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ABBREVIATIONS:

ADI, acceptable daily intake;

BMI, body max index;

CVUA, Chemisches und Veterinäruntersuchungsamt;

EFSA, European Food Safety Authority;

Ei,j, estimated daily exposure to pesticide j for individual i

EU, European Union;

IQR, interquartile range;

MRL, maximum residue level;

Org-FFQ, organic semi-quantitative food frequency questionnaire;

SD, standard deviation;

Abstract

Purpose: To examine dietary exposure to 25 pesticide residues in several diet groups including omnivores, pesco-vegetarians, vegetarians and vegans while accounting for the farming system (organic or conventional) of plant-based foods consumed.

Methods: Organic and conventional consumption data in combination with data on pesticide residues in plant-based foods were used to derive estimated dietary exposure to pesticide residues. Pesticide residue exposure was estimated based on observed data, and using two scenarios simulated for 100%-conventional and 100%-organic diets in 33,018 omnivores, 555 pesco-vegetarians, 501 vegetarians and 368 vegans from the NutriNet-Santé study. Pesticide residue exposure across groups was compared using Kruskal-Wallis tests.

Results: Exposure levels varied across diet groups depending on the pesticide studied. The highest exposure was observed for imazalil in all groups. Vegetarians appeared to be less exposed to the studied pesticides overall. Compared to omnivores – apart from pesticides authorised in organic farming – vegetarians had lowest exposure. The 100%-conventional scenario led to a sharp increase in exposure to pesticide residues, except for pesticides allowed in organic farming and conversely for the 100%-organic scenario.

Conclusions: Despite their high plant-based product consumption, vegetarians were less exposed to synthetic pesticides than omnivores, due to their greater propensity to consume organic.

SHORT TITLE: Dietary exposure to pesticide residues in vegetarians

KEYWORDS: pesticide exposure estimation; vegetarian diets; organic food, environmental exposure; pesticide residues

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ETHICS, CONSENT, PERMISSIONS

Informed consent has been provided from all the participants into the NutriNet-Santé cohort study. The study is conducted in accordance with the Declaration of Helsinki and all procedures have been approved by the Institutional Review Board of the French Institute for Health and Medical Research (IRB Inserm 0000388FWA00005831) and the Commission Nationale de l'Informatique et des Libertés (CNIL 908,450 and 909,216). This study is registered in ClinicalTrials.gov (NCT03335644).

Introduction

Vegetarian diets are characterised by the avoidance of meat and meat products [1]. A broad spectrum of subtypes exist that include individuals who consume fish, seafood, eggs and dairy products (pesco-vegetarians), individuals who consume eggs and dairy products (lacto-ovo-vegetarians – who can also be classified as standard vegetarians) as well as individuals who exclude any animal-derived products from their diet (vegans).

While there has been growing interest in research on the nutrient intake adequacy [2,3] and possible related health benefits of vegetarian patterns [1,4], only few studies have investigated dietary pesticide residue exposure in various vegetarian groups compared to omnivores. Pesticides are widely used for pest control, and include a large range of diverse compounds such as organophosphates, carbamates, pyrethroids, and neonicotinoids. Pesticides and their residues include active substances, as well as their metabolites and their degradation products, currently or formerly (banned by regulations) employed for farming. With regard to active substances allowed for pest control in organic farming, they are much less numerous and of lower toxicity [5,6].

Diet is considered to be the major contributor to pesticide exposure in the general population [7–9], and pesticide residues are found in plant-based foods and to a lesser extent in animal products [10]. Whereas occupational exposure to pesticides has been linked to several human diseases including cancers, neurodevelopmental abnormalities as well as neuro-degenerative and reproductive diseases, respiratory, and metabolic disorders [11,12], potential effects of chronic low-dose dietary exposure are still poorly documented [13,14].

Due to their high intakes of plant-derived products, vegetarians, and vegans in particular, are potentially at higher exposure risk than the omnivorous population [10,15]. Given the considerable growth of consumers adopting vegetarian diets worldwide in the recent decades [16] as well as the general scientific consensus on the need to a widespread global shift to more plant-based diets to reach sustainable development goals [17], assessing chronic dietary exposure of pesticide residues from plant products and associated health risks has central public health implications [18].

Exposure to various environmental contaminants (including organic pollutants, mycotoxins and trace elements) have been estimated by our group using NutriNet-Santé data [19]. However, to our knowledge, up until now only one study in France, based on diet survey data from 1999, has investigated the pesticide residue exposure of five diets (omnivorous, lacto-vegetarian, ovo-lacto-vegetarian, pesco-lacto-vegetarian and vegan), based on a relatively small sample of individuals. Their findings suggest that vegetarians may be more exposed to pesticides (except banned organochlorines) than the general population [20], however this study did not account for the farming system (organic *vs* conventional) of food consumed.

Vegetarianism and its avatars have been positively linked to several specific lifestyle-related factors, including organic food preferences [21–23]. This may shape the overall dietary pesticide exposure and its nature, since *e.g.* the latest European Food Safety Authority (EFSA) report on pesticides in food indicates that organic food products contain less pesticide residues than their conventional counterparts [10]. This observation has been largely confirmed in experimental human studies where organic diets have been repeatedly associated with reduced exposure to different families of pesticides in both adults and children [7,24–28].

Yet, to what extent organic food consumption may affect dietary pesticide residue exposure in vegetarian diets is largely unknown. To our knowledge, only one study, using urinary markers of pesticides, has evaluated whether a vegan/vegetarian diet was linked to increased pesticide exposure and the impact of organic food on pesticide residue exposure [29]. Still, large-scale observational studies are needed to comprehensively address the impact of organic food consumption on exposure to various classes of pesticides in omnivorous and vegetarian groups quantitively and qualitatively.

Hence, we aimed to estimate dietary exposure to 25 pesticide residues in different diet groups (omnivores, pesco-vegetarians, vegetarians and vegans) in a large sample of French adults comprising of more than 30,000 individuals, and – for the first time – to examine to what extent organic food consumption may affect dietary pesticide exposure.

Methods

The NutriNet-Santé study

The present analysis was carried out in the NutriNet-Santé study. The French NutriNet-Santé study is a web-based prospective cohort study that was launched in 2009 to investigate relationships between nutrition and health as well as determinants of dietary intakes [30]. The study is still ongoing. Study participants are Internet-using adult volunteers recruited from the general population. At enrolment and annually thereafter, participants are asked to complete a set of validated self-administered questionnaires via an online platform that query for sociodemographic, lifestyle, dietary, anthropometric and health data [30]. In addition, complementary questionnaires related to dietary behaviours, attitudes and practices as well as specific health issues are regularly sent to participants as part of their follow-up.

Ethics, consent and permissions

Informed consent was provided from all the participants into the NutriNet-Santé cohort study. The study is conducted in accordance with the Declaration of Helsinki and all procedures have been approved by the Institutional Review Board of the French Institute for Health and Medical Research

(IRB Inserm 0000388FWA00005831) and the Commission Nationale de l'Informatique et des Libertés (CNIL 908,450 and 909,216). This study is registered in ClinicalTrials.gov (NCT03335644).

Data collection

Assessment of dietary intakes and diet group definition

Dietary intakes were assessed using a semi-quantitative food frequency questionnaire (Org-FFQ)fully described elsewhere [31]. Briefly, in June 2014, participants were sent a food frequency questionnaire requesting their usual intake of 264 food and beverage items over the preceding year, as well as their organic food consumption frequency. For each item present in the questionnaire, participants were invited to report their frequency of consumption as well as the quantity consumed via the use of standard portion size or coloured validated photographs [32]. The Org-FFQ was elaborated on the basis of a previously validated frequency questionnaire [33] to which questions pertaining to organic food consumption frequency were incorporated. More precisely, for each item with organic options, participants were additionally asked to report how frequent was its consumption in organic, by choosing one of the following modalities: never, rarely, half-of-time, often or always. Nutrient intakes were derived from the NutriNet-Santé food composition table [34].

The definition of diet groups based on the Org-FFQ was previously described [35]. Briefly, participants were assigned to one of the four following diet groups: omnivores (participants who consumed meat or fish almost every day), pesco-vegetarians (participants who did not consume meat (<1 g/day) but did consume fish, dairy products and eggs), vegetarians (participants who did not consume meat (<1 g/day), fish (<1 g/day) but did consume dairy products and eggs), vegans (participants who did not consume meat (<1 g/day), fish (<1 g/day), eggs (<1 g/day), or dairy products (<1 g/day)).

The 264 Org-FFQ food items were grouped into 16 main food groups. Subcategories for fruit and vegetables were also created (dark green vegetables, red and orange vegetables, other vegetables, citrus fruits, pomaceous fruits, stone fruits, berries and other fruits).

The contribution of organic food intake (total and plant food groups) to total dietary intake was calculated as ratios. Two validated dietary scores were also calculated for each individual: the PNNS-GS2 (theoretical range $-\infty$ to 14.25), which measures adherence to the new French dietary guidelines [36] and the PANDiet score (theoretical range 0 to 100) which measures the probability of adequacy to nutrient-based dietary guidelines recently updated by de Gavelle et al [37] and includes 28 nutrients.

Assessment of sociodemographic, lifestyle and anthropometric information

Sex, age, location, education, monthly income per household unit, physical activity level (as measured by the IPAQ [38]) and smoking status were obtained from the repeated self-administered online questionnaires. Body mass index (BMI) was calculated as self-reported weight (kg) by height squared (m^2) using a validated questionnaire [39]. The most recent sociodemographic information closest to the Org-FFQ assessment was used.

Pesticide residue data

Pesticide residue data came from the Chemisches und Veterinäruntersuchungsamt (CVUA) Stuttgart database. A detailed description of its content has been published elsewhere [40]. In brief, the CVUA Stuttgart [41] is an official regional state food control and health laboratory located in Germany, the aim of which is to analyse pesticide residues and contaminants (around 1,000 substances are routinely analysed [42]) in food samples of plant origin sold in Germany (from European Union (EU) and outside). Since 2006, the CVUA Stuttgart has been appointed as an EU-Community Reference Laboratory for Pesticides using Single Residue Methods. A dedicated monitoring programme of the CVUA Stuttgart is designed to assess pesticide residues and contaminants in organic plant foods and the laboratory also provides data for the EU pesticide monitoring program.

We used pooled data from four years (2012-2015). The CVUA Stuttgart database was chosen over French government or EU agency records for its wide range of data regarding organic plant foods and its availability against national available data. Of note, German and French organic standards are based on the same EU regulations with some national features on marketed plant protection products [43].

Assessment of dietary pesticide residue exposure

Combining consumption data with pesticide residue CVUA data, we assessed dietary exposure to 25 pesticide residues. More information about the methodology used is available in the **Supplementary Material**.

