database

included only 3 concentrations for cheese, which belongs to the food group of dairy products and contributes to 27% of the dietary exposure to this substance, while it did not present any data for fish or butter, which accounted for 33% and 11% of the contribution, respectively. Similarly, for inorganic arsenic, while dairy products accounted for 29% of the contribution to exposure under the LB hypothesis, no data for arsenic in dairy products were found in the database. Looking forward, more quantitative studies are needed for organic foods, considering both the substances and the matrices of concern, and focusing on the authorized substances, as these, despite being of natural origin, may cause potential health effects (EFSA et al., 2018a; EFSA et al., 2018b).

In addition, a major limitation of this database is that the data for the matrices from the literature were mainly raw matrices and not matrices prepared as consumed, which is a major limitation for exposure assessment. This is critical because the concentration of chemicals may change during the food transformation process. For example, the use of preparation materials made of stainless steel or aluminum, as highlighted in the references, can increase the levels of contaminants such as aluminum or nickel in the final product (ANSES, 2011b). Similarly, contaminants from the cooking process, such as hazardous air pollutants (HAPs) or acrylamide, can contribute to exposure levels (ANSES, 2011c). Conversely, failure to account for factors such as washing and peeling can result in overestimation of exposure levels, as these practices can reduce contaminant concentrations as reported in the references (Chung, 2018).

Another limitation of this database is the inconsistency of the reported data. In addition to the non-convertible units (units included mg/kg, mg/kg fw (fresh weight), mg/kg ww (wet weight), etc.), the different methods of sample preparation (washed, peeled, etc. unspecified), the different analytical methods used, the number of samples analyzed, as well as the limits of detection and quantification were not always specified in the studies. In addition, some concentrations were reported as ranges, others as means, medians, a single value or sometimes not specified.

In summary, the current state of data in the literature poses significant challenges for assessing the impact of organic foods on the exposure profile of the populations studied. The presence of different and non-convertible units, the samples analyzed (mainly raw matrices), and the use of different origins and analytical techniques posed obstacles to the direct calculation of exposure levels. As a result, it is not possible to calculate exposure

levels from organic matrices from this database and compare them with those calculated using EAT2 values to assess the impact of organic food consumption on exposure levels.

In the future, there is an urgent need to establish standardized protocols for quantitative studies reporting contaminants in organic foods. Such protocols would increase the utility of the data, allow effective comparisons with other production methods, and facilitate the estimation of population exposure levels using these values. This standardization is essential to advance our understanding of the broader effects of organic food consumption on exposure profiles.

Given the current data limitations that prevent exposure calculations from organic foods, a methodology was developed that first involved sorting articles in the database that provided data for the same pair (substance, matrix) in both organic and conventional production (22/32 articles). Subsequently, an excess ratio (ER) metric was developed, expressed as ER $= [C_{org} - C_{conv}]/[C_{org} + C_{conv}]$ with C org and C conv are the concentrations of the chemical in the organic and conventional matrices, respectively. This metric, applied to pairs from the same study, allowed a direct and reliable comparison of contamination levels ensuring the same methods, same preparations, same limits of quantification and same origins are applied to both matrices. Consequently, ER > 0, ER = 0, or ER < 0 indicated higher, equal, or lower contamination in the organic matrix, respectively. This provided an accurate comparison between the levels of contamination for the same pair from two different modes of production.

Moreover, this approach allowed the pooling of data across studies, expressed as ER distributions, providing a comprehensive assessment of contamination differences between organic and conventional foods at the chemical category level.

For the large-scale results, for both the contaminants and the authorized chemical substances in conventional production, the overall ERs are -0.11and –0.16, respectively, reflecting a lesser contamination in organic food. As for the authorized substances in organic production, ER was equal to 0.07 reflecting higher levels in organic products (Chapter 4). Given the limited data in the literature, only one substance resulted in our work as authorized in organic food, which is copper, yet the results aligned with what was shown in the ESTEBAN study where the frequent consumption of organic vegetables was associated with higher levels of urinary copper (SPF et al., 2021).

As for the individual results, higher levels of contamination in organic foods were explained by the literature for plant foods, by the recent transition to organic farming in the area, which could have affected soil contamination levels, or by the proximity to polluted environments, as it was also due to the tendency of some species to accumulate more contaminants due to their composition (example of carrots and carotene content with the tendency to accumulate POPs) (Witczak & Abdel-Gawad, 2012). In the case of animal products, higher levels of POPs or metals/trace elements have been attributed to the organic method of raising animals, which requires animal raising for long periods and therefore more accumulation (Dervilly-Pinel et al., 2017). Furthermore, in the case of additives, some were justified by natural occurrence (sulphite in wine) rather than contamination (EFSA, Younes, et al., 2022).

With the limited data available and the scarcity of studies dealing with the chemical food safety aspect of organic food, it is difficult to really understand the reason behind the presence of these chemicals, and this hinders the preventive actions to reduce these contaminations. Therefore, **more data are needed for this purpose, especially since organic farming has now been shown to have a good and sustainable impact on the environment, as it has also been shown that it could increase the nutritional value of organically produced products.**

The ER ratio method is an innovative, universal method, because the concentrations of chemical pollutants may vary within the same farm or from one farm to another within the same year or from one year to another, for a number of reasons that are difficult to identify and list, regardless of the type of production of the food matrix. This ratio removed all barriers and allowed us to get an overview of the chemical content of organic foods from what is available in the literature. This adaptable approach has potential for broader application as more data become available in the literature. In addition, it served as a base for developing a methodology to assess the impact of organic food consumption on exposure levels.

Now to the second part of this thesis, which focuses on the assessment of specific populations. This thesis presents the first study to assess the exposure levels of a university student population. Previous studies have assessed the exposure profiles of the general population (specifically EAT (ANSES, 2011b, 2011c)), children and infants (specifically EATi (ANSES, 2016)), and pregnant women (specifically ELFE (de Gavelle et al., 2016) and EDEN (Chan-Hon-Tong et al., 2013)). The focus was based on three axes: the diet and therefore the

assessment of the diet quality, the food insecurity status, which was the core of the choice of this population, and the exposure profile of this population, as they could have irregular diets due to their vulnerability to food insecurity and therefore low food budgets.

Starting with the quality of the diet, the decision to develop a new score and not use the PNNS-GS2 (Chaltiel, Adjibade, et al., 2019) is mainly due to the questionnaire that was developed before the start of this thesis and the food consumption data were collected before the selection of the population. Therefore, the questionnaire hindered the evaluation of 4 recommendations of the PNNS, either because of the absence of related questions (salt, sugary foods, and sugary drinks) or because of the way the question was asked, with answers that cannot be translated into the recommendation (for added fats, which was asked as % of the type of oil consumed, while the recommendation is to avoid overeating). In addition, the new PNNS did not include recommendations for protein intake (meat, poultry, and fish). Given the high cost of poultry and meat, and the fact that the majority of students didn't follow a special diet (190/274), it was interesting to see the association between adherence to this criterion (from the first PNNS score (Estaquio et al., 2009)) and food insecurity.

In Chapter 5, the goal was not to calculate an overall score for compliance with national dietary guidelines. Instead, our focus was on a careful evaluation of the individual scores, criterion by criterion. Through this approach, we sought to highlight the importance of each criterion and provide a nuanced understanding of which specific dietary guidelines were being followed with precision. This method facilitated the identification of groups that received due attention, as well as areas for improvement. Furthermore, a percentage frequency of compliance was calculated to provide an overview of the overall compliance with the tested criteria of the PNNS.

As such, the adherence to the PNNS guidelines among the UGA student population is generally considered satisfactory, with an overall compliance rate of 62% across different dietary groups. The majority of students were respective of the recommendations of alcohol (98% of students compliant), cold cuts cold (75%), and red meat (90%) (Chapter 5) indicating that this population has no unhealthy habits regarding these food groups. As for other recommendations, overall, the rates of full compliance (score of 3) varied across different criteria for the entire population. Specifically, 13% met the recommendations for fruits and vegetables, 31% for organic foods, 12% for nuts, 51% for legumes (with higher compliance observed in vegetarian groups), 23% for cereals and cereal products, 42% for dairy products,

38% for meat, poultry, and eggs, while only 4% met the recommendations for fish and fish products. These findings can only be compared with outcomes from one study done in Rouen in 2021 that assessed changes in diet quality before and during the COVID-19 period, evaluating six components using the PNNS-GS2 score. Despite the different scoring system used, and compared to results from the pre-COVID assessment, the UGA population demonstrated higher compliance with the criteria for nuts, legumes, and dairy products (12%, 51%, and 41%, respectively) compared to the Rouen students (10%, 21%, and 31%, respectively) (L. Miller et al., 2022).

As for the FI, the prevalence among the UGA students (moderate and severe FI constituted 9%, while severe constituted only 1.2%) was considerably low compared to the prevalence among our population from Western and European countries ranging from 14.8% to 58.8% (Berg & Raubenheimer, 2015; Bruening et al., 2016; Chaparro et al., 2009; Freudenberg et al., 2009; Gaines et al., 2014; Hughes et al., 2011; Micevski et al., 2013; Morris et al., 2016; Patton-López et al., 2014; Payne-Sturges et al., 2018; Theodoridis et al., 2018). Yet, this prevalence was significantly associated with the compliances to some PNNS guidelines. As shown, FI students had lower adherence to the fruits and vegetables ($p = 0.004$), and the alcohol ($p = 0.005$) criteria, yet they exhibited higher adherence to the meat, poultry, and eggs criterion ($p = 0.021$) and the whole grain cereals criterion ($p = 0.01$).
The associations were explained by the higher cost of fruits and vegetables (Becker et al., 2017; IPSOS, 2022) and alcohol (Morrell et al., 2021; Xu & Chaloupka, 2011), higher egg consumption rather than meat satisfying the recommendation, as eggs are believed to be a good economical alternative animal proteins (CNPO, 2021), and higher fiber content for whole grain, providing longer satiety (Martini et al., 2018) to the FI students.

Limitation of this part included the small number of population as further research with a larger sample size is necessary to understand the factors influencing students' choices compliance. Also, the dietary assessment conducted in 2021 may have been influenced by the ongoing COVID pandemic, as people were still under its effects, potentially impacting both dietary habits and food insecurity prevalence.

In conclusion, although the adherences to the dietary recommendations and the FI prevalence were better compared to other studies, there is many room for increasing the compliance of the UGA students to the PNNS recommendations especially that FI has been significantly shown to increase or decrease the compliance to different food groups.

Furthermore, in the context of promoting healthy eating among university students, it would have been beneficial to also assess their intake of sugary foods and fast food, as students often exhibit tendencies towards unhealthy eating habits; besides, such dietary choices may pose potential exposure risks, as exemplified by the presence of cadmium in pastries from sugary foods or acrylamide in fries from the fast food category (ANSES, 2011b, 2011c), highlighting the importance of comprehensive dietary assessments to address potential health exposures.

Future efforts should be focused on the incorporation with the CROUS to develop comprehensive meal plans. These plans can be strategically designed to include a variety of fruits and vegetables as well as valuable proteins such as chicken, which may be perceived as expensive by the students. Incorporating a serving of fatty fish into meal plans can significantly improve the overall health profile to address low compliance rates for the fish criterion. Also, to meet the diverse needs of international students, a special focus on cultural food preferences can be incorporated into meal plans (for example offering some options from different cuisines) ensuring a well-rounded approach to promoting healthy eating habits among the student population. In addition, fostering partnerships with local grocery stores to offer discounts on products with a favorable NutriScore can make healthier food options more accessible to students. Moreover, this work highlighted the need to develop specific PNNS guidelines, which are required for particular dietary groups (vegans, vegetarians etc.), as eating habits are evolving especially in the young adult generation.

Now for the exposure characterization, a level of concern was shown for the following substances: dimethoate, cadmium, lead, inorganic arsenic and dioxins (PCDD/F +DL-PCBs) as some of the population individuals exceeded the TRVs or had low MOE for these substances. Compared to the general population (EAT2) (ANSES, 2011b, 2011c), when substances were evaluated for the same TRVs, the students showed lower % exceedance. Yet as some substances were also evaluated for more recent TRV, as TRVs decreased (by 16 folds, 7 and 8 folds respectively for dioxins, Cd and Ni), the %TRVs exceeded for the students for dioxins and Cd, highlighting the importance of performing a new EAT were the updated TRVs are used as references. Also, as mentioned in Chapter 6, new quantitative measurements have to be made because the data from EAT2 are now outdated: some substances were allowed at the time of the EAT2 measurements and are now banned (dimethoate), and other substances that were not

present at that time are now substances of concern in France (per- and polyfluoroalkyl substances (PFAS) (Berthou et al., 2023).

Similar to the results of INCA2 (Afssa, 2009), which includes the population studied in EAT2, and similar to those of INCA3 (ANSES, 2017a), the food groups most consumed by the UGA students were also vegetables, dairy products, fruits, and cereals, but with a different rate of consumption. Therefore, **in conclusion, although it was important to consider the university student population in terms of food insecurity, we cannot definitively conclude whether this group is vulnerable to chemical exposures from food. Despite showing a similar exposure profile to the general population, our findings are limited by the small and potentially unrepresentative sample size. Furthermore, other specific populations need to be evaluated, especially those that, to my knowledge, have never been studied in France in terms of dietary exposure, including the elderly or immunocompromised individuals.**

Finally, regarding the impact of organic food consumption, specifically on the exposure to the substances showing a level of concern, results were not conclusive. On an individual level, some results were interesting: for example, the consumption of organic cherries, which was a main contributor to the exposure to dimethoate, would reduce the exposure to this substance by 71%. Yet overall, results were varied and mixed, indicating a decrease in exposure levels with organic consumption for some matrices and an increase for others, as well as different results for specific substances of concern in different matrices.