The method used for data treatment of undetected and unquantified values has been previously described [44] and is based on the World Health Organization reference method described by Nougadère et al [45] (**Supplemental Figure**). The CVUA database contained variables allowing us to calculate the frequency of detection as well as the frequency of quantification. In brief, for left-censored data, when the censoring rate is higher than 60%, two scenarios are considered (upper- and lower-hypotheses). The estimated daily exposure ($E_{i,j}$) expressed per kilogram of body weight per day (µg/kg bw/d) is calculated for each pesticide j and for each individual i. In the present analysis, the upper-bound hypothesis was disregarded since it does not allow to capture pesticide exposure adequately in the case of organic agriculture [44], which is subject to strict standards. Under the upper-bound hypothesis, censored data are systematically imputed by limit of reporting values. Substituting censoring data would have led to artificially overestimate the true unobserved levels in organic produce, for which censored data are very frequent.

Twenty-five commonly used pesticides, including active substances allowed by organic standards, were retrieved from the CVUA database; the selection was based on sufficient diet coverage, acceptable daily intake (ADI) and frequency of detection above the maximum residue levels (MRL) [46]. A description of the selected pesticides is given in **Table 1**.

Pesticide	Pesticide family	Target pests[1]	ADI according to EU Pesticides Database (µg/kg bw/d)[2]	Authorisation status (EU)[2]
Acetamiprid	neonicotinoids	acaricide, miticide	25	Approved
Anthraquinone	Quinones	geese	X	Not approved since 2009
Azadirachtin†	limonoids	insecticide*	100	Approved
Azoxystrobin	aryloxypyrimidine	acaricide, miticide	200	Approved
Boscalid	carboxamide	fungicide	40	Approved
Carbendazim	carbamates	fungicide	20	Not approved Max. period of grace 31/05/2016
Chlorpropham	carbamates	herbicide plant growth regulator	50	Not Approved Withdrawal of authorisations by 8/01/2020 Max. period of grace: 8/10/2020
Chlorpyrifos	organophosphates	insecticide	1	Not Approved Withdrawal of authorisations by 16/02/2020 Max. period of grace: 16/04/2020
Cypermethrin	pyrethroids	insecticide	50	Approved
Cyprodinil	aminopyrimidine	fungicide	30	Approved
Difenoconazole	triazoles	fungicide	10	Approved
Dimethoate	organophosphates	acaricide, miticide insecticide	1[3]	Not Approved Withdrawal of authorisations by 17/01/2020 Max. period of grace: 17/07/2020
Fenhexamid	hydroxyanilides	fungicide	200	Approved
Glyphosate	phosphonic acids	herbicide	500	Approved
Imazalil	azoles	fungicide	25	Approved
Imidacloprid	neonicotinoids	insecticide	60	Approved
Iprodione	hydantoins	fungicide	20	Not Approved Withdrawal of authorisations by 5/03/2018 Max. period of grace: 5/06/2018
Malathion	organophosphates	acaricide, miticide insecticide	30	Approved
Methamidophos	organophosphates	acaricide, miticide insecticide	1	Not Approved since 2008
Profenofos	organophosphates	acaricide, miticide insecticide	30	Not Approved since 2007
Pyrethrins†	substance derived from *Chrysanthemum cinerariifolium*	acaricide, miticide insecticide	40	Approved
Spinosad†	mixture of two metabolites (spinosyns A and D) produced by *S. spinosa*	insecticide*	24	Approved
Tebuconazole	triazoles	Fungicide	30	Approved
Thiabendazole	carbamates	anthelminthic fungicide	100	Approved
lambda-Cyhalothrin	pyrethroids	insecticide	2.5	Approved

Table 1. Description of the studied pesticides
ADI, acceptable daily intake; EU, European Union
†Authorised in organic farming, https://eur-lex.europa.eu/legal-content/FR/TXT/PDF/?uri=CELEX:02008R0889-20181112&from=EN
[1]https://apps.who.int/pesticide-residues-jmpr-database/pesticide? *not retrieved from the WHO
[2]https://ec.europa.eu/food/plant/pesticides/eu-pesticides-db_en
[3]https://efsa.onlinelibrary.wiley.com/doi/epdf/10.2903/j.efsa.2018.5454

Furthermore, in order to estimate the combined dietary exposure to all of the studied pesticide residues (j), we created an aggregated indicator for each individual (i), using the following formula:

$$\sum_{j=1}^{24} \frac{E(j)}{ADI(j)}$$

ADI were retrieved from the EU pesticide database [47]. Of note, anthraquinone was not included in the indicator due to the absence of ADI for this molecule.

Statistical analysis

Sociodemographic, lifestyle, and dietary characteristics are presented by diet group, as mean and standard deviation (SD) for normally distributed variables, median and interquartile range (IQR) for skewed variables, and percentage for categorical variables. Normality was evaluated using Kolmogorov–Smirnov test, Q–Q plots and histograms. Sociodemographic, lifestyle, and dietary characteristics were compared according to diet group, using Kruskal-Wallis for continuous variables and Chi² tests for categorical variables.

Dietary pesticide residue exposure by diet group are presented as mean (SD).

The contribution of each plant food group to the estimated dietary pesticide residue exposure was calculated by diet group. Associations between pesticide residue exposure and plant food groups were calculated using partial Spearman's correlations. The estimated daily exposure to pesticide residues from observed conventional or organic food consumption, by diet group, was also assessed separately.

To determine to which extent consumption of organic products affected the estimated exposure, several simulated scenarios were carried out. For each pesticide residue and each diet group, we computed 100%-conventional and 100%-organic scenarios, by allocating conventional and organic pesticide residue values (respectively) to all plant ingredients consumed by the individual.

As explained above, the frequency modalities of the 5-point scale were translated into percentages to retrieve organic food consumption from each food item consumption. We also derived conventional and organic intakes from the Org-FFQ, by attributing the percentage 5% instead of 25% to the aforementioned 'rarely' modality to investigate the magnitude of the effect of the arbitrary allocation of the 25% on the total estimated pesticide residue exposure.

Pesticide residue exposure across diet groups were compared using Kruskal-Wallis tests.

Finally, a multivariable analysis of covariance (ANCOVA) was performed (using observed margins) to test for differences between diet groups while accounting for age, sex and energy intake. The adjustment for energy intake was carried out using the residual method [48].

Two-sided tests were considered and statistical significance was set at 5%. Data treatment and statistical analyses were performed using SAS®, version 9.4 and R, version 3.6.2.

Results

Study sample selection

In total, 37,685 participants had completed the Org-FFQ in 2014. Of these, we selected those with no missing covariates, no under/over-reporters (using cut-offs previously defined [31]), who were not living overseas and who had available data for the computation of the dietary scores. We thus retained 34,442 individuals (33,018 omnivores, 555 pesco-vegetarians, 501 vegetarians, 368 vegans) (see **Supplementary Figure 1**).

Participant characteristics

Participants' characteristics by diet group are presented in **Table 2**. The proportion of women was lowest in vegans and highest in pesco-vegetarians. Higher animal food avoidance was associated with lower age. Vegans were more likely to live in a population-dense urban unit than other diet groups. The lowest proportion of individuals with no high-school diploma was observed in vegetarians and the highest in omnivores. The more individuals avoided animal food the more likely they were to belong to the lowest income category (< 1200 euros per month). Vegans showed the highest proportion of highly physically active individuals and never-smokers.

	Diet groups			
	Omnivores n= 33,018	**Pesco-vegetarians** n= 555	**Vegetarians** n= 501	**Vegans** n= 368
Sociodemographics				
Female sex, %	75	88	84	71
Age [y], mean (SD)	53.7 (13.8)	48.7 (13.7)	41.6 (13.3)	37.8 (12.7)
Location, %				
Rural community	22	22	22	17
Urban unit with a population < 20,000 inhabitants	19	19	18	14
Urban unit with a population between 20,000 and 200,000 inhabitants	16	14	12	10
Urban unit with a population > 200,000	43	46	48	60

inhabitants				
Education, %				
< High-school diploma	21	15	11	13
High school diploma	15	17	14	18
Under-graduate	31	31	35	31
Post-graduate	33	37	40	38
Monthly income per household unit, %				
Unwilling to answer	6	7	8	13
< 1200 euros	11	20	24	29
1200–1800 euros	23	24	25	22
1801–2700 euros	27	26	24	18
> 2700 euros	32	22	20	18
Lifestyles				
Physical activity, %				
Missing	11	12	8	4
Low	19	16	15	16
Moderate	37	35	41	42
High	33	37	36	38
Ethanol intake [g/day], median (IQR)	4.16 (10.1)	1.82 (6.17)	1.48 (4.74)	0.729 (4.13)
Smoking status, %				
Never-smoker	49	44	52	55
Ex-smoker	40	45	37	33
Current smoker	11	11	12	13

Table 2. Characteristics of study participants, by diet group, N=34,442, NutriNet-Santé study[1,2]
[1]Values are means (SD), medians (IQR) or percent, as appropriate.
[2]All p-values $\leq 10^{-2}$. P-values derived from non-parametric Kruskal-Wallis tests for heterogeneity between diet groups (continuous variables) or Chi² test (categorical variables).

Vegans had the highest PANDiet score (reflecting adherence to nutrient-based guidelines) and omnivores the lowest (**Table 3**). Pesco-vegetarians had a slightly higher sPNNS-GS2 (reflecting adherence to food-based guidelines) than vegetarians and vegans. Pesco-vegetarians and vegans had the lowest BMI whereas omnivores had the highest. Mean energy intake was lowest among pesco-vegetarians and highest among omnivores. Logically, the intakes of plant-based products (whole-grain products, extra food, fruit and vegetables, starchy foods, soya-based products) increased with decreasing animal food consumption while intakes of alcohol, seafood, dairy products decreased. Of note, omnivores had higher intake of seafood than pesco-vegetarians. Omnivores showed the highest intakes of fatty and sweetened food and mixed dishes. The highest intake of eggs was observed among pesco-vegetarians. Pesco-vegetarians had also a high intake of fruit and vegetables. Overall, median organic ratios for plant-based foods and total food were much lower in omnivores compared to other diet groups, with the highest values observed among vegans.

	Omnivores	**Pesco-vegetarians**	**Vegetarians**	**Vegans**
Dietary intakes [g/d], median (IQR)				
Alcoholic beverages	50.6 (122)	22.7 (80.7)	22.0 (67.3)	11.3 (52.5)
Other fats	2.17 (3.63)	2.14 (3.96)	2.47 (4.94)	2.47 (5.94)
Non-alcoholic drinks	1638 (864.3)	1713 (1025)	1560 (890.5)	1455 (853.8)
Butter, margarine	5.00 (8.03)	1.37 (5.00)	1.59 (5.39)	0.329 (3.83)
Whole-grain products	29.1 (79.1)	59.0 (105)	57.7 (93.7)	69.3 (104)
Extra food (including snacks, chips, salted biscuits, dressing and sauces)	12.1 (14.4)	12.9 (24.45)	17.5 (25.9)	24.2 (31.2)
Fruit and vegetables (including juices and soups)	644 (460)	810 (567)	726 (564)	900 (712)
Of which fruit juices	23.9 (146)	24.7 (147)	31.3 (145)	64.3 (148)
Of which dark green vegetables	81.3 (83.5)	105 (109)	93.4 (110)	117 (135)
Of which red and orange vegetables	51.6 (56.5)	73.5 (84.5)	71.4 (81.9)	85.9 (88.0)
Of which citrus fruits	17.1 (35.5)	17.1 (47.5)	17.1 (47.5)	17.1 (62.0)
Of which pomaceous fruits	50.0 (90.9)	72.8 (135)	50.0 (107)	66.4 (139)
Of which stone fruits	31. 38 (64.4)	29.2 (62.3)	24.0 (58.5)	30.5 (81.6)
Of which berries	6.16 (17.7)	4.93 (19.0)	4.93 (17.3)	4.93 (12.3)
Of which other fruits	42.4 (63.9)	56.4 (86.5)	53.8 (88.9)	83.5 (116)
Starchy foods	153. (129)	148 (147)	188 (156)	233 (228)
Oil	15.8 (18.8)	18.5 (19.7)	17.1 (19.3)	20.6 (23.2)
Fatty and sweetened foods	57.6 (54.4)	42.1 (45.6)	484 (47.0)	31.4 (42.0)
Fast food/mixed dishes	26.2 (29.5)	15.2 (25.1)	21.7 (33.0)	11.4 (35.3)
Seafood	37.7 (41.5)	30.7 (59.7)	0.000 (0.000)	0.000 (0.000)
Dairy products	206 (242)	108 (248)	62.1 (169)	0.000 (0.000)
Eggs	8.50 (10.9)	10.4 (18.1)	7.14 (12.9)	0.000 (0.000)
Soya-based products and meat substitutes	0.000 (6.03)	52.7 (144)	102 (217)	248 (313)
Meat, poultry, processed meats	102 (88.1)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)
Organic ratios [/1], median (IQR)				

Bread	0.00 (0.25)	0.032 (0.35)	0.25 (0.49)	0.25 (0.50)
Cereals	0.17 (0.49)	0.75 (0.72)	0.72 (0.7)	0.77 (0.56)
Fruit juice	0.25 (0.5)	0.50 (0.92)	0.50 (0.79)	0.73 (0.75)
Fruit	0.25 (0.51)	0.57 (0.66)	0.61 (0.64)	0.74 (0.61)
Grains	0.00 (0.75)	1.00 (1.00)	1.00 (1.00)	1.00 (0.25)
Legumes	0.00 (0.50)	0.75 (0.75)	0.75 (0.75)	0.75 (0.5)
Dairy substitutes	0.00 (0.00)	0.75 (1.00)	0.96 (1.00)	1.00 (0.25)
Nuts	0.00 (0.50)	0.75 (1.00)	0.75 (0.75)	0.75 (0.75)
Vegetable oils	0.25 (0.75)	0.82 (0.64)	0.82 (0.70)	0.93 (0.50)
Potatoes	0.19 (0.56)	0.54 (0.87)	0.51 (0.85)	0.60 (0.71)
Soups	0.25 (0.75)	0.75 (0.75)	0.75 (0.75)	0.75 (0.75)
Meat substitutes	0.00 (0.25)	1.00 (0.50)	1.00 (0.20)	0.99 (0.2)
Vegetables	0.25 (0.57)	0.58 (0.66)	0.61 (0.65)	0.70 (0.63)
Wholegrain products	0.25 (0.63)	0.75 (0.75)	0.75 (0.72)	0.87 (0.47)
Diet-related and anthropometric characteristics				
Energy intake [kcal/d], mean (SD)	2002 (627.3)	1859 (632.7)	1874 (614.9)	1975 (671.9)
Proportion of organic food in the diet [/1], median (IQR)	0.219 (0.397)	0.579 (0.550)	0.604 (0.489)	0.714 (0.441)
PANDiet3, mean (SD)	64.8 (7.83)	69.7 (7.65)	68.7 (7.42)	73.1 (5.55)
sPNNS-GS2, mean (SD)	2.62 (3.41)	5.68 (2.19)	5.48 (2.12)	5.51 (2.06)
BMI [kg/m²], mean (SD)	24.3 (4.63)	21.7 (3.76)	22.2 (5.53)	21.7 (3.65)

Table 3. Dietary characteristics of study participants, by diet group, N=34,442, NutriNet-Santé study[1,2]
[1]Values are means (SD) or medians (IQR), as appropriate.
[2]All p-values $<5\times10^{-3}$. P-values derived from non-parametric Kruskal-Wallis tests for heterogeneity between diet groups.

Spearman correlation between pesticide exposure and plant-based foods

Supplementary Table 1 depicts the Spearman's correlations between major plant food groups and pesticide exposure levels. The strongest correlation was found between imidacloprid and fruit juice (ρ=0.77, p-value $<10^{-4}$) and the lowest between chlorpropham and meat substitutes (ρ=-0.24, p-value $<10^{-4}$) while the correlation coefficient was particularly high between glyphosate and legumes. As indicated by the aggregated indicator, the highest correlations with overall exposure to studied pesticides were observed for fruit and vegetables. Spearman's correlations between the different studied pesticides and dairy and meat substitutes were mostly negative, except for pesticides authorised in organic farming (azadirachtin, pyrethrins and spinosad). **Supplementary Table 2** shows the Spearman's correlation coefficients between specific subgroups of fruit and vegetables and pesticide exposure levels. Citrus fruit intake was strongly positively correlated with imazalil (ρ=0.58, p-value $<10^{-4}$) while stone fruit intake was positively correlated with iprodione and tebuconazole (ρ=0.61 and ρ=0.67, respectively, p-values <10^{-4}). Red and orange vegetable intake was also positively correlated with exposure to azadirachtin.

Estimated daily exposure to pesticide residues based on observed diet, by diet group

Table 4 shows the mean (SD) estimated daily exposure to pesticide residues (µg/kg bw/day) according to the different diet groups, under the lower-bound hypothesis. The highest mean $E_{i,j}$ was observed for imazalil, regardless of the diet group. Pesco-vegetarians exhibited the highest daily exposure to acetamiprid, carbendazim, and cypermethrin while omnivores showed the highest exposure to boscalid, chlorpropham, fenhexamid, imazalil, iprodione and tebuconazole. Vegans had the highest exposure to azoxystrobin, glyphosate, spinosad and thiabendazole. $E_{i,j}$ of malathion, methamidophos, profenofos were very low for all diet groups. The aggregated indicator expressing combined exposure to the studied pesticides ranged from 0.1835 (0.1406) µg/kg bw/day (vegetarians) to 0.2217 (0.1670) µg/kg bw/day (pesco-vegetarians).

	Omnivores	**Pesco-vegetarians**	**Vegetarians**	**Vegans**
Acetamiprid	0.0505 (0.0683)	0.0526 (0.0896)	0.0355 (0.0638)	0.0282 (0.0441)
Anthraquinone	0.0006 (0.0016)	0.0005 (0.0011)	0.0004 (0.0009)	0.0006 (0.0017)
Azadirachtin	0.0003 (0.0004)	0.0007 (0.0008)	0.0007 (0.0009)	0.0009 (0.0010)
Azoxystrobin	0.0426 (0.0449)	0.0372 (0.0449)	0.0409 (0.0587)	0.0615 (0.1605)
Boscalid	0.1147 (0.1049)	0.1031 (0.1110)	0.0861 (0.1099)	0.0860 (0.1080)
Carbendazim	0.0486 (0.0520)	0.0554 (0.0689)	0.0401 (0.0491)	0.0353 (0.0367)
Chlorpropham	0.0632 (0.0657)	0.0337 (0.0528)	0.0360 (0.0490)	0.0415 (0.0638)
Chlorpyrifos	0.0655 (0.0592)	0.0625 (0.0684)	0.0491 (0.0502)	0.0547 (0.0852)
Cypermethrin	0.0745 (0.0977)	0.0853 (0.1294)	0.0591 (0.0915)	0.0500 (0.0640)
Cyprodinil	0.0697 (0.0769)	0.0620 (0.0703)	0.0493 (0.0668)	0.0487 (0.0640)
Difenoconazole	0.0166 (0.0161)	0.0169 (0.0215)	0.0145 (0.0232)	0.0128 (0.0164)
Dimethoate	0.0032 (0.0035)	0.0032 (0.0040)	0.0026 (0.0033)	0.0024 (0.0033)
Fenhexamid	0.0910 (0.1302)	0.0662 (0.0942)	0.0653 (0.1344)	0.0570 (0.1253)
Glyphosate	0.0035 (0.0047)	0.0039 (0.0072)	0.0049 (0.0080)	0.0069 (0.0111)
Imazalil	0.7543 (0.9088)	0.5612 (0.8582)	0.5219 (0.7437)	0.7188 (10.834)
Imidacloprid	0.0782 (0.0731)	0.0833 (0.0834)	0.0816 (0.0868)	0.0972 (0.1118)
Iprodione	0.1306 (0.1557)	0.1082 (0.1420)	0.0873 (0.1402)	0.0921 (0.1773)
Malathion	0.0003 (0.0004)	0.0003 (0.0004)	0.0002 (0.0004)	0.0003 (0.0006)
Methamidophos	0.0003 (0.0004)	0.0002 (0.0004)	0.0002 (0.0003)	0.0003 (0.0005)
Profenofos	0.0000 (0.0000)	0.0000 (0.0000)	0.0000 (0.0000)	0.0000 (0.0001)
Pyrethrins	0.0021 (0.0019)	0.0024 (0.0022)	0.0028 (0.0024)	0.0030 (0.0027)
Spinosad	0.1367 (0.1637)	0.2349 (0.2634)	0.1956 (0.2210)	0.2401 (0.3484)
Tebuconazole	0.0320 (0.0398)	0.0262 (0.0334)	0.0219 (0.0353)	0.0231 (0.0405)
Thiabendazole	0.2703 (0.2929)	0.2098 (0.2841)	0.2120 (0.2771)	0.3069 (0.7425)
lambda-Cyhalothrin	0.0098 (0.0096)	0.0092 (0.0119)	0.0072 (0.0089)	0.0080 (0.0117)
Aggregated indicator	0.1960 (0.1425)	0.2217 (0.1670)	0.1835 (0.1406)	0.2168 (0.2439)

Table 4. Estimated daily exposure to pesticide residues (µg/kg bw/day), by diet group, observed diets, N=34,442, NutriNet-Santé study[1,2]

[1]Values are means (SD) under the lower-bound hypothesis

[2]All p-values <10^{-4}, except imidacloprid (p-value=0.14). P-values derived from non-parametric Kruskal-Wallis tests for heterogeneity between diet groups.