The major limitation of the percent relative exposure method is that it is solely dependent on the ER value generated in chapter 4 and does not consider the consumption data of the individuals. Thus, the increased exposure to the chemical with increased consumption of organic foods is attributed solely to the higher chemical content in the organic matrix. This underscores the need for further studies to confirm and understand these contaminations, potentially leading to preventive measures such as regulatory changes as seen in the case of organic animal-based foods (Dervilly-Pinel et al., 2017). This part was bound by the limited availability of data on contamination of organic products in the literature, which prevented an evaluation based on a 100% organic diet. Also, the different origins of the matrices, affected our results as the variation in matrices origins can significantly impact the composition and concentrations of contaminants present in food.

In summary, the method developed to assess the impact of organic food consumption on exposure levels was a way to overcome the limitations of existing data and to provide a framework for a better understanding of how such consumption may affect exposure, especially to substances of concern. The strengths of this method are that it is universal and overcomes all the data limitations that made it impossible to calculate the exposure level from the existing values. However, the weaknesses of this method are the paucity of data on organic foods and the fact that it does not consider individual consumption data. The best way to obtain a more comprehensive assessment is to establish a national database quantifying the levels of different classes of chemicals in all matrices consumed by the French population and purchased as organic, in order to see the impact of an all-organic diet on the population's exposure profile.

External Perspectives for Future Research and Conclusion

In addition to the perspectives due to the limitations, further perspectives could be considered in the development of the database:

Inclusion of mycotoxins: Mycotoxins are substances of concern in organic production, and their presence in food products can pose a threat to global food security(Giannioti et al., 2023). Including mycotoxins in the MeSH (Medical Subject Headings) of the database would provide a more comprehensive understanding of the substances of concern in organic production

Assessing the relationship between adherence to PNNS and exposure to substances of concern: An important consideration in the improvement of nutrition is the careful assessment of the balance of risks and benefits. Adequate dietary intake is fundamental to maintaining overall health and well-being. However, the foods we consume, regardless of how they are produced, may contain various chemicals that may pose potential health risks through cumulative exposure. Therefore, in addition to prioritizing adequate food consumption, it's imperative to remain mindful of potential chemical exposures. In this context, the study of the relationship between compliance with the guidelines of the PNNS and exposure to the substances of concern is of great importance for our population. Investigating this relationship could provide valuable insights into how closely adherence to the PNNS guidelines correlates with exposure to harmful chemicals, thereby elucidating the effectiveness of dietary recommendations in mitigating health risks. This investigation could include conducting correlation tests or implementing different models of adherence to the PNNS to assess different

levels of exposure to substances of concern. This will allow researchers to better understand the interplay between dietary habits and chemical exposures, and ultimately inform evidence-based strategies to promote healthier and safer food choices.

The goal of this thesis was mainly to develop and implement a methodology for assessing the influence of organic versus conventional food consumption on a consumer's exposure to chemical substances. As the students showed a level of concern for dimethoate, Cd, Pb, Asi and dioxins (PCDD/F +DL-PCBs), the impact of consumption of organic on the level of exposure to these substances was not conclusive. Further research considering all the previously stated limitations is needed.

In summary, this thesis conducted a comprehensive assessment of the nutritional quality of the diets of UGA students, it examined the influence of food insecurity on the adherence to French dietary guidelines and it explored the students' chemical exposure profile. Moreover, this thesis introduced an innovative methodology that broke barriers of the existing data to demonstrate the impact of organic food consumption, which is an emerging sustainable agriculture method. Organic farming methods have shown positive effects on the environment and some studies suggest that they may offer superior nutritional quality compared to foods from other production methods. Consequently, this thesis serves as an illustrative example of a multifaceted exploration that addresses numerous challenges related to dietary practices.

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Contributions to the Field

Scientific Articles

Status: Submitted

Choueiri J, Petit P, Balducci F, Bicout DJ, Demeilliers C. (2024). Literature-based inventory of chemical substances concentrations measured in organic food consumed in Europe.

Choueiri J, Bicout DJ , Dias D, Balducci F, Couturier K, Royer N , Trancher V, Huc M , Clavier V, Christine Demeilliers C. (2024). Food habits of university students in Grenoble, France: adherence to the National Nutritional Program and association with food insecurity

Oral Communications

Joanna Choueiri, Pascal Petit, Franck Balducci, Dominique J. Bicout, Christine Demeilliers. Human exposure to environmental pollutants through food. Journée des doctorants TIMC, 12 avril 2022, Grenoble

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Posters

Joanna Choueiri, Pascal Petit, Franck Balducci, Dominique J. Bicout, Christine Demeilliers. Chemical contaminants in organic food : a database. 20ème Journée de l'EDISCE, 2 juin 2022, Faculté de Médecine et Pharmacie de Grenoble, La Tronche (Isère) (Poster preview in next page)

E-Poster

Joanna Choueiri, Pascal Petit, Franck Balducci, Dominique J. Bicout, Christine Demeilliers. Contaminants chimiques dans les aliments issus de l'agriculture biologique : Construction d'une base de données à partir de la littérature scientifique. Journée Francophone de Nutrition, 16-18 novembre 2022, Toulouse

Chemical contaminants in organic food: a database

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*Demeilliers C and Bicout D J contributed equally to this work as thesis supervisors

Introduction

- The **exposome** is " The cumulative measure of environmental influences and associated biological responses throughout the lifespan, including exposures from the environment, **diet**, behavior, and endogenous processes " (Miller and Jones, 2013).
- More than **90%** of human exposure to contaminants is **through food** (WHO, 2021).
- It is important to characterize and quantify the exposure of a population from chemicals in food.
- In France, national studies on exposure evaluation from food (TDS) have been conducted but **did not distinguish organic food** from conventional food.
- 14% of the French population consumed organic products daily (Agence Bio, 2020)
- While databases on chemical contaminants in food exist in the literature, **few data on chemical contaminants in organic food is found** (Food & Authority, 2018).

"The Exposome: Understanding the Effect of the Environment on Our Health", ISGLOBAL, 2020

Objectives

·**Gather** in a **database all chemical substances** and their levels in **organic food** in the **EU region retreived from literature**
·**Assessing the contamination levels in organic food**

Methodology

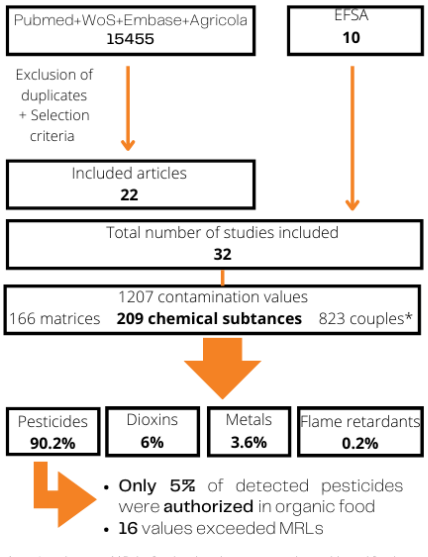
Literature search

Literature search was done from June 10 to June 15, 2021 in PubMed, Web of Science (WoS), Embase and Agricola + EFSA

Included articles

- written either in English or in French
- study organic food AND any residue, contamination OR chemical detection
- study food in the EU market regardless the country of production

Results



*matrix, substance **Baby foods other than processed cereal-based foods

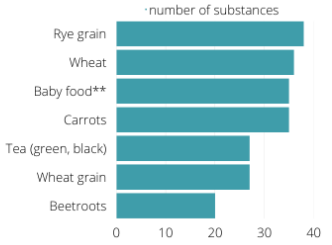


Figure 1. Organic matrices with the highest number of substances

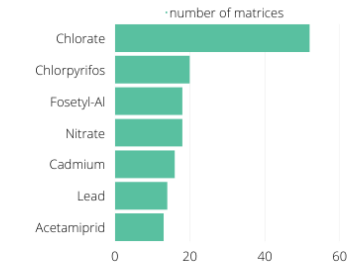


Figure 2. Substances emerging most in the organic matrices

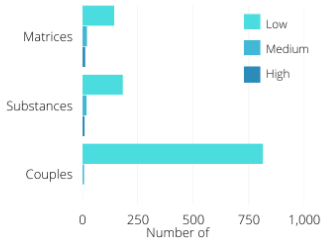


Figure 3. Distribution of organic matrices, substances, and couples in terms of low, medium and high scientific prominence. Variables were classified as high if they occur > 20 x in the DB table, low if they occur < 10x and medium if they occur in between 10 and 20x.

Table 1. Variables with high prominence

Matrices	Baby food**, Tea (green, black), Wheat, Carrots, wheat flour, Rye grain, Tomato, Lettuces, Wheat grain, spinach, Beetroots, Rice, grapes
Substances	Chlorate, Fosetyl-Al , Nitrate, Chlorpyrifos, Cadmium, Lead, Copper, Bromide ion, Anthraquinone, Spinosad
Couples	Baby foods other than processed cereal-based foods,Fosetyl-Al

Conclusion

- Results showed **little diversity** in terms of matrices and substances **highly studied in organic food.**
- Chemical quantification studies are lacking in organic food.
- This database will serve as a **tool to study exposure levels** from **organic food.**

Limitations

- Some chemicals were not included in the search (i.e. mycotoxins).
- The data were not all of the same scientific prominence which is a major limitation of this database .

References: in supplemental appendix

Acknowledgement: I would like to thank Ms. Sylvette Liaudy for helping me with the bibliography work.

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Appendices

I. Appendix- Chapter 1 (A1)

Description of chemical substances included in this thesis

Inorganic contaminants

Inorganic pollutants include metals that occur naturally in the environment, water, and soil and can result from human activities such as agriculture (use of pesticides and fertilizers, e.g., cadmium in fertilizers), industry (mining and metal processing, e.g., aluminum and lead), car exhaust (e.g., lead and mercury in fuel), or during food processing (e.g., nickel and aluminum from utensils) and storage (e.g., aluminum in food contact materials) (EFSA, 2024b). Unlike minerals, they are not necessary for the body function, moreover, their sustainable and long-term bioaccumulation poses a huge threat to human health (Pandey et al., 2016; Rodríguez et al., 2009). Metals can be present in inorganic and/or organic forms, and their toxicity is of significant concern in their inorganic form, except for mercury (Abd Elnabi et al., 2023).

Table A1-1 provides details on the toxicology of inorganic contaminants assessed in this thesis. Copper has also been included in the table as it was the only mineral evaluated since it is permitted in organic production and has been assigned a TRV for chronic liver inflammatory effects.

Table A1- 1. Toxicological information for inorganic contaminants and the mineral assessed in this thesis.			
Chemicals	**TRV s**	**TRV effects**	**TRV References**
Cadmium	0,357 µg/kg bw/day	Renal effects in human	(EFSA, 2009a)
Lead	0,5 µg/kg bw/day	Neurotoxic in fetuses/infants	(EFSA Journal, 2010)
	0,63 µg/kg bw/day	Nephrotoxic in humans	
	1,5 µg/kg bw/day	Cardiovascular in humans	
Nickel	22 µg/kg bw/day*	reduced relative organ weights (liver and kidney)	(WHO, 2005)
	13 µg/kg bw/day**	post-implantation fetal loss in rats	**(EFSA 2020)**
Aluminum	143 µg/kg bw/day	neurodevelopmental effects in mice	(JECFA, 2006)
Inorganic Arsenic	0,3 – 8 µg/kg bw/day	Lung cancer in human	(EFSA Journal, 2009)
Methylmercury	0,23 µg/kg bw/day	Neurodevelopmental effects in human	(JECFA, 2004)
Copper	150 µg/kg bw/day	Chronic liver inflammation in rats	(EFSA et al., 2018)
TRV: Toxicological Reference value *TRV used in the French Total Diet Study (EAT2) **Updated TRV value			

Phytosanitary Products

There is no unique definition to define pesticides as many definitions exist today. Yet as defined by the FAO in 2022, pesticide means “any substance intended for preventing, destroying, attracting, repelling, or controlling any pest including unwanted species of plants or animals during the production, storage, transport, distribution and processing of food, agricultural commodities, or animal feeds or which may be administered to animals for the control of ectoparasites. Including substance intended for use as a plant growth regulator, defoliant, desiccant, fruit thinning agent, or sprouting inhibitor and substances applied to crops either before or after harvest to protect the commodity from deterioration during storage and transport”. In agriculture, pesticides are referred to as "phytosanitary products" (ANSES, 2023b).

In Europe, all information on phytosanitary products including the regulation, approval for sale, usage, and control were initially established by Council Directive 91/414/EEC on July 15, 1991. This directive was later abrogated on June 13, 2011, and replaced by Regulation (EC) No. 1107/2009. Therefore, only substances explicitly listed in this regulation are allowed on the European market. As a result, any substance not on this list that is found in food today is considered a contaminant rather than a residue. Although banned, certain pesticides can persist in the environment and enter the food chain. A good example is the group of organochlorines, which have been banned since the 1970s, but continue to persist in the environment (POPs) (ANSES, 2011c). It's important to note that this group is just one of many examples of pesticides that are banned but still exist in the environment. Table A1-2 details 11 authorized and non-authorized pesticides that were assessed in this thesis, highlighting their authorization status along with some toxicological information.