Estimated daily exposure to pesticide residues based on 100%-conventional and 100-organic diets, by diet group

Table 5 presents the mean $E_{i,j}$ together with SD, according to 100% conventional and organic scenarios, across diet groups. When compared to the observed values, apart from profenofos for which estimated exposure was very low and azadirachtin, spinosad and to a lower extent pyrethrins for which the inverse trend was observed, attributing conventional values to all pesticide residue values greatly increased pesticide exposure (up to almost 4 times the observed value for glyphosate in vegans). Conversely, a 100%-organic diet allowed to markedly reduce overall exposure, except for azaridachtin and spinosad in all groups, and pyrethrins among pesco-vegetarians. The reduction was particularly important for chlorpropham as well as for fenhexamid and imazalil in all groups. In both scenarios, vegans had the highest overall exposure and omnivores the lowest, as reflected by the aggregated indicator.

	Omnivores		**Pesco-vegetarians**		**Vegetarians**		**Vegans**	
	100%-conventional	100%-organic	100%-conventional	100%-organic	100%-conventional	100%-organic	100%-conventional	100%-organic
Acetamiprid	0.0790 (0.0906)	0.0016 (0.0021)	0.1223 (0.1370)	0.0021 (0.0026)	0.0918 (0.1028)	0.0018 (0.0025)	0.0835 (0.1000)	0.0022 (0.0034)
Anthraquinone	0.0007 (0.0018)	0.0000 (0.0000)	0.0010 (0.0020)	0.0000 (0.0000)	0.0011 (0.0018)	0.0000 (0.0000)	0.0023 (0.0044)	0.0000 (0.0000)
Azadirachtin	0.0001 (0.0001)	0.0007 (0.0006)	0.0002 (0.0002)	0.0011 (0.0010)	0.0002 (0.0002)	0.0010 (0.0011)	0.0003 (0.0003)	0.0013 (0.0013)
Azoxystrobin	0.0690 (0.0607)	0.0009 (0.0005)	0.1179 (0.1422)	0.0011 (0.0009)	0.1269 (0.1867)	0.0010 (0.0007)	0.2151 (0.3724)	0.0012 (0.0007)
Boscalid	0.1763 (0.1312)	0.0095 (0.0099)	0.2473 (0.1727)	0.0110 (0.0103)	0.2209 (0.1844)	0.0095 (0.0106)	0.2676 (0.2918)	0.0098 (0.0129)
Carbendazim	0.0718 (0.0694)	0.0086 (0.0104)	0.1142 (0.1041)	0.0151 (0.0177)	0.0893 (0.0787)	0.0124 (0.0130)	0.0883 (0.0765)	0.0116 (0.0146)
Chlorpropham	0.0934 (0.0755)	0.0003 (0.0002)	0.0809 (0.0727)	0.0002 (0.0002)	0.0868 (0.0696)	0.0003 (0.0002)	0.1025 (0.0964)	0.0003 (0.0003)
Chlorpyrifos	0.1039 (0.0825)	0.0052 (0.0053)	0.1678 (0.1262)	0.0092 (0.0090)	0.1404 (0.1088)	0.0078 (0.0071)	0.1697 (0.1674)	0.0087 (0.0086)
Cypermethrin	0.1094 (0.1282)	0.0150 (0.0185)	0.1724 (0.1948)	0.0245 (0.0275)	0.1300 (0.1456)	0.0185 (0.0209)	0.1214 (0.1393)	0.0176 (0.0203)
Cyprodinil	0.1057 (0.1044)	0.0067 (0.0084)	0.1368 (0.1228)	0.0089 (0.0104)	0.1178 (0.1098)	0.0073 (0.0094)	0.1310 (0.1293)	0.0079 (0.0111)
Difenoconazole	0.0260 (0.0216)	0.0009 (0.0008)	0.0432 (0.0394)	0.0012 (0.0011)	0.0399 (0.0445)	0.0012 (0.0016)	0.0441 (0.0610)	0.0015 (0.0021)
Dimethoate	0.0046 (0.0044)	0.0009 (0.0019)	0.0067 (0.0060)	0.0010 (0.0017)	0.0054 (0.0049)	0.0009 (0.0019)	0.0054 (0.0054)	0.0010 (0.0027)
Fenhexamid	0.1390 (0.1754)	0.0013 (0.0015)	0.1641 (0.1806)	0.0013 (0.0015)	0.1602 (0.1940)	0.0010 (0.0012)	0.1662 (0.2212)	0.0010 (0.0014)
Glyphosate	0.0055 (0.0067)	0.0003 (0.0003)	0.0141 (0.0251)	0.0004 (0.0005)	0.0166 (0.0160)	0.0004 (0.0004)	0.0264 (0.0235)	0.0005 (0.0007)
Imazalil	10.119 (10.131)	0.0186 (0.0248)	10.590 (10.763)	0.0207 (0.0287)	10.468 (10.735)	0.0230 (0.0308)	20.345 (30.562)	0.0301 (0.0407)
Imidacloprid	0.0945 (0.0782)	0.0498 (0.0650)	0.1238 (0.0969)	0.0551 (0.0750)	0.1162 (0.0958)	0.0610 (0.0806)	0.1353 (0.1262)	0.0792 (0.1060)
Iprodione	0.1992 (0.1995)	0.0007 (0.0007)	0.2625 (0.2588)	0.0011 (0.0012)	0.2363 (0.2507)	0.0010 (0.0014)	0.2808 (0.3554)	0.0012 (0.0015)
Malathion	0.0005 (0.0007)	0.0000 (0.0000)	0.0009 (0.0012)	0.0000 (0.0000)	0.0008 (0.0011)	0.0000 (0.0000)	0.0010 (0.0011)	0.0000 (0.0000)
Methamidophos	0.0004 (0.0004)	0.0000 (0.0000)	0.0005 (0.0006)	0.0000 (0.0000)	0.0005 (0.0004)	0.0000 (0.0000)	0.0006 (0.0007)	0.0000 (0.0000)
Profenofos	0.0001 (0.0001)	0.0000 (0.0000)	0.0001 (0.0001)	0.0000 (0.0000)	0.0001 (0.0001)	0.0000 (0.0000)	0.0001 (0.0001)	0.0000 (0.0000)

Pyrethrins	0.0019 (0.0020)	0.0019 (0.0016)	0.0019 (0.0023)	0.0028 (0.0024)	0.0023 (0.0025)	0.0028 (0.0026)	0.0022 (0.0024)	0.0032 (0.0026)
Spinosad	0.0738 (0.0881)	0.2257 (0.2296)	0.1136 (0.1336)	0.3052 (0.3009)	0.0837 (0.1004)	0.2571 (0.2544)	0.0747 (0.0964)	0.3054 (0.3634)
Tebuconazole	0.0479 (0.0512)	0.0017 (0.0014)	0.0634 (0.0631)	0.0016 (0.0012)	0.0578 (0.0638)	0.0016 (0.0013)	0.0677 (0.0845)	0.0015 (0.0017)
Thiabendazole	0.4022 (0.3695)	0.0167 (0.0224)	0.5884 (0.6428)	0.0186 (0.0260)	0.5810 (0.6920)	0.0207 (0.0279)	0.9758 (10.486)	0.0272 (0.0368)
lambda-Cyhalothrin	0.0153 (0.0127)	0.0004 (0.0003)	0.0237 (0.0208)	0.0006 (0.0004)	0.0209 (0.0190)	0.0005 (0.0004)	0.0246 (0.0269)	0.0006 (0.0004)
Aggregated indicator	0.2444 (0.1762)	0.1040 (0.0974)	0.3697 (0.2596)	0.1421 (0.1278)	0.3145 (0.2354)	0.1205 (0.1096)	0.3888 (0.3832)	0.1424 (0.1560)

Table 5. Estimated daily exposure to pesticide residues (µg/kg bw/day), by diet group, according to 100%-conventional and 100%-organic scenarios, N=34,442, NutriNet-Santé study[1,2]

[1]Values are means (SD) under the lower-bound hypothesis.

[2]All p-values $<10^{-2}$, except for the 100-organic scenario: anthraquinone (p-value=1) and profenofos (p-value =1). P-values are derived from non-parametric Kruskal-Wallis tests for heterogeneity between diet groups within a scenario.

In most cases, the estimated dietary pesticide exposure came from conventional sources (**Table 6**). Predictably, for active substances allowed in organic farming, organic food consumption was the main contributor to the overall dietary exposure.

	Omnivores		**Pesco-vegetarians**		**Vegetarians**		**Vegans**	
	Conventional	Organic	Conventional	Organic	Conventional	Organic	Conventional	Organic
Acetamiprid	0.0499 (0.0683)	0.0005 (0.0012)	0.0514 (0.0897)	0.0012 (0.0019)	0.0345 (0.0640)	0.0010 (0.0018)	0.0269 (0.0440)	0.0013 (0.0020)
Anthraquinone	0.0006 (0.0016)	0.0000 (0.0000)	0.0005 (0.0011)	0.0000 (0.0000)	0.0004 (0.0009)	0.0000 (0.0000)	0.0006 (0.0017)	0.0000 (0.0000)
Azadirachtin	0.0001 (0.0001)	0.0002 (0.0004)	0.0001 (0.0002)	0.0006 (0.0008)	0.0001 (0.0001)	0.0006 (0.0009)	0.0001 (0.0001)	0.0008 (0.0010)
Azoxystrobin	0.0424 (0.0449)	0.0002 (0.0003)	0.0365 (0.0450)	0.0006 (0.0006)	0.0403 (0.0588)	0.0006 (0.0005)	0.0607 (0.1605)	0.0008 (0.0006)
Boscalid	0.1116 (0.1051)	0.0031 (0.0060)	0.0967 (0.1118)	0.0064 (0.0085)	0.0806 (0.1102)	0.0055 (0.0081)	0.0799 (0.1080)	0.0061 (0.0101)
Carbendazim	0.0445 (0.0516)	0.0041 (0.0079)	0.0444 (0.0680)	0.0110 (0.0168)	0.0308 (0.0481)	0.0093 (0.0127)	0.0267 (0.0358)	0.0086 (0.0120)
Chlorpropham	0.0632 (0.0657)	0.0001 (0.0001)	0.0336 (0.0528)	0.0001 (0.0002)	0.0359 (0.0491)	0.0002 (0.0002)	0.0413 (0.0638)	0.0002 (0.0002)
Chlorpyrifos	0.0631 (0.0593)	0.0024 (0.0041)	0.0559 (0.0689)	0.0066 (0.0086)	0.0433 (0.0510)	0.0058 (0.0072)	0.0484 (0.0847)	0.0064 (0.0072)
Cypermethrin	0.0687 (0.0963)	0.0058 (0.0115)	0.0704 (0.1268)	0.0148 (0.0200)	0.0471 (0.0902)	0.0120 (0.0165)	0.0377 (0.0610)	0.0122 (0.0179)
Cyprodinil	0.0672 (0.0768)	0.0025 (0.0054)	0.0566 (0.0698)	0.0054 (0.0080)	0.0448 (0.0667)	0.0045 (0.0071)	0.0437 (0.0636)	0.0050 (0.0072)
Difenoconazole	0.0163 (0.0161)	0.0003 (0.0005)	0.0162 (0.0216)	0.0007 (0.0009)	0.0138 (0.0233)	0.0007 (0.0015)	0.0118 (0.0163)	0.0010 (0.0018)
Dimethoate	0.0029 (0.0033)	0.0003 (0.0011)	0.0026 (0.0039)	0.0006 (0.0014)	0.0020 (0.0030)	0.0006 (0.0014)	0.0017 (0.0022)	0.0007 (0.0026)
Fenhexamid	0.0906 (0.1302)	0.0004 (0.0008)	0.0655 (0.0942)	0.0007 (0.0013)	0.0647 (0.1345)	0.0006 (0.0010)	0.0564 (0.1252)	0.0007 (0.0011)
Glyphosate	0.0034 (0.0047)	0.0001 (0.0002)	0.0036 (0.0073)	0.0003 (0.0004)	0.0046 (0.0081)	0.0002 (0.0003)	0.0065 (0.0111)	0.0004 (0.0006)