Table A1-2. Authorization Status and Toxicological information for phytosanitary products assessed in this thesis

Chemicals	Status under Reg. (EC) No 1107/2009	References	TRVs (µg/kg bw/day)	TRV effects	TRV References
Dimethoate	Not listed	Reg. (EU) 2019/1090	1	Inhibition of cholinesterase activity in rats	(EFSA, 2006)
Deltamethrin	Listed	Reg. (EU) 2023/1757	10	short- and medium-term effects on the nervous system in dogs	(EFSA, 2009c)
Pyrethrins	Listed	Reg. (EU) 2023/1446	40	Hepatic effects in rats	(EFSA, 2013a)
Hexachlorobenzene	Banned/ out of Dir.	Reg. (EU) 850/2004	0,8*		(Arnold et al., 1985)
			0,07**		**(ATSDR, 2015)**
Chlorothalonil	Not listed	Reg. (EU) 2019/677	15	occurrence of pre-neoplastic lesions of the kidney and forestomach	(EFSA, 2018)
Chlorpyriphos-ethyl	Not listed	Reg. (EU) 2020/18	10*	Inhibition of brain cholinesterase activity in rats	(European Comission, 2005a)
			1**		**(EFSA, 2014a)**
Chlorpyriphos-methyl	Not listed	Reg. (EU) 2020/17	10		(European Comission, 2005b)
Pirimiphos-methyl	Listed	Reg. (EU) 2023/918	4		(EFSA, 2005a)
Ethion	Not listed	2002/2076/EC	2	Effects on fetal development	(WHO-JMPR, 1990)
Bifenthrin	Not listed	2009/887/EC	15	Developmental toxicity	(EFSA, 2011)
λ-cyhalothrin	Listed	Reg. (EU) No 2019/724	5*	Hepatic and central nervous system effects in dogs	(European Comission, 2001)
			2,5**	Decrease in body weight gain in rats	**(EFSA, 2014b)**

TRV: Toxicological Reference value
*TRV used in the French Total Diet Study (EAT2)
**Updated TRV value

Dioxins, Furans & Polychlorinated biphenyls

Dioxins (PCDDs), furans (PCDFs) and polychlorinated biphenyls (PCBs) are persistent organic pollutants with 75, 135 and 209 congeners respectively. These congeners, characterized by the number and position of chlorine atoms on the aromatic rings, are very stable chemically, insoluble in water but highly soluble in lipids.

PCDDs and PCDFs are mainly the by-products of industrial and combustion processes (such as incinerations, metallurgical processing, bleaching of paper pulp, and the manufacturing of some herbicides and pesticides) but they can also result from natural processes like volcanic eruptions and forest fires . Waste incineration is one of the largest contributors to the release of PCDDs and PCDFs into the environment, especially when combustion is incomplete (Srogi, 2008).

PCBs, on the other hand, result from industrial activities, as they were produced and used for heat transfer and electrical insulating properties, flame retardant properties, as well as, and their chemical and physical stability (in inks, paints). Their production was banned in 1987, yet they can still be found in the environment (Reddy et al., 2019).

From a toxicological point of view, PCBs are often divided into two categories: 12 PCB congeners that share the same type of toxicity (common mechanism) as PCDD/Fs are referred to as "dioxin-like" PCBs (DL-PCBs). The others are referred to as "non-dioxin-like" PCBs (PCB-NDL). Accordingly, the term "dioxins" may often be used in the literature to refer to PCDDs, PCDFs and DL-PCBs.

Due to their similar planar structure, hydrophobicity, persistence, and common mode of action, namely the activation of the aryl hydrocarbon receptor [AhR], PCDDs, PCDFs and DL-PCBs are considered together in risk assessment studies. Consequently, the WHO developed the Toxic Equivalency Factor (TEF) approach to characterize the toxicity of each dioxin in relation to a reference congener, primarily TCDD (Ring et al., 2023). Historically, 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD) has been considered the most toxic congener of dioxins. Most dioxins are considered to be less toxic than TCDD, with the exception of 1,2,3,7,8-pentachlorodibenzo-p-dioxin, which is considered to be approximately as toxic as 2,3,7,8-TCDD. Consequently, a TEF value have been assigned to each dioxin congener, comparing its relative toxicity in "orders of magnitude" to that of 2,3,7,8-TCDD, based on detailed scientific review of chemical structures and toxicological databases.

TEF equivalent factors where set in 1998 by the WHO expertise (Van et al., 1998), but values were then re-evaluated in 2005 (Van den Berg et al., 2006). In 2022, the WHO convened an expert consultation in Lisbon, Portugal, to re-evaluate the Toxic Equivalency Factors (TEFs) for dioxin-like compounds, mainly after the EFSA recommended re-evaluation of the 2005-TEFs values, to harmonize data on an international level. The WHO review of TEFs was expected to be completed in 2023 (WHO, 2022c), however the peer-review paper with the new TEFs is not yet published. Table A1-3 retrieved from (Van den Berg et al., 2006) is a summary of WHO 1998 and WHO 2005 TEF values.

Table A1-3. TEF values from WHO 1998 and WHO 2005 (Van den Berg et al., 2006).		
	WHO 1998 TEF	**WHO 2005 TEF**
Chlorinated dibenzo-p-dioxins		
2,3,7,8-TCDD	1	1
1,2,3,7,8-PeCDD	1	1
1,2,3,4,7,8-HxCDD	0.1	0.1
1,2,3,6,7,8-HxCDD	0.1	0.1
1,2,3,7,8,9-HxCDD	0.1	0.1
1,2,3,4,6,7,8-HpCDD	0.01	0.01
OCDD	0,0001	**0,0003**
Chlorinated dibenzofurans		
2,3,7,8-TCDF	0.1	0.1
1,2,3,7,8-PeCDF	0.05	**0.03**
2,3,4,7,8-PeCDF	0.5	**0.3**
1,2,3,4,7,8-HxCDF	0.1	0.1
1,2,3,6,7,8-HxCDF	0.1	0.1
1,2,3,7,8,9-HxCDF	0.1	0.1
2,3,4,6,7,8-HxCDF	0.1	0.1
1,2,3,4,6,7,8-HpCDF	0.01	0.01
1,2,3,6,7,8,9-HpCDF	0.01	0.01
OCDF	0.0001	**0.0003**
Non-ortho substituted PCBs		
3,3’,4,4’-tetraCB (PCB 77)	0.0001	0.0001
3,4,4’,5-tetraCB (PCB 81)	0.0001	**0.0003**
3,3’,4,4’,5-pentaCB (PCB 126)	0.1	0.1
3,3’,4,4’,5,5’-hexaCB (PCB 169)	0.01	**0.03**
Mono-ortho substituted PCBs		
2,3,3’,4,4’-pentaCB (PCB 105)	0.0001	**0.00003**
2,3,4,4’,5-pentaCB (PCB 114)	0.0005	**0.00003**
2,3’,4,4’,5-pentaCB (PCB 118)	0.0001	**0.00003**
2’,3,4,4’,5-pentaCB (PCB 123)	0.0001	**0.00003**
2,3,3’,4,4’,5-hexaCB (PCB 156)	0.0005	**0.00003**
2,3,3’,4,4’,5’-hexaCB (PCB 157)	0.0005	**0.00003**
2,3’,4,4’,5,5’-hexaCB (PCB 167)	0.00001	**0.00003**
2,3,3’,4,4’,5,5’-heptaCB (PCB 189)	0.0001	**0.00003**
Values in bold indicate a change in the TEF value		

When estimating the level of exposure to dioxins, the concentrations of dioxin congeners are multiplied with corresponding TEF values and then the sum of all these products is calculated generating a TEQ value which is the toxic equivalent for the environmental sample.

$$TEQ = \sum_j (PCDDj * TEFj) + \sum_j (PCDFj * TEFj) + \sum_j (PCBj * TEFj)$$

(TEQ: contamination level of the food in TCDD equivalent expressed in pg TEQ.kg^{-1} of material; PCDDj: concentration of the PCDDj congener expressed in pg.kg^{-1}; TEFj: weighting coefficient assigned to congener j)

Further, table A1-4 provides toxicological information for dioxins and NDL-PCBs.

Table A1-4. Toxicological information on Dioxins and Non-Dioxin-Like Polychlorinated Biphenyls.

Chemicals	TRVs	TRV effects	TRV References
PCDD/F & DL-PCB	2,33 pg TEQ WHO_{98} / kg bw/day*	Reproductive toxicity in rats	(JECFA, 2001)
	0,29 pg TEQ WHO_{05}/kg bw/day**	Effects on semen quality in human	**(EFSA CONTAM Panel et al., 2018)**
NDL-PCB	10 ng/kg bw/day	Effects in brain development in rat fetuses	(Afssa, 2007)

PCDD/F: Polychlorinated Dibenzo-p-Dioxins and Polychlorinated Dibenzofurans
PCB-DL: Polychlorinated Biphenyls - Dioxin-Like
NDL-PCB: Non-Dioxin-Like Polychlorinated Biphenyls
TRV: Toxicological Reference value
*TRV used in the French Total Diet Study (EAT2)
**Updated TRV value

Food additives

Food additives are chemical compounds integrated into food products, serve multifaceted roles, including the prevention of spoilage, the preservation of nutritional value, and the enhancement of appearance. Despite their deliberate inclusion, these additives can have implications for human health (Sambu et al., 2022).

In the last total diet study in France, and among the food additives tested, sulfites emerged as a high-risk food additive for the adult population (ANSES, 2011c). Sulfites are approved in the European Union as food additives according to Annex II and Annex III of Regulation (EC) No 1333/2008 (EFSA ANS, 2016). Table A1-5 provides toxicological information on sulfites.

Table A1- 5. Toxicological information on Sulfites.

Chemical	TRV	TRV effects	TRV Reference
Sulfites	700 µg/kg bw/day	Gastrointestinal effects in in rats	(SCF, 1996)

TRV: Toxicological Reference value

II. Appendix- Chapter 3 (A3)

Table A3-1. Criteria for selection based on language, title, abstract and data availability

Question	Description	Response	
		Yes	**No**
Level 1 : Language			
Q1	Is the paper written in French or English?	1	0
L1= Q1 ; Elligible for L1=1			
Level 2 : Title			
Q1	Does the title mention terms related to organic food OR organic products?	1	0
Q2	Does the title mention terms related to any of the following terms : Pesticide* OR contaminant* OR pollutant* OR chemical* OR herbicide* OR fertilizer* OR fungicide* OR insecticide* OR metals* OR residue* OR contamination* OR Food Safety*?	1	0
L2=Q1+ Q2; Elligible for L2=2			
Level 3 : Abstract			
Q1	Does the abstract describe a focused analysis on contamination or residues in organic products?	1	0
Q2	Quantification notion* : Value, data, level, concentration, measure, profile, quantification, composition, sampling, sample, amount	1	0
L3= Q2*Q1 ; Elligible for L3=1			
Level 4: Data availability			
Q1	Does the article contain contamination values ?	1	0
Q2	Does these values correspond to organic food from EU markets ?	1	0
L4=Q1+Q2 ; Eligible for L4=2			

Table A3-2. Elements of the database table columns

Country of origin	**Unknown, South Korea, Italy, France, Germany, EU, Turkey, Spain, Mexico, Netherlands, Switzerland, Bulgaria, UK, Ecuador, Dominican Republic, Norway, India, China, Greece, Egypt, Poland, Bolivia, Peru, Lutuania, USA,EU,AUS, IsraeI, USA, Portugal, Serbia, Colombia, Nicaragua, New Zealand, Sri Lanka, Belgium, Iran, Thailand, Romania, Slovenia, Czech republic, Extra-EU, Chile, South Africa, Kazakhstan, Hungary, Russia, Finland, Cyprus, Denmark, Nepal, Togo, Benin, Austria, Estonia, Croatia, Pakistan, Canada, Albania, Malta, Uzbekistan, Japan, Brazil, Vietnam, Madagascar**
COO by continents	Unknown, Asia, Europe, North America, South America, Africa, Australia/Oceania
FX_L2	Algae and prokaryotes organisms, Fruit used as fruit, Fruit / vegetable juices and nectars, Processed fruit products, Stems/stalks eaten as vegetables, Infant and follow-on formulae Food products for young population, Ready-to-eat meal for infants and young children, Processed cereal-based food for infants and young children, Cereals and cereal primary derivatives, Herbs and edible flowers Legumes, Legumes with pods, Root and tuber vegetables (excluding starchy- and sugar-), Mammals and birds meat, Nuts, oilseeds and oilfruits, Flowering brassica, Leafy vegetables, Coffee, cocoa, tea and herbal ingredients, Milk and dairy products, Cheese, Unprocessed eggs , Fruiting vegetables, Spices, Sprouts, shoots and similar, Fungi, mosses and lichens, Bulb vegetables Sugar and other sweetening ingredients (excluding intensive sweeteners), Processed or preserved vegetables and similar, Milk, whey and cream, Animal and vegetable fats/oils, Starchy roots and tubers, Wine and wine-like drinks, Savoury extracts and sauce ingredients
code_FX	A00VA, A04RK, A0BX9, A01ML, A00RR, A03PY, A03PV, A03RC, A03QX, A000K, A00VQ, A04RG, A00PB, A00QF, A0EYH, A04RH, A00FL, A00KR, A03GH, A02LR, A02QE, A04NY, A00HN, A014K, A016S, A00SF, A00TC , A00GX, A00FY, A04PA, A00ZA, A04NN, A00XZ, A0F3D, A0BX10, A00JA, A00ZS, A03MS, A0EQE
Food matrices	Algae and prokaryotes organisms, Apples, Apricot, Artichoke, Asparagus, Avocados, Baby Follow-on formulae, Baby food, Baby foods other than processed cereal-based foods, Baby Processed cereal-based foods, Bananas, Barley, Basil (holy, sweet), Basil and edible flowers, Beans, Beans (dry), Beans with pods, Beetroots, Bovine meat, Brazil nuts, Broccoli, Brussels sprouts, buckwheat, Buckwheat and other pseudo-cereals, Cabbage, Camomille flowers, Carrots, Castor beans, Cattle, cattle milk and milk products, Cauliflowers, Celery, Cereals, not specified, Chamomile, Chards, Chards/beet leaves, Cheese, Cherries, Chicken eggs, Chili peppers, Citrus fruits, Cocoa beans, Coconuts, Coffee beans, Coriander leaves, Coriander seed, Courgettes, Cresses and other sprouts and shoots, Cucumbers, Cultivated fungi, Cumin seed, Curry leaves, Dates, Dried apricots, Dried herbal infusions, Dry parsley, Eggplant, Eggs, Endive lettuce, Fennel seed, Fennels, Figs, French beans, Fresh herbs, Fungi, not specified, Garlic, Ginger, Goji berries, Granate, apples/pomegranates, Grape leaves and similar species, Grapefruit, Grapes, Green lentil, Guavas, Head cabbage, Hemp seeds, Herbal infusions, Herbs and edible flowers, Honey, Horseradishes, Kiwi, Kohlrabies, Lamb's lettuces, Land cresses, Laurel/bay leave, Leafy brassica, not specified, Leek, Lemons, Lentils, Lentils, dry, Lettuces, Limes, Linseeds, Lychee, Maize, Mandarins, Meat, Melons, Milk (sheep) , Milk and milk products, Mint, Oats, Oil seeds, Olive oil, Onions, Oranges, Oregano, Papaya, Parsley , Passion fruit, Peaches, Pears, Peas, Peppercorn (black, green and white), Peppers, Pine nuts, Pineapples, Plums, Pomegranates, Poppy seeds, Porcine meat, Potatoes,

	Poultry, Poultry meat, Pulses (dry), not specified, Pumpkin seeds, Pumpkins, Radishes, Rapeseeds, Raspberries, Red wine, Rice, rocket, Rucola, Rooibos leaves, Rucola, Rye, Rye grain, Sage, Scarole, Sea beat, Small fruit and berries, Soyabeans, Spices, Spinach, Strawberries, Sweet bell peppers, Sweet potato, Swiss chard, Table grapes, Table olives, Tea, Tea (green, black), Tea leaves, Thyme, Tomato, Turnips, Vanilla pods, vegetable marrow, Vegetables, Wheat, Wheat flour, Wheat grain, White wine, Wild fungi, Wine, Wine grapes
Chemicals Substances	Methacrifos, Diuron, Prometryn, Terbuthylazine, Cadmium, Lead, Chlorate, Chlorpyrifos, Chlorpropham, Azinophos methyl , Dimethoate, Diphenylamine (DPA), Carbaryl, Carbendazim, Boscalid, Iprodione, 2-phenylphenol, Thiabendazole, Acetamiprid, Cypermethrin, Spinosad, Fenhexamid, Dithiocarbamate, Glyphosate, Bromide ion, Permethrin BAC (RD), DDAC, DDT (RD), Endosulfans, Hexachlorobenzene, λ-cyhalothrin, Fosetyl-Al , Trimethyl-sulfonium cation, Difenoconazole, Chlormequate, Tridemorph, Gibberellic acid, Imazalil, Fenpropimorph, Dikegulac, Dichlorvos, Tetramethrin, Cyprodinil, Cobalt , Chromium, Nitrate, Copper, Manganese, Zinc, Iron, Epoxiconazole, Nicotine, Profenofos , Propargite, PCB 105, PCB 138, PCB 118, PCB 157, PCB 180, PCB 81 , PCB 77, PCB 101, PCB 153 PCB 126, PCB 28, PCB 156, DDE o,p', PCB 52 , α-HCH, DDD p,p', DDT o,p', β-HCH, DDE p,p', Dieldrin, DDT p,p', Endrin, Lindane, Heptachlor, Aldrin, 3∑HBCD, PCDD/F, DL-PCB, 6∑NDL-PCB, Mercury, Arsenic, Anthraquinone, Carbofuran (sum of carbofuran (including any carbofuran generated from carbosulfan, benfuracarb or furathiocarb) and 3-OH carbofuran expressed as carbofuran), Phenthoate, Haloxyfop , PCB 169, PCB 114, Fludioxonil, Metalaxyl, Methoxyfenozide, Imidacloprid, Bifenazate, Fluopicolide, Propamocarb, PCB 167, α- endosulfan, Methoxychlor, ∑DDTs, ∑OCP residues, Chlorothalonil, Pyrimethanil, Pirimiphos-methyl, Chlordane (sum of cis- and trans-chlordane), Flutriafol, Cyproconazole, Hexaconazole, Clothianidin, Thiamethoxam, Aldrin and Dieldrin (Aldrin and dieldrin combined expressed as dieldrin), Piperonyl butoxide, Monocrotophos, Kresoxim-methyl, Propiconazole (sum of isomers), Tricyclazole, Quinalphos, Clofentezine, Thiophanate methyl, Quinoxyfen, Flonicamid , Flupyradifurone, Malathion, Propoxur, Prometon, Phoxim, Tebufenpyrad, Azoxystrobin, Fenvalerate, Metrafenone, Biphenyl, Mandipropamid, Formetanate, Spiroxamine, Penconazole, Dimethomorph, Pyraclostrobin, Pendimethalin, Propyzamide , Prothioconazole: prothioconazole-desthio (sum of isomers), Coumaphos, Diazinon, Amitraz, Thiacloprid, Thymol, Isoproturon, Methiadathion , Fenitrothion, 2,4-D (sum of 2,4-D, its salts, its esters and its conjugates, expressed as 2,4-D), Triadimenol , Isofenphos-methyl, Fenthion, Deltamethrin, Phosmet, Bromopropylate, Fluazifop, Methamidophos, Acephate, Prosulfocarb, Carboxin, Fenazaquin , Tetradifon, Dicofol , Trifluralin, Bitertanol, Azadirachtin, Pyrethrins, Free sulphite, Total sulphite , Pyrethrins (Cinerine 1), Triflumizole, Fenpyrazamine, Phorate, Nitrite, β-endosulfan, Tebuconazole, Fenpropidin , Dinotefuran, Chlorfluazuron, Ametryn, Fenobucarb, Fipronil , Hexaflumuron , Buprofezin, Fenbuconazole, Lufenuron, Teflubenzuron, Quintozene , Rotenone, Pyridaben, Chlorantraniliprole, Tolfenpyrad, Flubendiamide, MCPA, 2-Naphthoxyacetic acid, Esfenvalerate, Cyfluthrin, Triasulfuron, Veratridine, Diflubenzuron, Pyrethrins (Cinerine 2), Pyrethrins (Jasmoline 1), Ethephon, Atrazine desethyl, Aluminium, Isoprothiolane, Iprovalicarb
Chemical classes	Organophosphate, Phenylamide , Triazine, Inorganic contaminants, Chlorates, Carbamate, Amine, Benzimidazole, Carboxamide, Hydroxybiphenyls, Neonicotinoid, Pyrethroid, Micro-organism derived, Hydroxyanilide , Dithiocarbamate, Quaternary ammonium compounds

	(QACs) , Organochlorine, Triazole, Morpholine, Imidazoles, Anilino-pyrimidine, Minerals, Nitrate, Plant derived, Sulfite Ester, PCB, Brominated compounds, Dioxins/Furans , PAH, Aryloxyphenoxypropionate, Phenylpyrrole, Diacylhydrazine, Hydrazine carboxylate, Benzamides, Chloronitrile, Cyclic aromatic, Strobilurin, Triazolobenzothiazole, Tetrazine, Quinolines, Pyridine compound, Butenolide, Methoxytriazine, Pyrazolium, Benzophenone, Aromatic hydrocarbon, Mandelamides, Formamidine, Dinitroaniline, Triazolinthione, Amidine, Urea, Alkylchlorophenoxy, Phenol, Benzylate, Thiocarbamate, Oxathiin, Quinazolines, Bridged diphenyl, Sulphite, Piperidines, Benzoylurea, Phenylpyrazole, Thiadiazine, Chlorophenyl, Pyridazinone , Anthranilic diamide, Pyrazole, Benzene-dicarboxamide, Aryloxyalkanoic acid, Synthetic auxin, Sulfonylurea, Phosphorothiolate
General Groups	Pesticides, Minerals, Inorganic contaminants, PCBs, Dioxins, Brominate flame retardants
Measurement type	Median, mean, range
Chemical type	Contaminant, residue

Table A3-3. Elements of the FoodEx 2 level 2 groups

FoodEx2 L2 group	Food Matrices
Algae and prokaryotes organisms	Algae and prokaryotes organisms
Animal and vegetable fats/oils	Oil seeds; Olive oil
Bulb vegetables	Garlic; Kohlrabies; Onions
Cereals and cereal primary derivatives	Barley; buckwheat; Buckwheat and other pseudo-cereals; Cereals, not specified; Maize; Oats; Rice; Rye grain; Wheat flour; Wheat grain
Cheese	Cheese
Coffee, cocoa, tea and herbal ingredients	Camomille flowers; Chamomile; Cocoa beans; Coffee beans; Dried herbal infusions; Herbal infusions; Rooibos leaves; Tea; Tea (green, black)
Flowering brassica	Broccoli; Cauliflowers
Food products for young population	Baby food
Fruit / vegetable juices and nectars	Apples ; Granate apples/pomegranates ; Oranges
Fruit used as fruit	Apples; Apricot; Avocados; Bananas; Cherries; Citrus fruits; Figs; Grapefruit; Grapes; Guavas; Kiwi; Lemons; Limes; Lychee; Mandarins; Oranges; Papaya; Passion fruit; Peaches; Pears; Pineapples; Plums; Pomegranates; Raspberries; Strawberries; Table grapes; Wine grapes
Fruiting vegetables	Chili peppers; Courgettes; Cucumbers; Eggplant; Goji berries; Melons; Peppers; Pumpkins; Sweet bell pepper; tomato
Fungi, mosses and lichens	Cultivated fungi; Fungi, not specified; Wild fungi
Herbs and edible flowers	Basil (holy, sweet); Basil and edible flowers; Celery; Coriander leaves; Curry leaves; Dry parsley; Fresh herbs; Herbs and edible flowers; Laurel/bay leave; Mint; Oregano; Parsley; Sage; Thyme
Infant and follow-on formulae	Baby Follow-on formulae
Leafy vegetables	Brussels sprouts; Cabbage; Cauliflowers; Chards; Chards/beet leaves; Endive lettuce; Grape leaves and similar species; Head cabbage; Lamb's lettuces; Land cresses; Leafy brassica, not specified; Lettuces; rocket, Rucola; Rucola; Scarole; Sea beat; Spinach; Swiss chard
Legumes	Beans ; Beans (dry) ; Lentils ; Lentils, dry ; Pulses (dry), not specified
Legumes with pod	Beans with pods; French beans; Green lentil; Peas
Mammals and birds meat	Bovine meat; Cattle; Meat; Porcine meat; Poultry; Poultry meat
Milk and dairy products	cattle milk and milk products; Milk and milk products
Milk, whey and cream	Milk (sheep)
Nuts, oilseeds and oilfruits	Brazil nuts; Castor beans; Coconuts; Hemp seeds; Linseeds; Pine nuts; Poppy seeds; Pumpkin seeds; Rapeseeds; Soyabeans