Imazalil	0.7478 (0.9090)	0.0065 (0.0140)	0.5479 (0.8606)	0.0133 (0.0236)	0.5077 (0.7439)	0.0142 (0.0233)	0.6975 (10.831)	0.0213 (0.0350)
Imidacloprid	0.0608 (0.0618)	0.0173 (0.0366)	0.0483 (0.0618)	0.0350 (0.0616)	0.0443 (0.0590)	0.0373 (0.0609)	0.0413 (0.0567)	0.0558 (0.0911)
Iprodione	0.1303 (0.1558)	0.0002 (0.0004)	0.1076 (0.1421)	0.0006 (0.0008)	0.0867 (0.1402)	0.0007 (0.0013)	0.0913 (0.1773)	0.0007 (0.0008)
Malathion	0.0003 (0.0004)	0.0000 (0.0000)	0.0002 (0.0004)	0.0000 (0.0000)	0.0002 (0.0004)	0.0000 (0.0000)	0.0003 (0.0006)	0.0000 (0.0000)
Methamidophos	0.0003 (0.0004)	0.0000 (0.0000)	0.0002 (0.0004)	0.0000 (0.0000)	0.0002 (0.0003)	0.0000 (0.0000)	0.0003 (0.0005)	0.0000 (0.0000)
Profenofos	0.0000 (0.0000)	0.0000 (0.0000)	0.0000 (0.0000)	0.0000 (0.0000)	0.0000 (0.0000)	0.0000 (0.0000)	0.0000 (0.0001)	0.0000 (0.0000)
Pyrethrins	0.0014 (0.0017)	0.0007 (0.0010)	0.0009 (0.0016)	0.0016 (0.0018)	0.0011 (0.0018)	0.0016 (0.0020)	0.0010 (0.0019)	0.0020 (0.0023)
Spinosad	0.0470 (0.0666)	0.0898 (0.1537)	0.0485 (0.0871)	0.1864 (0.2594)	0.0315 (0.0615)	0.1642 (0.2191)	0.0246 (0.0434)	0.2155 (0.3503)
Tebuconazole	0.0315 (0.0397)	0.0005 (0.0008)	0.0253 (0.0335)	0.0009 (0.0010)	0.0211 (0.0353)	0.0009 (0.0010)	0.0221 (0.0405)	0.0010 (0.0016)
Thiabendazole	0.2645 (0.2927)	0.0059 (0.0126)	0.1979 (0.2851)	0.0119 (0.0213)	0.1993 (0.2764)	0.0127 (0.0210)	0.2877 (0.7376)	0.0193 (0.0316)
lambda-Cyhalothrin	0.0096 (0.0097)	0.0002 (0.0003)	0.0088 (0.0120)	0.0004 (0.0004)	0.0069 (0.0090)	0.0004 (0.0004)	0.0075 (0.0117)	0.0005 (0.0004)
Aggregated indicator	0.1545 (0.1349)	0.0415 (0.0667)	0.1338 (0.1543)	0.0879 (0.1118)	0.1058 (0.1220)	0.0777 (0.0960)	0.1162 (0.1966)	0.1006 (0.1510)

Table 6. Estimated daily exposure to pesticide residues (µg/kg bw/day), by diet group, disentangling conventional and organic sources, N=34,442, NutriNet-Santé study[1,2]

[1]Values are means (SD) under the lower-bound hypothesis.

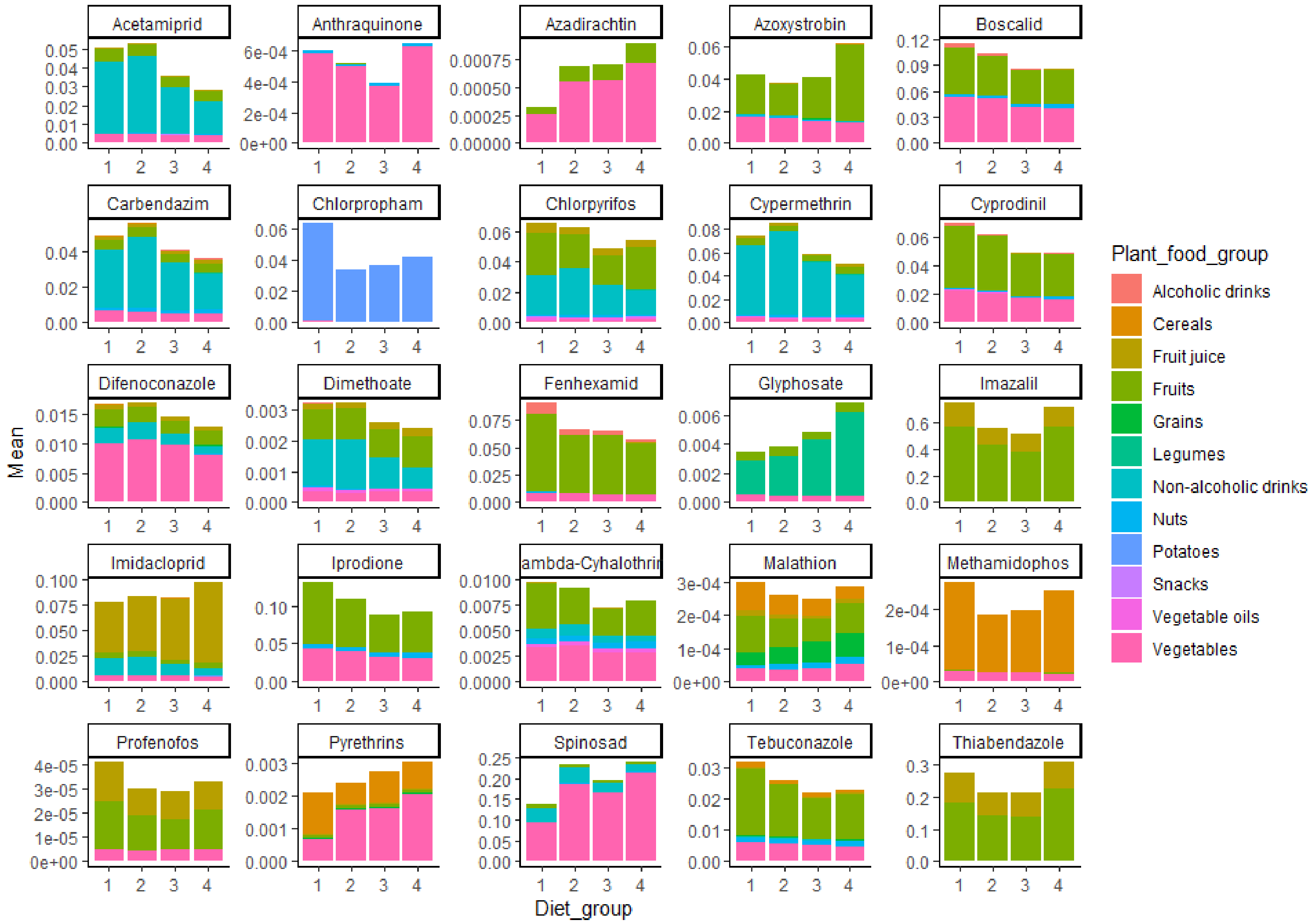
[2]All p-values $<10^{-4}$, except for organic sources: anthraquinone (p-value=1) and profenofos (p-value=1). P-values are derived from non-parametric Kruskal-Wallis tests for heterogeneity between diet groups within a farming system.

Sensitivity analyses

When compared to figures from the main analysis (Table 4), changing the allocated percentage to rarely when assessing organic food consumption did not substantially change the mean $E_{i,j}$ for most pesticides (**Supplementary Table 3**). However, it should be noted that exposure to azaridachtin, imidacloprid, pyrethrins and spinosad was lower for all diet groups when using 5% to the rarely modality. Multivariable-adjusted models yielded similar trends (**Supplementary Table 4).**

Contribution of plant food groups to total dietary pesticide exposure, by diet group

Figure 1 presents the contribution of plant-based food groups to the exposure levels, by diet group. The main food contributors were the same regardless of diet group while varying across pesticides. The major plant contributor to exposure to acetamiprid, carbendazim, chlorpyrifos, cypermethrin,



dimethoate were non-alcoholic drinks. Vegetable intake was responsible for exposure to anthraquinone, azadirachtin, boscalid, difenoconazole, pyrethrins (except for omnivores), spinosad while fruit intake was the main contributor to azoxystrobin, boscalid, cyprodinil, fenhexamid, imazalil, iprodione, tebuconazole, thiabendazole. Intake of potatoes was a very strong contributor to chlorpropham levels. Legume consumption accounted for most of the exposure to glyphosate and cereal consumption accounted for most of the exposure to methamidophos and pyrethrins in omnivores.