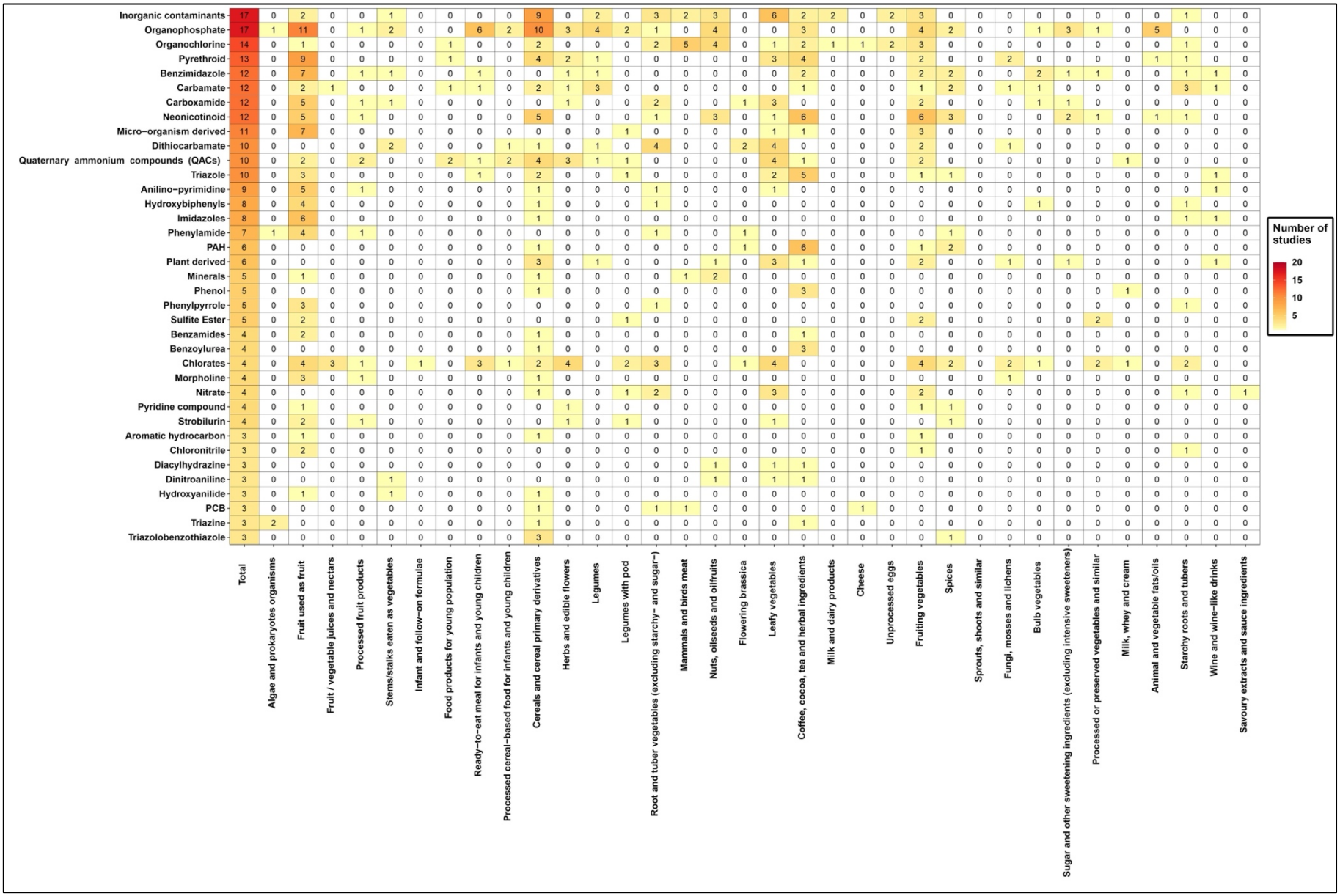
Processed cereal-based food for infants and young children	Baby Processed cereal-based foods
Processed fruit products	Apricot; Dates; Dried apricots; Figs; Kiwi; Small fruit and berries; Strawberries; Table grapes; Wine grapes
Processed or preserved vegetables and similar	Lettuces; Spinach; Sweet bell peppers; Tomato; Vegetables
Ready-to-eat meal for infants and young children	Baby foods other than processed cereal-based foods
Root and tuber vegetables (excluding starchy- and sugar-)	Beetroots; Carrots; Horseradishes; Radishes; Turnips
Savoury extracts and sauce ingredients	vegetable marrow
Spices	Coriander seed; Cumin seed; Fennel seed; Ginger; Peppercorn (black, green and white); Spices; Thyme; Vanilla pods
Sprouts, shoots and similar	Cresses and other sprouts and shoots
Starchy roots and tubers	Potatoes, Sweet potato
Stems/stalks eaten as vegetables	Artichoke; Asparagus; Fennels; Leek
Sugar and other sweetening ingredients (excluding intensive sweeteners)	Honey
Unprocessed eggs	Eggs
Wine and wine-like drinks	Red wine; White wine; Wine

Table A3-4. Elements of the chemical classes

Chemical class	Chemical substance
Alkylchlorophenoxy	2,4-D (sum of 2,4-D, its salts, its esters and its conjugates, expressed as 2,4-D)
Amidine	Amitraz
Amine	Diphenylamine (DPA)
Anilino-pyrimidine	Cyprodinil,Pyrimethanil
Anthranilic diamide	Chlorantraniliprole
Aromatic hydrocarbon	Biphenyl
Aryloxyalkanoic acid	MCPA
Aryloxyphenoxypropionate	Haloxyfop , Fluazifop
Benzamides	Fluopicolide, Propyzamide
Benzene-dicarboxamide	Flubendiamide
Benzimidazole	Carbendazim, Thiabendazole, Thiophanate methyl
Benzophenone	Metrafenone
Benzoylurea	Chlorfluazuron, Diflubenzuron, Hexaflumuron, Lufenuron, Teflubenzuron
Benzylate	Bromopropylate
Bridged diphenyl	Tetradifon
Brominated compounds	3∑HBCD
Butenolide	Flupyradifurone
Carbamate	Chlorpropham, Iprovalicarb, Carbaryl, Carbofuran (sum of carbofuran (including any carbofuran generated from carbosulfan, benfuracarb or furathiocarb) and 3-OH carbofuran expressed as carbofuran), Propamocarb, Propoxur, Fenobucarb
Carboxamide	Boscalid, Iprodione
Chlorates	Chlorate
Chloronitrile	Chlorothalonil
Chlorophenyl	Quintozene
Cyclic aromatic	Piperonyl butoxide
Diacylhydrazine	Methoxyfenozide
Dinitroaniline	Pendimethalin, Trifluralin
Dioxins/Furans	PCDD/F
Dithiocarbamate	Dithiocarbamate
Formamidine	Formetanate
Hydrazine carboxylate	Bifenazate
Hydroxyanilide	Fenhexamid
Hydroxybiphenyls	2-phenylphenol
Imidazoles	Imazalil, Triflumizole
Inorganic contaminants	Cadmium, Lead, Bromide ion, Cobalt, Mercury, Arsenic, Aluminium
Mandelamides	Mandipropamid
Methoxytriazine	Prometon
Micro-organism derived	Spinosad, Gibberellic acid
Minerals	Chromium, Copper, Manganese, Zinc, Iron
Morpholine	Tridemorph, Fenpropimorph, Spiroxamine, Dimethomorph

Neonicotinoid	Acetamiprid, Imidacloprid, Clothianidin, Thiamethoxam, Thiacloprid, Dinotefuran
Nitrate	Nitrate
Not specified	Dikegulac
Organochlorine	DDT (RD), Endosulfans, Hexachlorobenzene, DDE o,p', α-HCHDDD, p,p'DDT, o,p'DDE, β-HCH, p,p'DDT, Dieldrin, p,p'DDT (RD), α-endosulfan, ∑DDTs, ∑OCP residues, Methoxychlor, Chlordane (sum of cis- and trans-chlordane), Aldrin and Dieldrin (Aldrin and dieldrin combined expressed as dieldrin), Dicofol, Phorate, β-endosulfan, Endrin, Lindane, Heptachloraldrin
Organophosphate	Methacrifos, Chlorpyrifos, Azinophos methyl, Glyphosate, Dimethoate, Fosetyl-Al, Trimethyl-sulfonium cation, Dichlorvos, Profenofos, Phenthoate, Pirimiphos-methyl, Monocrotophos, Quinalphos, Malathion, Phoxim, Coumaphos, Diazinon, Methiadathion, Fenitrothion, Isofenphos-methyl, Fenthion, Phosmet, Methamidophos, Acephate, Ethephon
Oxathiin	Carboxin
PAH	Anthraquinone
PCB	PCB 105, PCB 138, PCB 118, PCB 157, PCB 180, PCB 81, PCB 77, PCB 101, PCB 153, PCB 126, PCB 28, PCB 156, PCB 52, DL-PCB, 6∑NDL-PCB, PCB 169, PCB 114, PCB 167
Phenylamide	Diuron,Metalaxyl
Phenylpyrazole	Fipronil
Phenylpyrrole	Fludioxonil
Phosphorothiolate	Isoprothiolane
Piperidines	Fenpropidin
Plant derived	Nicotine, Thymol, Azadirachtin, Pyrethrins, Pyrethrins (Cinerine 1), Rotenone, Veratridine, Pyrethrins (Cinerine 2), Pyrethrins (Jasmoline 1)
Pyrazole	Tolfenpyrad
Pyrazolium	Tebufenpyrad, Fenpyrazamine
Pyrethroid	Cypermethrin, Permethrin, λ-cyhalothrin, Fenvalerate, Tetramethrin, Deltamethrin, Esfenvalerate, Cyfluthrin
Pyridazinone	Pyridaben
Pyridine compound	Flonicamid
Quaternary ammonium compounds (QACs)	BAC (RD), DDAC, Chlormequate
Quinazolines	Fenazaquin
Quinolines	Quinoxyfen
Strobilurin	Kresoxim-methyl, Azoxystrobin, Pyraclostrobin
Sulfite Ester	Propargite
Sulfonylurea	Triasulfuron
Sulphite	Total sulphite, Free sulphite
Synthetic auxin	2-Naphthoxyacetic acid
Tetrazine	Clofentezine
Thiadiazine	Buprofezin
Thiocarbamate	Prosulfocarb
Triazine	Prometryn, Terbuthylazine, Ametryn, Atrazine desethyl

Triazole	Difenoconazole, Epoxiconazole, Flutriafol, Cyproconazole, Hexaconazole, Propiconazole (sum of isomers), Penconazole, Triadimenol, Bitertanol, Tebuconazole, Fenbuconazole
Triazolinthione	Prothioconazole: prothioconazole-desthio (sum of isomers)
Triazolobenzothiazole	Tricyclazole
Urea	Isoproturon


Inorganic contaminants 17 0 2 0 0 1 0 0 0 0 9 0 2 0 3 2 3 0 6 2 2 0 2 3 0 0 0 0 0 0 0 0 1 0 0
Organophosphate 17 1 11 0 1 2 0 0 6 2 10 3 4 2 1 0 4 0 0 3 0 0 0 4 2 0 0 1 3 1 0 5 0 0 0
Organochlorine 14 0 1 0 0 0 0 1 0 0 2 0 0 0 2 5 4 0 1 2 1 1 2 3 0 0 0 0 0 0 0 0 1 0 0
Pyrethroid 13 0 9 0 0 0 0 1 0 0 4 2 1 0 0 0 0 0 3 4 0 0 0 2 0 0 2 0 0 0 0 1 1 0 0
Benzimidazole 12 0 7 0 1 1 0 0 1 0 0 1 1 0 0 0 0 0 0 2 0 0 0 2 2 0 0 2 1 1 0 0 1 1 0
Carbamate 12 0 2 1 0 0 0 1 1 0 2 1 3 0 0 0 0 0 0 1 0 0 0 1 2 0 1 1 0 0 0 0 3 1 0
Carboxamide 12 0 5 0 1 1 0 0 0 0 0 1 0 0 2 0 0 1 3 0 0 0 0 2 0 0 0 1 1 0 0 0 0 0 0
Neonicotinoid 12 0 5 0 1 0 0 0 0 0 5 0 0 0 1 0 3 0 1 6 0 0 0 6 3 0 0 0 2 1 0 1 1 0 0
Micro−organism derived 11 0 7 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 1 1 0 0 0 3 0 0 0 0 0 0 0 0 0 0 0
Dithiocarbamate 10 0 0 0 0 2 0 0 0 1 1 0 1 0 4 0 0 2 4 0 0 0 0 2 0 0 1 0 0 0 0 0 0 0 0
Quaternary ammonium compounds (QACs) 10 0 2 0 2 0 0 2 1 2 4 3 1 1 0 0 0 0 4 1 0 0 0 2 0 0 0 0 0 0 1 0 0 0 0
Triazole 10 0 3 0 0 0 0 0 1 0 2 0 0 1 0 0 0 0 2 5 0 0 0 1 1 0 0 0 0 0 0 0 0 1 0
Anilino−pyrimidine 9 0 5 0 1 0 0 0 0 0 1 0 0 0 1 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0
Hydroxybiphenyls 8 0 4 0 0 0 0 0 0 0 1 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 1 0 0
Imidazoles 8 0 6 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 1 0
Phenylamide 7 1 4 0 1 0 0 0 0 0 0 0 0 0 1 0 0 1 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0
PAH 6 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 1 0 6 0 0 0 1 2 0 0 0 0 0 0 0 0 0 0
Plant derived 6 0 0 0 0 0 0 0 0 0 3 0 1 0 0 0 1 0 3 1 0 0 0 2 0 0 1 0 1 0 0 0 0 1 0
Minerals 5 0 1 0 0 0 0 0 0 0 1 0 0 0 0 1 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Phenol 5 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 3 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0
Phenylpyrrole 5 0 3 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0
Sulfite Ester 5 0 2 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 2 0 0 0 0 0 2 0 0 0 0 0
Benzamides 4 0 2 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Benzoylurea 4 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 3 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Chlorates 4 0 4 3 1 0 1 0 3 1 2 4 0 2 3 0 0 1 4 0 0 0 0 4 2 0 2 1 0 2 1 0 2 0 0
Morpholine 4 0 3 0 1 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0
Nitrate 4 0 0 0 0 0 0 0 0 0 1 0 0 1 2 0 0 0 3 0 0 0 0 2 0 0 0 0 0 0 0 0 1 0 1
Pyridine compound 4 0 1 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 1 1 0 0 0 0 0 0 0 0 0 0
Strobilurin 4 0 2 0 1 0 0 0 0 0 0 1 0 1 0 0 0 0 1 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0
Aromatic hydrocarbon 3 0 1 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0
Chloronitrile 3 0 2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 1 0 0
Diacylhydrazine 3 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 1 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
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PCB 3 0 0 0 0 0 0 0 0 0 1 0 0 0 1 1 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0
Triazine 3 2 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
Triazolobenzothiazole 3 0 0 0 0 0 0 0 0 0 3 0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0
Total
Algae and prokaryotes organisms
Fruit used as fruit
Fruit / vegetable juices and nectars
Processed fruit products
Stems/stalks eaten as vegetables
Infant and follow−on formulae
Food products for young population
Ready−to−eat meal for infants and young children
Processed cereal−based food for infants and young children
Cereals and cereal primary derivatives
Herbs and edible flowers
Legumes
Legumes with pod
Root and tuber vegetables (excluding starchy− and sugar−)
Mammals and birds meat
Nuts, oilseeds and oilfruits
Flowering brassica
Leafy vegetables
Coffee, cocoa, tea and herbal ingredients
Milk and dairy products
Cheese
Unprocessed eggs
Fruiting vegetables
Spices
Sprouts, shoots and similar
Fungi, mosses and lichens
Bulb vegetables
Sugar and other sweetening ingredients (excluding intensive sweeteners)
Processed or preserved vegetables and similar
Milk, whey and cream
Animal and vegetable fats/oils
Starchy roots and tubers
Wine and wine−like drinks
Savoury extracts and sauce ingredients
Number of studies
20
15
10
5