Figure. 1. Contribution of plant food groups to pesticide residue exposure (µg/kg bw/day), by diet group, N=34,442, NutriNet-Santé study

1: omnivores, 2: pesco-vegetarians, 3: vegetarians, 4: vegans

Discussion

Summary of findings

In this large population-based study, we assessed exposure to pesticides from plant-based food among four diet groups (omnivores, pesco-vegetarians, vegetarians and vegans), based on detailed data on conventional and organic food consumption and pesticide residue concentrations in foodstuffs. Unlike omnivores, vegans, and to a lesser extent, vegetarians and pesco-vegetarians, tended to consume more organic produce. Varying levels of exposure were observed across diet groups depending on the pesticide studied. Overall, vegetarians seemed the least exposed to the studied pesticides (notably, due to their relatively high level of organic food intake) while the results were more contrasted for other groups. Both plant-based food consumption and farming system appeared to play a role in the total exposure by increasing or diminishing it. Thus, in vegetarians and vegans, higher intakes of plant-based products led to higher dietary exposure to synthetic pesticide residues but their higher organic food consumption lowered their exposure to synthetic pesticides.

Observed diets and 100% conventional and organic scenarios

Daily intakes of pesticides estimated in our sample were higher overall than those obtained in the previous total French diet study on pesticide residues [45]. This can be explained by different methodological approaches (study population, assessment period, dietary tools, pesticide database). In particular, the use of a FFQ may have led to an overestimation of fruit and vegetable intakes, and thus also to an overestimation of resulting pesticide exposure, explaining the increased exposure observed in our study. Nonetheless, in both studies, imazalil, iprodione and thiabendazole exposure levels were high.

To our knowledge, few studies have investigated exposure to pesticides from plant-based foods accounting for the farming system, and only one compared dietary pesticide residue exposure between French vegetarians and the general population [20]. In the latter study based on consumption data from the 1997 French individual consumption survey, conducted in a representative sample, a very broad range of pesticides (>400), including organochlorine pesticides, were examined across five diets (omnivorous, lacto-vegetarian, ovolacto-vegetarian, pesco-lacto-vegetarian and vegan) in a rather small number of individuals (1,474 in total, with vegetarian groups all below 50). In that study [20], theoretical maximum daily intakes based on MRL were calculated as a percentage of the ADI to assess exposure. The non-consideration of farming system (generic foods were used) in the study by Audenhaege et al [20] and the non-consideration of animal-based products in ours as well as the differences in collection dates make comparisons difficult. However, interestingly, authors observed that pesticide exposure was higher among vegetarian populations than among omnivores. The high dietary exposure observed among vegans in that study [20] may be – partly – explained by the non-consideration of organic food. Interestingly, in our population, even omnivores were large fruit and vegetable eaters, which can explain their high exposure to imazalil and iprodione (although the latter has been recently banned in the EU) which are largely spayed on fruit crops, especially citrus crops.

Contribution of farming system to the exposure

Studies rarely integrate the farming system in their exposure assessment while some specific eating patterns, such as vegetarianism, and organic food consumption are strongly correlated [21–23]. Unsurprisingly, our data indicate a strong impact of the choice of organic produce in overall pesticide exposure. In a pilot study [29] conducted in 42 adults of a vegetarian community in Israel, a positive association was found between vegetable intake and urinary levels of a chlorpyrifos metabolite. Lower levels of total dimethylphosphate in individuals with higher intake of organic foods were observed, showing that the organic farming system lowered pesticide exposure among vegetarian populations. These findings are in line with ours to some extent. We observed in our study that vegans, despite their large amount of plant-based food consumption, were not systematically the group with the highest pesticide exposure, illustrating the attenuating role of organic food consumption. In particular, vegetarians exhibited the lowest overall exposure to the studied pesticides, as assessed by the aggregated indicator, due to a relatively good balance between plant-based product intakes and consumption of organic foods.

The 100%-conventional and organic scenarios corroborated the findings from observed data. Thus, a full conventional diet increased exposure levels of most pesticides - except those useable in organic farming - while the opposite was observed for a 100%-organic diet. Vegans were thus those with the highest exposure, for both 100%-conventional and 100%-organic scenarios, and omnivores those with the lowest. In vegans, the very high consumption of plant-based products mitigated the low contents of synthetic pesticides in organic products. Due to their high intake of fruits and stone fruits in particular, pesco-vegetarians showed their exposure to cyprodinil notably increased - cyprodinil is a fungicide used against apple scab among others.

Respective contribution of food groups to the exposure

In the present work, the most important food group contributors were generally the same for a given pesticide across all diet groups. They, however, differed across pesticides. Fruit and vegetables as well as cereals and legumes were the major contributors for most pesticide exposure, except chloprophamfor which it was potatoes. Chlorpropham is indeed a highly effective potato sprout inhibitor [49], widely used at post-harvest stage [50]. Additionally, acetamiprid, carbendazim, chlorpyrifos, cypermethrin, dimethoate were strongly linked to non-alcoholic drinks. This is mainly due to the high intake of tea and infusions in all groups. In the study by Van Audenhaege [20] et al, a strong link between fruit and vegetable consumption and carbamates was observed in all diet groups. In line with this, we found herein a strong correlation between thiabendazole (of the carbamate family) and fruit and fruit juices. In the above-mentioned study, cereals were the main contributors to chlorpyrifos exposure unlike in the present study where non-alcoholic drinks and fruits were the main food contributors. Comparison with other studies might be challenging given differences in study objectives, populations and collection date. However, our results show some similarity with those of the French total diet study on pesticide residues [45]. In that study, fruit and vegetables were also the major contributors to the exposure of a large number of the studied substances and potato products were also the main contributor to exposure to chlorpropham [45]. In contrast to our study, wheat flour-based products largely contributed to the exposure to the organophosphate insecticide chlorpyrifos-methyl. We, nevertheless, observed herein that cereal intake was the greatest contributor to exposure to the organophosphate methamidophos.

Methodological considerations

Several limitations and uncertainties should be noted. First, our study included healthy volunteers, generally more aware of food and health-related issues than the general population. It has thus been shown that NutriNet-Santé participants are more often highly educated and exhibit healthier dietary patterns than the general French population [51,52]. However, the large sample size and this specific nutrition-conscious population enabled us to have access to a wide range of diet behaviours, including vegetarian and vegan dietary patterns, leading to a very acceptable size for these subgroups. Another limitation is the use of the food frequency questionnaire and a 5-point scale to estimate total and organic food consumption. In particular, the scale pertaining to organic food consumption frequency was not validated. This may have caused a misestimation of the actual proportion of organic food consumption in the diet. Nevertheless, the estimation of mean proportion of organic food consumption was impacted to a limited extent when conducting sensitivity analyses with regard to the weighting [31]. Therefore, generalisation of our results to other populations may require caution as they are estimates based on a health-conscious population. It must be nonetheless noted that in a previous work by our group using the same questionnaire to measure organic food consumption, concentrations in certain pesticide biomarkers were significantly lower in organic food consumers [53]. Furthermore, self-report is prone to measurement error. The FFQ used in our study included an extensive list of food items (>260), which made it possible to cover a wide variety of eating habits, particularly those associated with organic food consumption and vegetarian eating habits. For instance, the FFQ included numerous meat- and dairy- substitute food items. FFQs are also very useful to classify individuals according to their usual food intakes [54]. Nonetheless, it is very likely that fruit and vegetable consumption as well as organic food consumption were somewhat overestimated [31,55]. The completion of the FFQ may have led to decreased motivation and social desirability response bias [55–57], though Internet-based studies have been associated with reduced desirability bias [58]. However, the Org-FFQ was based on a previous validated FFQ [33] to which the organic food frequency scale was added. In the sensitivity analysis where we attributed 5% to the rarely modality in response to the question relating to organic food consumption frequency, the trends were essentially unchanged. However, importantly, this was not the case for pesticides allowed in organic farming – in particular spinosad for which the exposure considerably decreased. This may be explained by the fact that spinosad is sprayed on many organic crops and, in turn, a small change when assessing organic food consumption may affect intakes of individuals with low organic food consumption. It should be noted that spinosad is of low acute and long-term toxicity to wild mammals, fish and birds but is of high acute and chronic risk for aquatic organisms (*i.e.* aquatic invertebrates) [59]. Nevertheless, most pesticides used in organic farming are of lower toxicity than synthetic pesticides [5,6]. We estimated usual daily intakes of pesticide residues and therefore our findings are of relevance for long-term chronic health effects. To assess acute health effects, short-term intake data and the consideration of the mixture of foods consumed together would have been required. With regard to pesticide data collection, it is noteworthy that animal-based products were not considered in our study. This may have resulted in an underestimation of exposure, for example to organochlorines, especially among omnivores and pesco-vegetarians, although plant foods remain the main contributors to pesticide exposure [10]. Of note, a recent study showed that dietary exposure from cereals and animal foods were among the major sources of pyrethroid exposure [9]. Furthermore, we based our analysis on German data gathered on their national market, as very few databases on pesticide residues in plant-based foods include the organic parameter. The same European regulations regarding organic standards apply for both France and Germany while differences exist in marketed plant protection products, though this bias is not easily quantifiable. We also selected emblematic pesticides for which we had enough data available, reducing the exposure coverage (*e.g.* banned organochlorines or copper were not considered), although a wide range of various family compounds were investigated. Finally, we did not account for transfer factors (concentration or dilution effects [60] during food processing (washing, peeling, heating or cooking)) due to lack of systematic data on these aspects. Strengths of our study include its large sample size, allowing to have access to large subgroups of vegetarians, the detailed food data and the consideration of the farming system (organic versus conventional). We also included three pesticides widely used in organic farming in our exposure assessment in order to better cover exposure from organic food and performed two scenarios. Importantly, the date of collection of the residue pesticide data used in our study overlapped with the date of completion of the questionnaire. Of note, in our study, we did not consider other sources of exposure that may exist. New approaches characterising the risk of single chemicals and chemical mixtures while taking into consideration their specific sources of interest have been recently developed [61].

Conclusion

In conclusion, we observed that dietary pesticide exposure was explained by both the amount of plant-based foods consumed and the quantities consumed in organic or conventional. High intake of food of plant origin increased dietary exposure to conventional pesticides while organic food consumption lowered it. It should be noted that dietary intakes of active substances allowed in organic farming however increased in full organic diets. Heavy organic food consumption could be therefore seen as a leverage to reduce exposure to synthetic pesticides in particular among high consumers of plant-based foods. Our study stresses the importance of refining pesticide exposure modelling, by notably collecting consumers' preferences as regards organic produce. The main issue would also be to determine whether and to what extent these differences in exposures translate into health outcomes. Further studies in other settings are warranted and should attempt to consider the farming system in their estimation.

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CONFLICT OF INTEREST None.

AUTHORSHIP

MT, SH, EK-G contributed to study design. RV, DL, JB and EK-G conducted the research and implemented databases. JB performed statistical analyses and drafted the manuscript. PR, BA, J-PC, MT, SH, DL, RV and EK-G contributed to the interpretation of data. All authors critically revised the manuscript and provided relevant intellectual input. JB had full access to all data and had primary responsibility for the final content.

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Supplementary Material. Estimation of dietary pesticide exposure

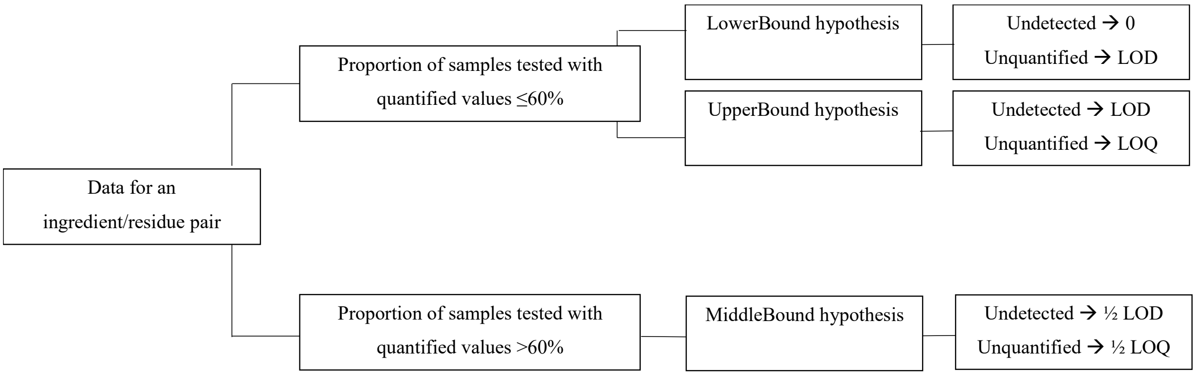
Dietary exposure of plant origin to 25 pesticide residues was assessed using NutriNet-Santé consumption data and pesticide residue data of the CVUA. The CVUA database comprises more than 6.7 million datapoints (a datapoint referring to a couple pesticide residue/product), including 1 million related to organic data. A more extensive description of the methodology used to derive dietary pesticide residue exposure has been provided elsewhere [44]. To do so, the 264 items of the Org-FFQ were broken down into 776 "ingredients", 442 of which were retained as they made up at least 5% of an item. Of these, only plant ingredients were included, giving a total of 180 ingredients, as the CVUA Stuttgart database only comprises foods of plant origin. Ingredients referred here to both raw foods (*e.g.* apple) and so-called "typical" ingredients from mixed foods (*e.g.* wheat flour and tomato sauce from pizza). The 180 NutriNet-Santé ingredients were then linked to their CVUA database equivalents and were attributed a pesticide residue value in conventional and organic (as the mean of corresponding datapoint). Of note, whenever data were missing for a couple ingredient/pesticide residue for one or another form (organic or conventional), the corresponding organic or conventional value was attributed. This may have led to an overestimation of pesticide residue levels in the case of some organic products.

Cooking and edibility coefficients [62,63] were assigned whenever needed to the ingredients, as NutriNet-Santé data referred to foods as consumed (*i.e.* peeled, bone-free or cooked products).

The same coefficients were allocated to both conventional and organic foods. Of note, preparation or cooking processes which may affect pesticide residue content in food [60] were not accounted for.

The method used for data treatment of undetected and unquantified values has been previously described [44] and is summarised in the **Supplemental Figure**. The methodology used was based on the international guidelines [64] and the method used by Nougadère et al [45].

Supplemental Figure. Decision tree for pesticide residue data treatment



The estimated daily intake expressed per kilogram of body weight per day (µg/kg bw/d) was calculated for each pesticide and for each individual, under the lower- and upper-hypotheses, using the following formula:

$$\sum_{k=1}^{n\_i} \mathrm{E\ i,j} = (\mathrm{Ci,k\ x\ Lk,j}) / \mathrm{Bwi}$$

$E_{i,j}$ estimated daily exposure to pesticide j for individual i (µg/kg bw/day)

n_i number of plant foods in the diet of individual i

$C_{i,k}$ mean daily intake of plant food k by individual i (g/day)

$L_{k,j}$ concentration of pesticide j in food k (mg/kg)

Bw_i body weight of individual i (kg)

Supplementary Table 1. Spearman's correlations between pesticide residue exposure and major plant food groups, N=34,442, NutriNet-Santé study

	Bread	Cereals	Vegetable oils	Potatoes	Snacks	Vegetables	Wholegrain products	Fruit juice	Fruit	Grains	Legumes	Dairy substitutes	Nuts	Soups	Meat substitutes
Acetamiprid	0.03	0.02	0.05	-0.06	0.00	0.13	0.00	0.02	0.22	-0.02	-0.02	-0.05	0.06	0.12	-0.07
Anthraquinone	0.05	0.17	0.01	0.00	0.13	0.04	0.00	0.10	-0.06	0.03	0.06	0.03	0.12	-0.02	0.07
Azadirachtin	-0.19	0.07	0.30	0.01	0.12	0.64	0.26	0.03	0.41	0.30	0.26	0.22	0.29	0.30	0.28
Azoxystrobin	0.10	0.09	0.07	0.09	0.02	0.29	-0.06	0.02	0.35	-0.07	0.03	-0.08	0.05	0.12	-0.13
Boscalid	0.07	0.02	0.13	0.06	0.01	0.40	-0.03	-0.02	0.49	-0.05	0.02	-0.09	0.08	0.17	-0.13
Carbendazim	-0.03	0.04	0.08	-0.08	0.04	0.17	0.05	0.05	0.23	0.06	0.03	0.02	0.16	0.13	0.01
Chlorpropham	0.23	0.20	-0.02	0.59	0.07	0.02	-0.16	0.03	0.01	-0.21	0.03	-0.17	-0.09	0.19	-0.24
Chlorpyrifos	0.03	0.02	0.06	-0.04	0.02	0.13	-0.01	0.13	0.37	-0.02	-0.01	-0.06	0.10	0.14	-0.09
Cypermethrin	-0.04	0.03	0.10	-0.08	0.04	0.13	0.06	0.07	0.20	0.07	0.03	0.03	0.15	0.13	0.02
Cyprodinil	0.07	0.00	0.12	0.04	0.01	0.34	-0.04	0.01	0.52	-0.06	-0.01	-0.09	0.07	0.15	-0.13
Difenoconazole	0.05	0.06	0.10	0.03	0.01	0.37	0.00	0.06	0.33	-0.02	0.05	-0.06	0.08	0.25	-0.09
Dimethoate	0.02	0.01	0.11	-0.02	0.03	0.18	0.01	0.11	0.33	0.00	0.01	-0.05	0.10	0.15	-0.07
Fenhexamid	0.06	-0.01	0.07	0.05	0.04	0.21	-0.02	0.03	0.41	-0.04	-0.01	-0.09	0.07	0.11	-0.13
Glyphosate	0.09	0.23	0.08	0.18	0.07	0.20	-0.02	0.00	0.21	-0.02	0.57	-0.04	0.08	0.13	-0.07
Imazalil	0.10	0.03	0.01	0.04	0.01	0.09	-0.06	0.31	0.37	-0.09	-0.02	-0.11	0.04	0.11	-0.16
Imidacloprid	0.04	0.05	0.00	-0.01	0.05	0.06	-0.01	0.77	0.13	-0.02	-0.02	-0.02	0.08	0.08	-0.04
Iprodione	0.07	-0.01	0.12	0.04	0.00	0.33	-0.04	-0.02	0.51	-0.06	0.00	-0.10	0.09	0.16	-0.14
Malathion	0.09	0.08	0.04	0.04	0.02	0.17	0.14	0.12	0.35	-0.01	0.02	-0.09	0.07	0.11	-0.13
Methamidophos	0.24	0.54	-0.04	0.20	0.02	0.00	-0.23	0.06	-0.08	-0.17	0.06	-0.14	-0.11	-0.04	-0.17

Profenofos	0.11	0.05	-0.01	0.05	0.01	0.10	-0.08	0.42	0.27	-0.11	-0.02	-0.12	0.01	0.08	-0.17
Pyrethrins	0.09	0.58	0.11	0.20	0.08	0.27	-0.02	0.11	0.06	0.06	0.19	0.02	0.04	0.07	0.03
Spinosad	-0.15	-0.03	0.28	-0.03	0.07	0.44	0.20	-0.01	0.29	0.25	0.16	0.15	0.24	0.23	0.18
Tebuconazole	0.09	0.01	0.07	0.05	0.01	0.25	-0.03	0.02	0.55	-0.07	0.00	-0.11	0.10	0.21	-0.15
Thiabendazole	0.09	0.04	-0.01	0.04	0.01	0.07	-0.07	0.41	0.34	-0.09	-0.02	-0.10	0.04	0.09	-0.15
lambda-Cyhalothrin	0.05	0.01	0.13	0.02	0.01	0.34	-0.02	0.00	0.48	-0.03	0.02	-0.07	0.12	0.17	-0.11
Aggregated indicator	-0.06	-0.01	0.21	-0.02	0.04	0.40	0.09	0.11	0.52	0.11	0.08	0.02	0.20	0.24	0.01

Supplementary Table 2. Spearman's correlations between pesticide residue exposure and fruit and vegetable food subgroups, N=34,442, NutriNet-Santé study

	Dark green vegetables	Red and orange vegetables	Other vegetables	Citrus fruits	Pomaceous fruits	Stone fruits	Berries	Other fruits
Acetamiprid	0.12	0.11	0.11	0.15	0.14	0.19	0.13	0.13
Anthraquinone	0.00	0.07	0.05	0.04	-0.07	-0.02	0.02	0.01
Azadirachtin	0.53	0.61	0.55	0.28	0.28	0.35	0.29	0.32
Azoxystrobin	0.28	0.24	0.23	0.21	0.17	0.25	0.18	0.42
Boscalid	0.43	0.30	0.29	0.26	0.32	0.42	0.26	0.33
Carbendazim	0.17	0.13	0.14	0.17	0.16	0.16	0.11	0.15
Chlorpropham	0.02	0.03	0.03	0.01	-0.02	0.04	0.01	0.02
Chlorpyrifos	0.11	0.11	0.12	0.43	0.21	0.26	0.14	0.25
Cypermethrin	0.12	0.11	0.12	0.19	0.13	0.17	0.11	0.14
Cyprodinil	0.36	0.27	0.25	0.26	0.23	0.51	0.41	0.36
Difenoconazole	0.32	0.33	0.35	0.24	0.22	0.27	0.16	0.22
Dimethoate	0.16	0.15	0.18	0.30	0.16	0.33	0.26	0.24
Fenhexamid	0.20	0.17	0.17	0.24	0.18	0.38	0.36	0.45
Glyphosate	0.16	0.16	0.22	0.17	0.22	0.11	0.06	0.14
Imazalil	0.08	0.09	0.08	0.58	0.16	0.27	0.15	0.29
Imidacloprid	0.04	0.06	0.05	0.10	0.02	0.13	0.10	0.14
Iprodione	0.35	0.25	0.25	0.29	0.24	0.61	0.30	0.37
Malathion	0.14	0.15	0.16	0.44	0.17	0.30	0.14	0.22
Methamidophos	-0.02	0.04	0.02	-0.02	-0.08	-0.06	-0.06	-0.04
Profenofos	0.08	0.10	0.10	0.46	0.10	0.21	0.13	0.18
Spinosad	0.50	0.30	0.33	0.22	0.24	0.24	0.21	0.20
Tebuconazole	0.22	0.23	0.23	0.30	0.26	0.67	0.34	0.38
Thiabendazole	0.06	0.07	0.06	0.47	0.14	0.23	0.13	0.30
lambda-Cyhalothrin	0.33	0.25	0.28	0.31	0.24	0.52	0.26	0.33
Aggregated indicator	0.43	0.29	0.31	0.49	0.32	0.41	0.26	0.36