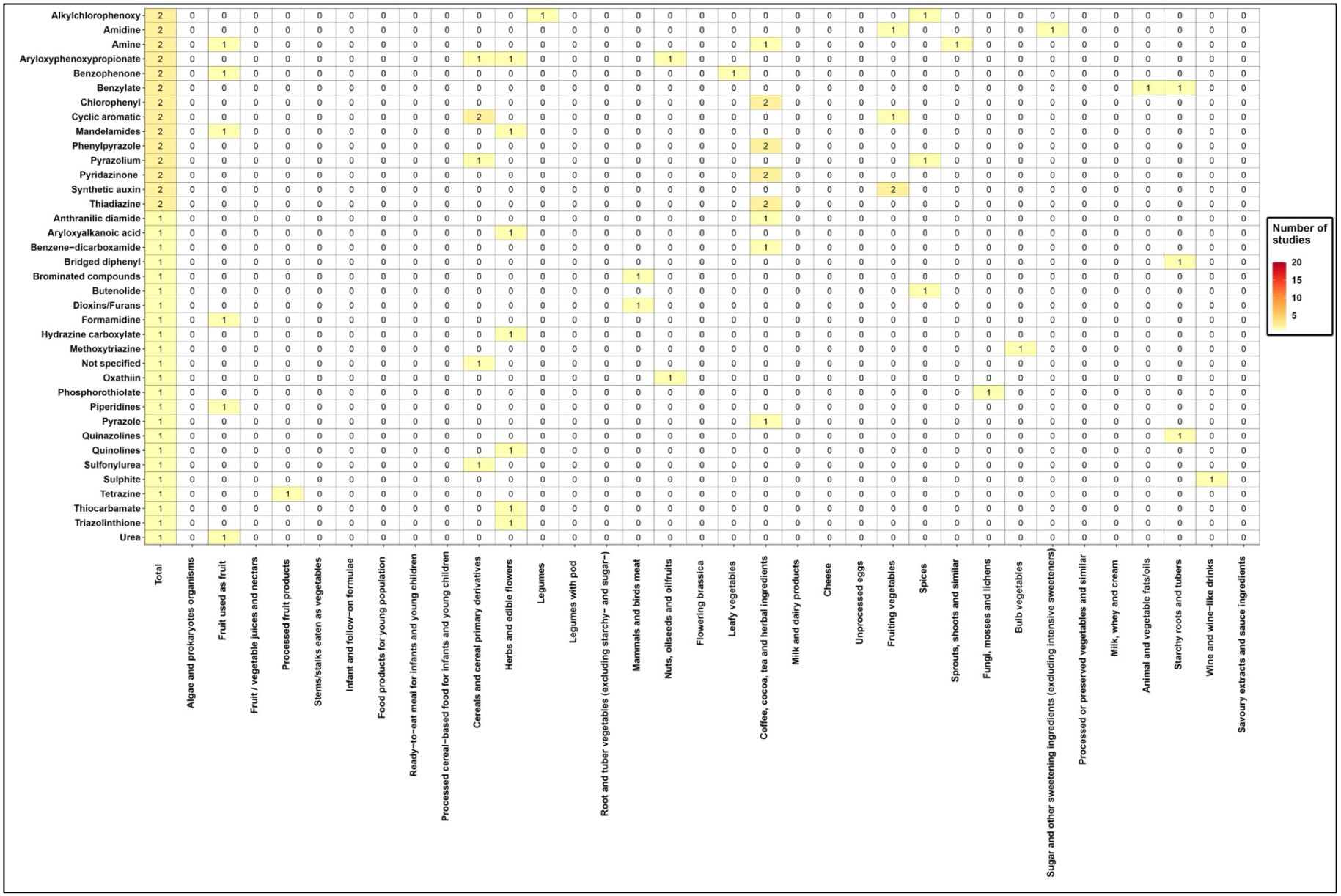

Figure A3-1. Number of studies by chemical class per FoodEx2 group

III. Appendix- Chapter 5 (A5)

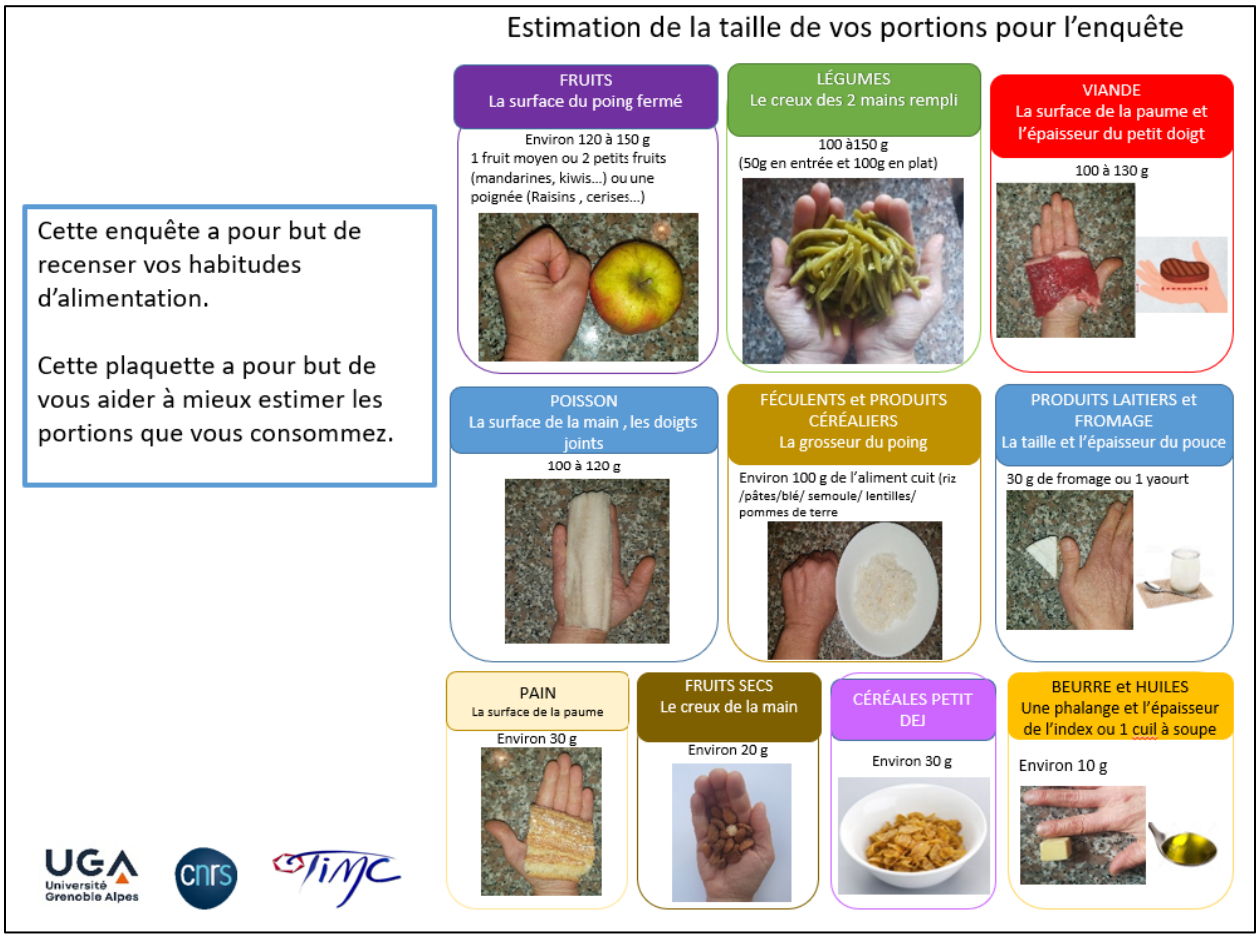

Figure A5-1. Portion size reference guide for the PEANUTS questionnaire

Table A5-1. Special diets involved in this study (with n≥3) (McRae, 2019)

Diet type	Description
Ovo-Lacto	Eliminates meat, fish and poultry but allows eggs and dairy product
Ovo-Lacto Pescitarian	Allows fish and seafood but eliminates red meat, white meat, and poultry
Flexitarian	Allows meat and other animal products occasionally
Lactose Free	Eliminates lactose, a type of sugar found in milk and milk products (Facioni et al., 2020)
Vegan	Eliminates all animal products, including meat, fish, poultry, dairy, eggs, and honey and also eliminates the use of animal goods (leather goods, wool, and silk)

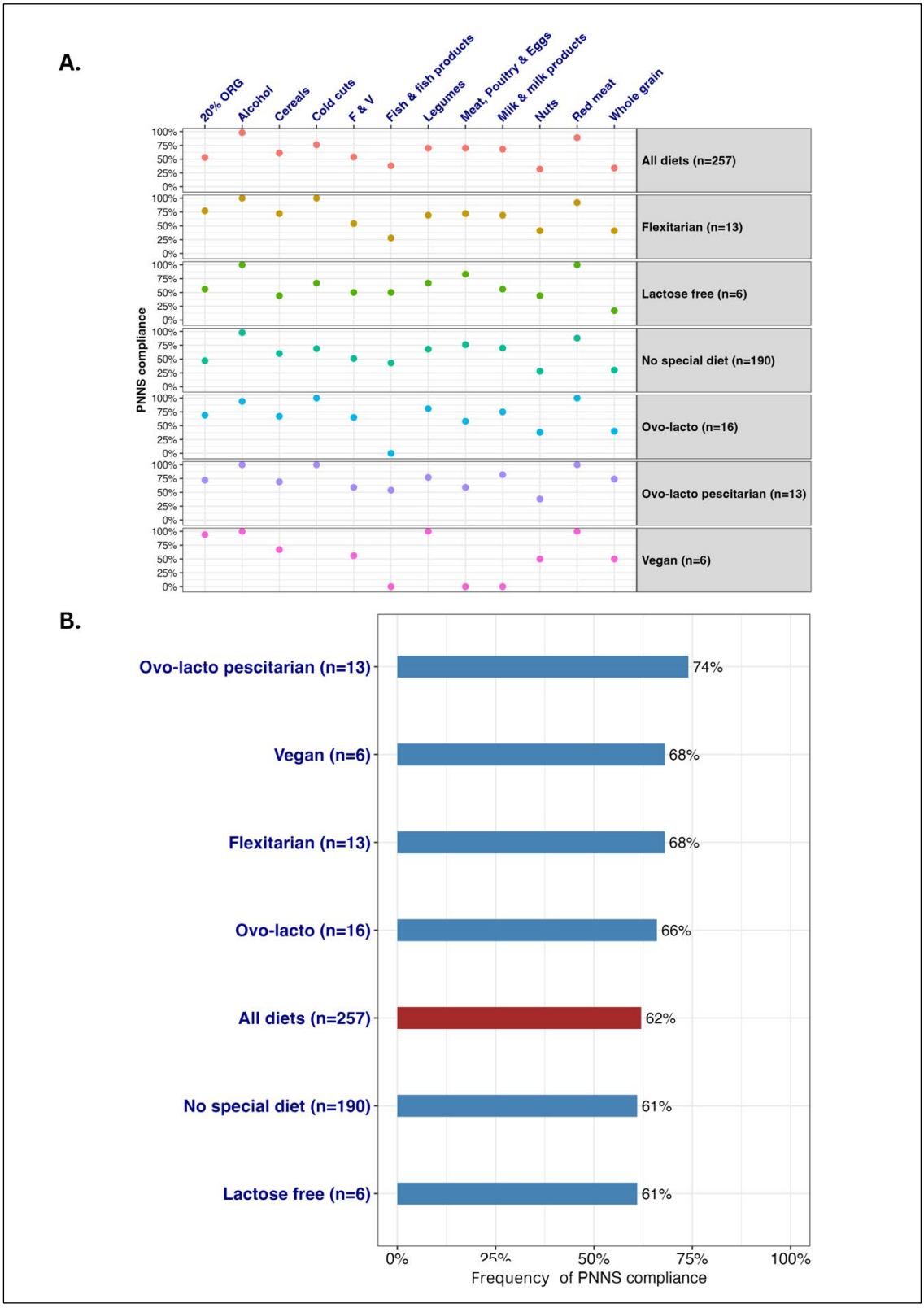

Figure A5-2. (A) Percentage PNNS compliance distribution for the total students' population and for the different diet type groups per PNNS criterion. (B) Frequency of compliance to the overall PNNS criteria tested for the total students' population and for the different diet type group

IV. Appendix- Chapter 6 (A6)

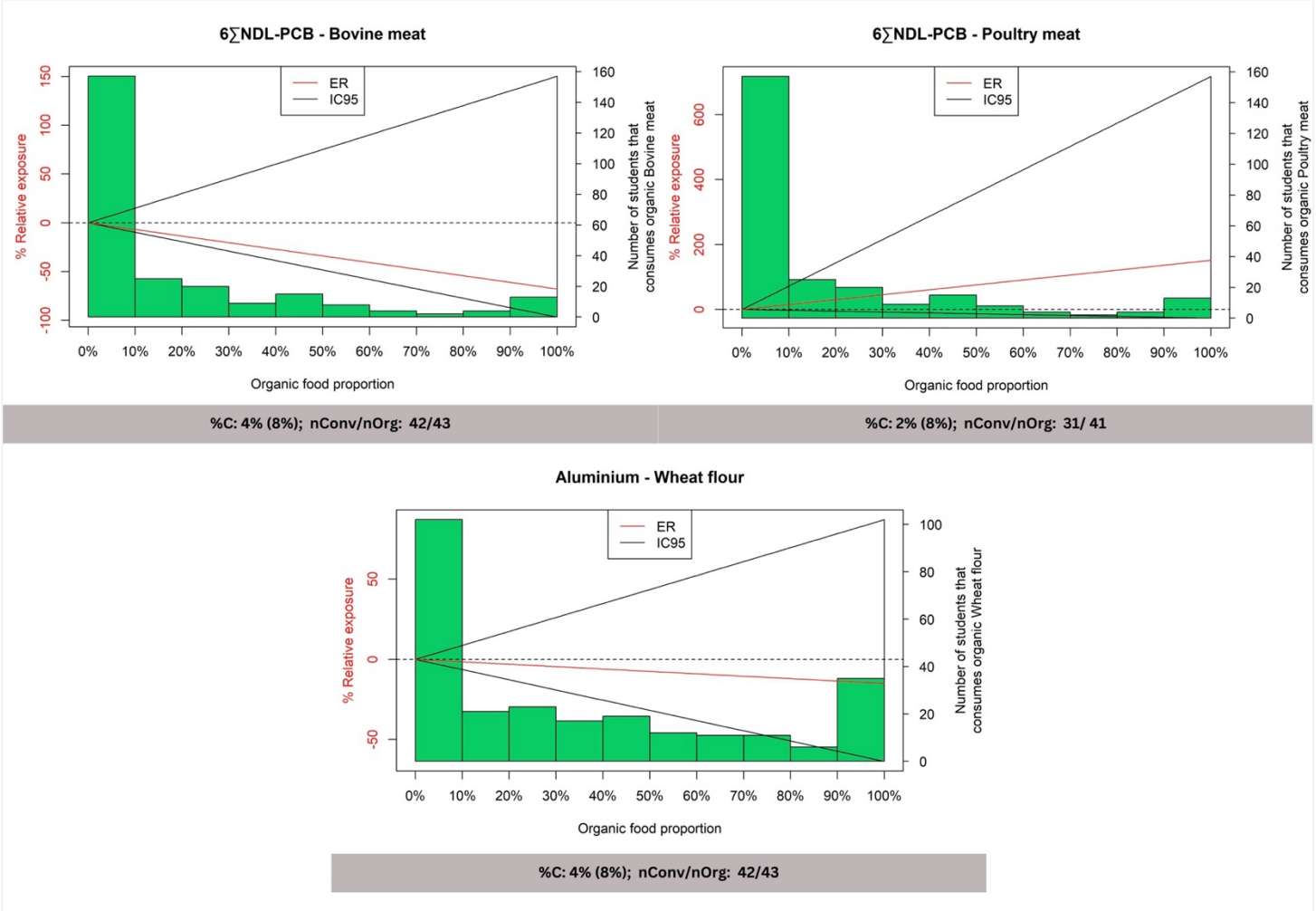

Figure A6-1. Relative exposure to the non-dioxin like substances and aluminum according to the proportion of organic food consumed for meat and wheat flour respectively; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value.

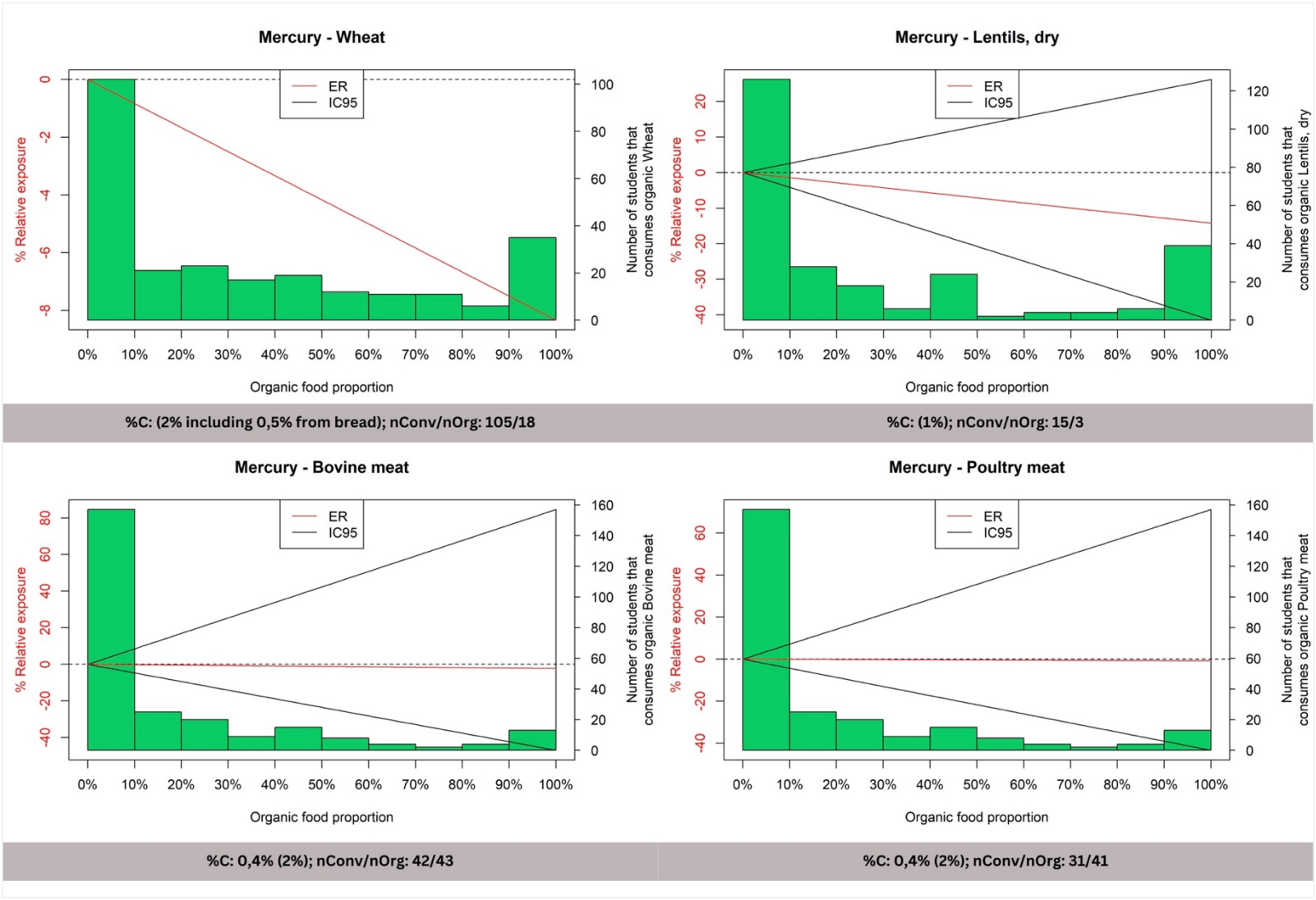

Figure A6-2. Relative exposure to mercury according to the proportion of organic food consumed for different food; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value.

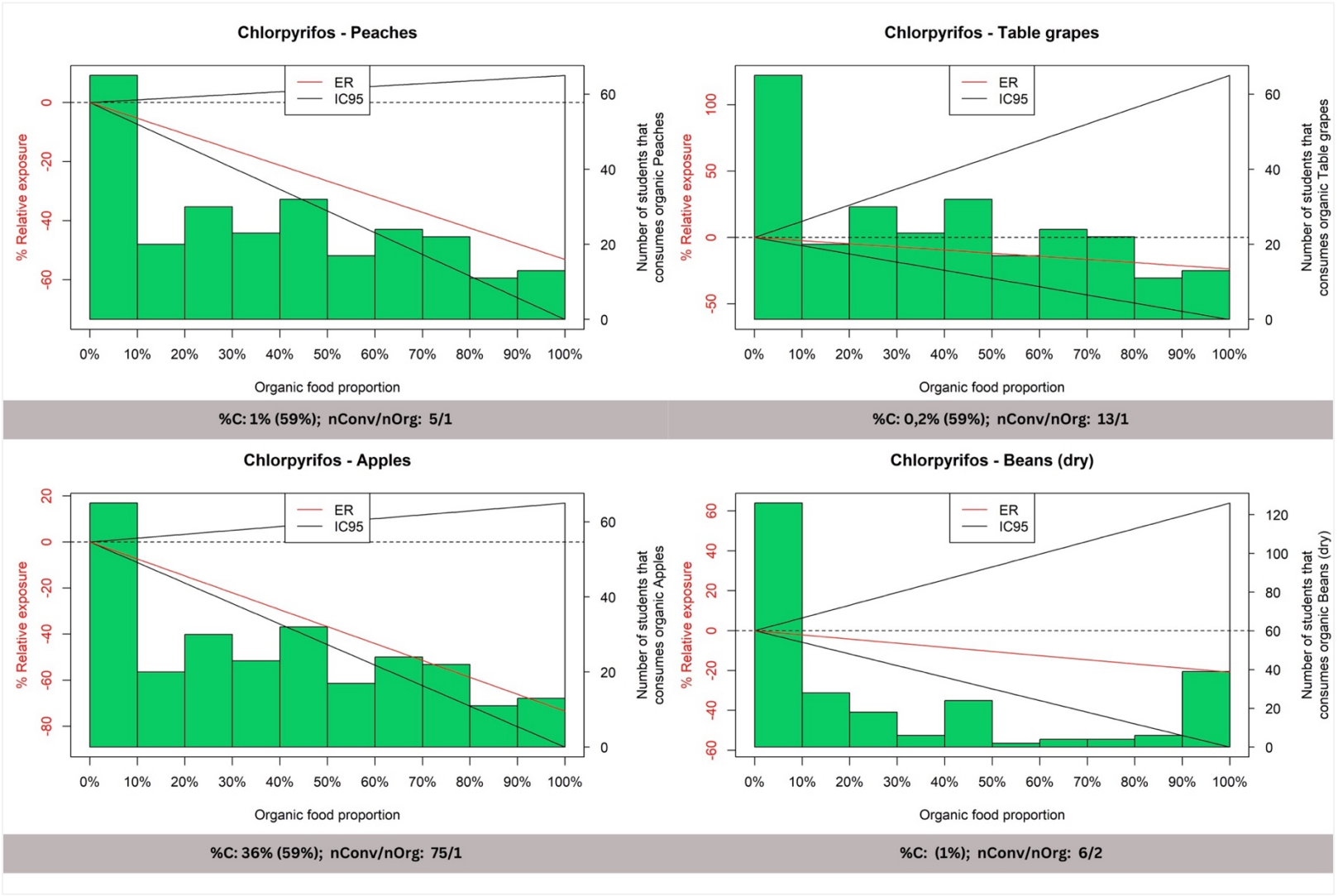

Figure A6-3. Relative exposure to chlorpyrifos according to the proportion of organic food consumed for different food; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

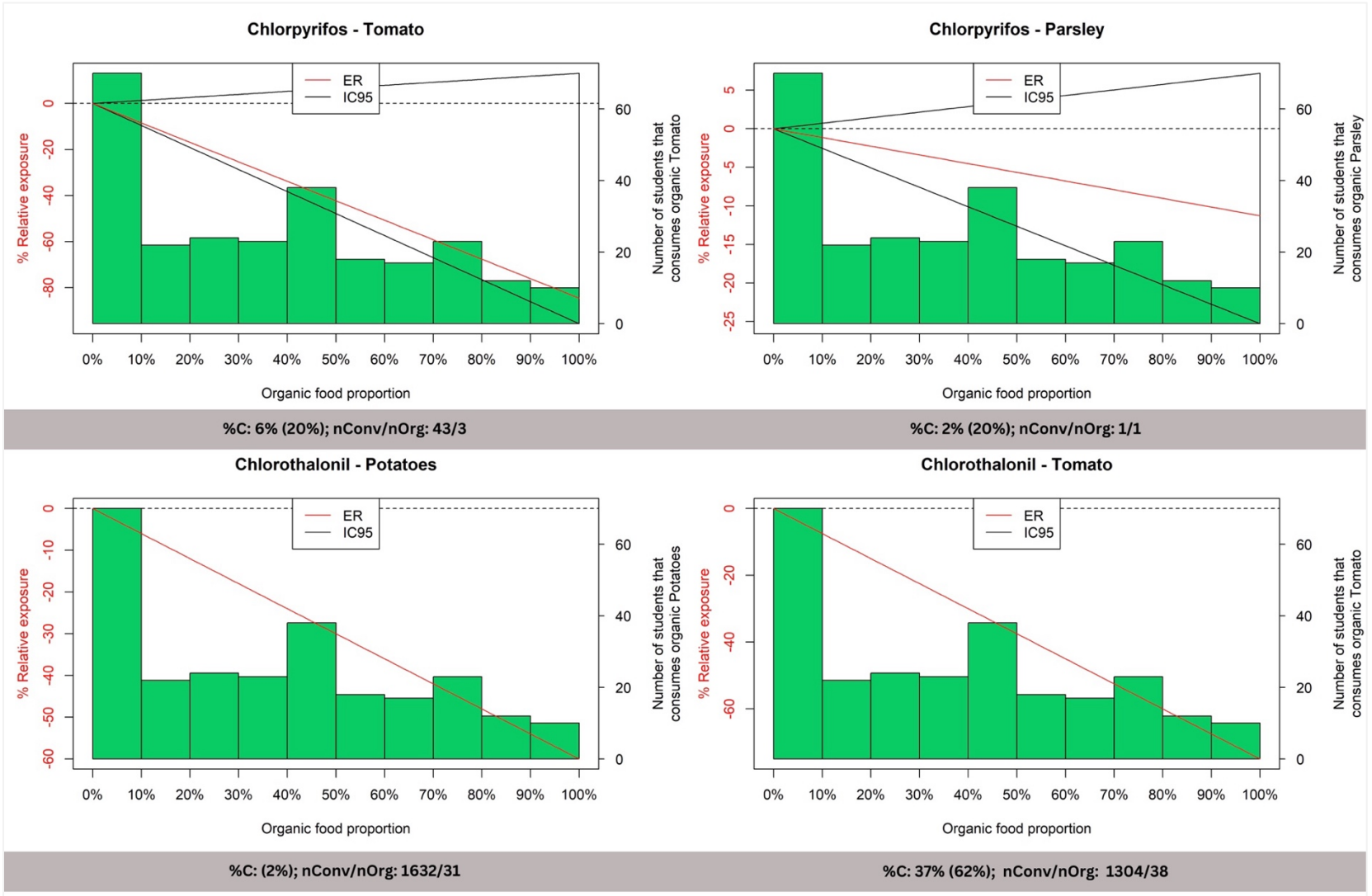

Figure A6-4. Relative exposure to chlorpyrifos and chlorothalonil according to the proportion of organic food consumed for different food; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

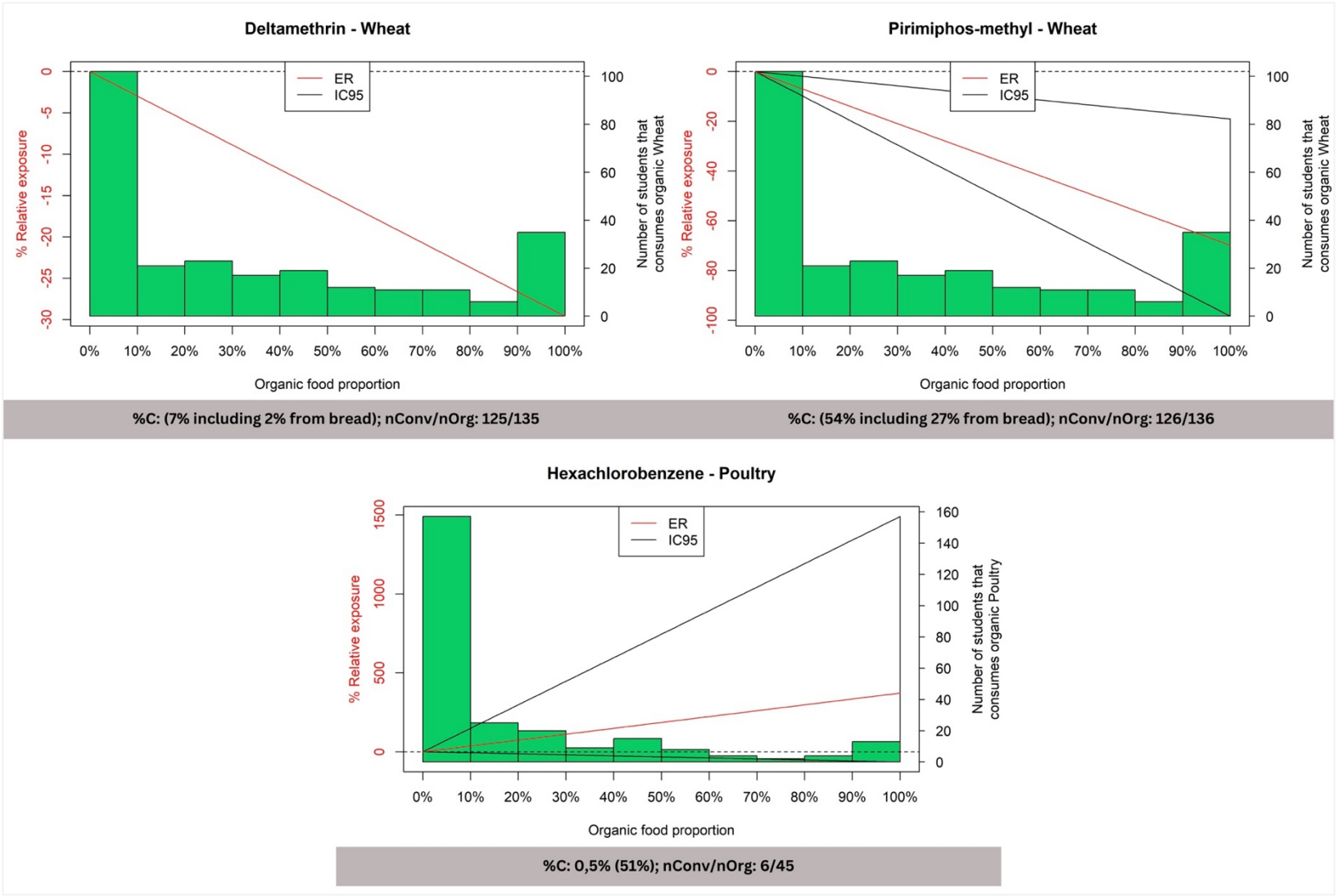

Figure A6-5. Relative exposure to different pesticides according to the proportion of organic food consumed for different food; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

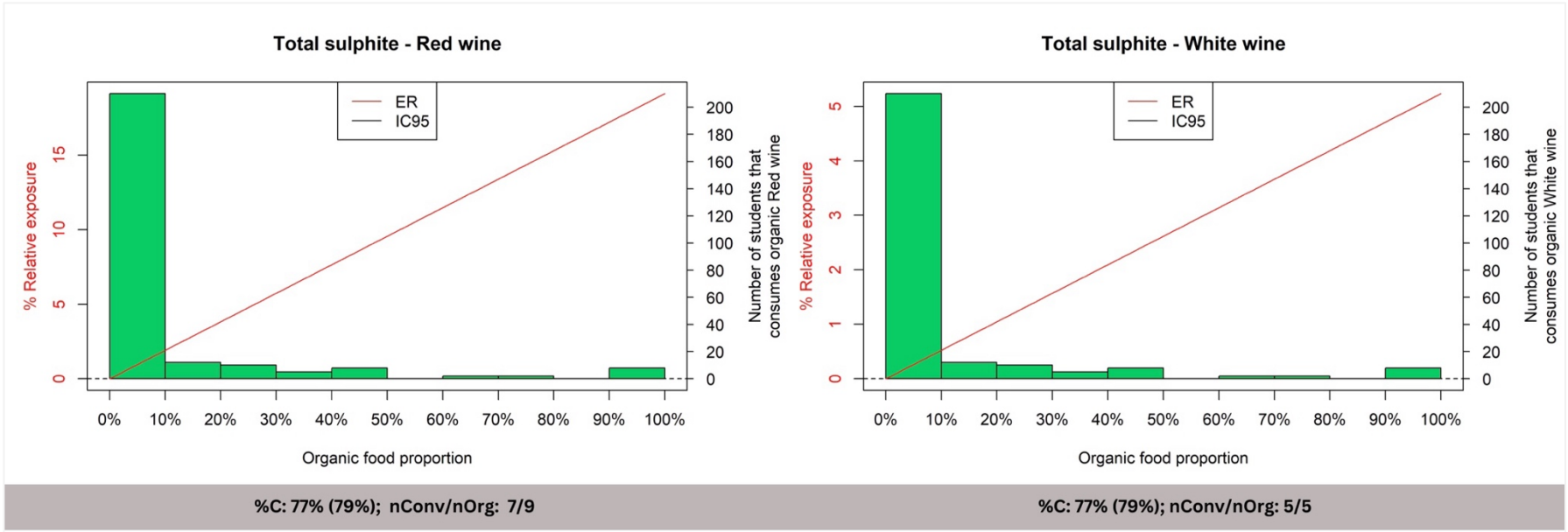

Figure A6-6. Relative exposure to total sulphite according to the proportion of organic food consumed for red and white wine; The %Relative exposure (left y-axis) is represented by the red line with the two lines of IC95, the distribution of organic food consumption (right y-axis) is represented by the histograms; %C: the percentage contribution of the specific matrix to the exposure of the chemical substance (percentage contribution of the food group), nConv/nOrg: correspond to the number of samples for both conventional and organic matrices used to generate the ER value

Title: Evaluation of human exposure to environmental pollutants through food

ABSTRACT: The main challenges linked to food include guaranteeing an optimal diet in terms of nutrition, health safety and environmental impact. Indeed, food, as a component of the exposome, is a major source of exposure to environmental pollutants for the general population. The aim of this thesis was to provide some answers to the above-mentioned challenges by: (1) developing a methodology to assess the impact of organic vs. conventional food consumption on consumer exposure to environmental pollutants in Europe, and (2) assessing the nutritional quality of students at the Université Grenoble Alpes (UGA) via a score based on the Programme National Nutrition Santé (PNNS), while studying the impact of food insecurity on adherence to these national dietary recommendations, as well as the levels of exposure of this population to certain environmental pollutants. To achieve these objectives, a database was first created to collect data from the literature on contamination levels of organically produced foods in Europe, as no comprehensive databases were available. Secondly, as the data in the database had a number of limitations that prevented the direct calculation of exposure levels from the available data, an Excess Ratio (ER) approach was developed to assess the impact of organic food consumption on the population's level of exposure to chemicals, compared to the consumption of conventionally farmed food. In addition, a population of UGA students was studied and their nutritional adequacy to the PNNS was assessed using a score we developed. The association between food insecurity and PNNS adherence was also examined. In addition, the dietary exposure profile of students was assessed using the French Total Diet Study (EAT2) contamination database for certain substances (data provided by ANSES), and the methodology developed to assess the impact of organic food consumption was applied to this population. Particular emphasis was placed on dimethoate, cadmium, lead, inorganic arsenic and dioxins. In terms of diet quality, the overall compliance rate of students with PNNS guidelines was 62%, with food-insecure students showing lower compliance for fruit and vegetables ($p = 0.004$), alcohol ($p = 0.005$) and higher compliance for meat, poultry and eggs ($p = 0.021$) and wholegrain cereals ($p = 0.01$). This thesis led to the development of a method that overcame some of the limitations of currently available data, making it possible to assess the effect of organic food consumption. This thesis also serves as an illustrative example of multi-faceted research that addresses several challenges related to food practices and agricultural production.

Keywords: Organic food, chemicals, contaminants, residues, food insecurity, PNNS score, UGA students

Titre : Évaluation de l'exposition humaine aux polluants de l'environnement par l'alimentation

RÉSUMÉ : Les principaux défis liés à l'alimentation comprennent la garantie d'un régime alimentaire optimal sur le plan nutritionnel, en termes de sécurité sanitaire mais aussi en terme d'impact environnemental. En effet, l'alimentation, en tant que composante de l'exposome, est une source majeure d'exposition aux polluants environnementaux pour la population en général. Cette thèse a eu pour objectif d'apporter quelques éléments de réponse sur les défis pré-cités en : (1) développant une méthodologie permettant d'évaluer l'impact de la consommation d'aliments biologiques par rapport aux aliments conventionnels sur l'exposition des consommateurs aux polluants environnementaux en Europe, et (2) en évaluant la qualité nutritionnelle des étudiants de l'Université Grenoble Alpes (UGA) via un score basé sur le Programme National Nutrition Santé (PNNS) tout en étudiant l'impact de l'insécurité alimentaire sur l'adhésion à ces recommandations diététiques nationales ainsi que les niveaux d'expositions de cette population à certains polluants environnementaux. Pour atteindre ces objectifs, une base de données a d'abord été créée pour recueillir les données de la littérature sur les niveaux de contamination des aliments produits en agriculture biologique en Europe, car il n'existait pas de bases de données exhaustives disponibles. Ensuite, comme les données de la base présentaient un certain nombre de limites qui empêchaient le calcul direct des niveaux d'exposition à partir des données disponibles, une approche de type Excess Ratio (ER) a été développée afin d'évaluer l'impact de la consommation d'aliments biologiques sur le niveau d'exposition de la population aux produits chimiques par rapport à la consommation d'aliments issus de l'agriculture conventionnelle. De plus, une populatio n d'étudiants de l'UGA a été étudiée et leur adéquation nutritionnelle au PNNS a été évaluée à l'aide d'un score que nous avons élaboré. L'association entre l'insécurité alimentaire et l'adhésion au PNNS a également été examinée. D'autre part, le profil d'exposition alimentaire des étudiants a été évalué à l'aide de la base de données de contamination de l'Étude de l'alimentation totale française (EAT2) pour certaines substances (données fournies par l'ANSES), et la méthodologie développée pour évaluer l'impact de la consommation d'aliments biologiques a été appliquée à cette population. Un focus particulier a été fait sur le diméthoate, le cadmium, le plomb, l'arsenic inorganique et les dioxines. En termes de qualité de l'alimentation, le taux global de conformité des étudiants aux lignes directrices du PNNS était de 62%, avec des étudiants en situation d'insécurité alimentaire présentant une conformité plus faible pour les fruits et légumes ($p = 0,004$), l'alcool ($p = 0,005$) et une conformité plus élevée pour la viande, la volaille et les œufs ($p = 0,021$) et les céréales complètes ($p = 0,01$). Cette thèse a permis le développement d'une méthode qui a permis de surmonter certaines limites des données actuellement disponibles, permettant d'évaluer l'effet de la consommation d'aliments biologiques. Cette thèse sert également d'exemple illustratif d'une recherche à multiples facettes qui aborde plusieurs défis liés aux pratiques alimentaires et à la production agricole.

Mots clés : Alimentation biologique, produits chimiques, contaminants, résidus, insécurité alimentaire, score PNNS, étudiants de l'UGA