Supplementary Table 3. Estimated daily exposure to pesticide residues (µg/kg bw/day) by diet group, observed diets, allocating 5% to the rarely frequency modality, N=34,442, NutriNet-Santé study[1,2]

	Omnivores	**Pesco-vegetarians**	**Vegetarians**	**Vegans**
Acetamiprid	0.0536 (0.0735)	0.0565 (0.0994)	0.0372 (0.0680)	0.0297 (0.0504)
Anthraquinone	0.0006 (0.0016)	0.0005 (0.0011)	0.0004 (0.0009)	0.0007 (0.0018)
Azadirachtin	0.0001 (0.0001)	0.0001 (0.0002)	0.0001 (0.0002)	0.0001 (0.0001)
Azoxystrobin	0.0451 (0.0474)	0.0397 (0.0485)	0.0438 (0.0639)	0.0675 (0.1914)
Boscalid	0.1195 (0.1105)	0.1063 (0.1268)	0.0877 (0.1209)	0.0860 (0.1164)
Carbendazim	0.0504 (0.0557)	0.0568 (0.0761)	0.0400 (0.0522)	0.0347 (0.0412)
Chlorpropham	0.0666 (0.0684)	0.0363 (0.0564)	0.0383 (0.0523)	0.0449 (0.0721)
Chlorpyrifos	0.0693 (0.0635)	0.0652 (0.0766)	0.0504 (0.0545)	0.0562 (0.0988)
Cypermethrin	0.0785 (0.1041)	0.0895 (0.1413)	0.0606 (0.0968)	0.0502 (0.0719)
Cyprodinil	0.0718 (0.0805)	0.0625 (0.0763)	0.0491 (0.0728)	0.0479 (0.0703)
Difenoconazole	0.0175 (0.0172)	0.0177 (0.0235)	0.0150 (0.0249)	0.0129 (0.0179)
Dimethoate	0.0031 (0.0036)	0.0029 (0.0043)	0.0022 (0.0032)	0.0018 (0.0025)
Fenhexamid	0.0964 (0.1404)	0.0708 (0.1046)	0.0703 (0.1466)	0.0620 (0.1489)
Glyphosate	0.0036 (0.0050)	0.0039 (0.0080)	0.0050 (0.0089)	0.0071 (0.0128)
Imazalil	0.8032 (0.9681)	0.5950 (0.9619)	0.5498 (0.8118)	0.7735 (20.201)
Imidacloprid	0.0656 (0.0666)	0.0526 (0.0673)	0.0485 (0.0658)	0.0455 (0.0638)
Iprodione	0.1398 (0.1656)	0.1182 (0.1568)	0.0943 (0.1516)	0.0990 (0.2044)
Malathion	0.0003 (0.0004)	0.0003 (0.0004)	0.0002 (0.0004)	0.0003 (0.0007)
Methamidophos	0.0003 (0.0004)	0.0002 (0.0004)	0.0002 (0.0004)	0.0003 (0.0006)
Profenofos	0.0000 (0.0001)	0.0000 (0.0001)	0.0000 (0.0000)	0.0000 (0.0001)
Pyrethrins	0.0015 (0.0018)	0.0010 (0.0017)	0.0013 (0.0020)	0.0012 (0.0021)
Spinosad	0.0506 (0.0716)	0.0536 (0.0967)	0.0344 (0.0654)	0.0279 (0.0499)
Tebuconazole	0.0339 (0.0420)	0.0280 (0.0370)	0.0232 (0.0378)	0.0244 (0.0463)
Thiabendazole	0.2833 (0.3119)	0.2143 (0.3150)	0.2157 (0.3038)	0.3182 (0.8762)
lambda-Cyhalothrin	0.0104 (0.0103)	0.0100 (0.0136)	0.0078 (0.0098)	0.0085 (0.0129)
Aggregated indicator	0.1678 (0.1438)	0.1515 (0.1717)	0.1190 (0.1305)	0.1319 (0.2297)

[1]Values are means (SD) under the lower bound-hypothesis.

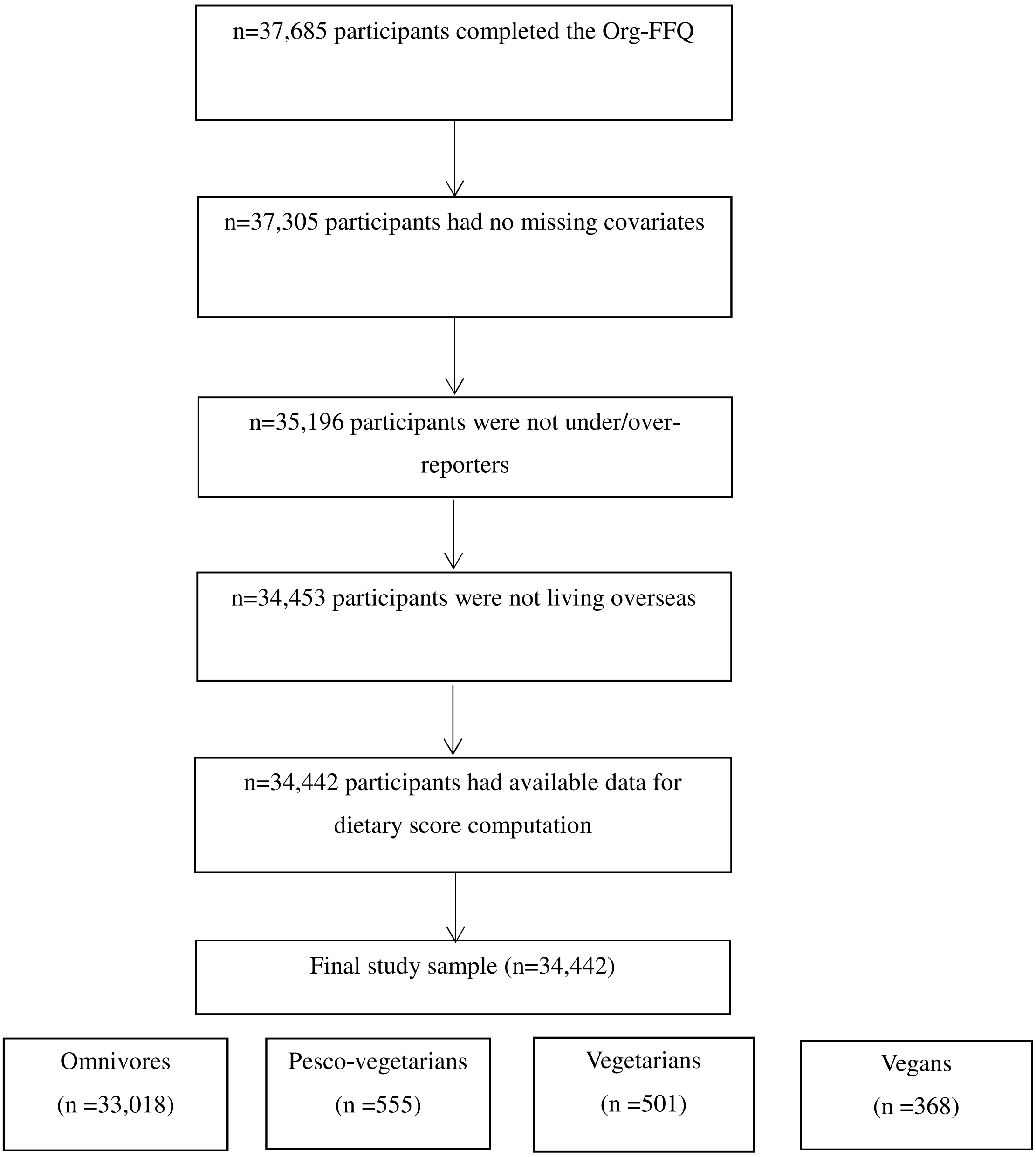
[2]All P-values $<10^{-4}$. P-values are derived from non-parametric Kruskal-Wallis tests.

Supplementary Table 4. Estimated daily exposure to pesticide residues (µg/kg bw/day), by diet group, adjusted model, observed diets, N=34,442, NutriNet-Santé study[1]

	Omnivores	**Pesco-vegetarians**	**Vegetarians**	**Vegans**	**P**
Acetamiprid	0.0504 (0.0497; 0.0511)	0.0512 (0.0456; 0.0568)	0.0372 (0.0313; 0.0432)	0.034 (0.0271; 0.0409)	<.0001
Anthraquinone	0.0006 (0.0006; 0.0006)	0.0005 (0.0004; 0.0006)	0.0003 (0.0002; 0.0004)	0.0005 (0.0004; 0.0007)	0.0001
Azadirachtin	0.0003 (0.0003; 0.0003)	0.0007 (0.0007; 0.0007)	0.0007 (0.0007; 0.0008)	0.0009 (0.0009; 0.001)	<.0001
Azoxystrobin	0.0425 (0.042; 0.043)	0.0394 (0.0355; 0.0433)	0.0445 (0.0404; 0.0486)	0.0652 (0.0604; 0.07)	<.0001
Boscalid	0.1141 (0.113; 0.1152)	0.1103 (0.102; 0.1186)	0.1036 (0.0948; 0.1123)	0.109 (0.0988; 0.1193)	0.07
Carbendazim	0.0485 (0.048; 0.0491)	0.0539 (0.0497; 0.0582)	0.0408 (0.0363; 0.0453)	0.0394 (0.0341; 0.0446)	<.0001
Chlorpropham	0.063 (0.0623; 0.0637)	0.0391 (0.0338; 0.0443)	0.0425 (0.037; 0.048)	0.0456 (0.0392; 0.052)	<.0001
Chlorpyrifos	0.0654 (0.0648; 0.066)	0.0622 (0.0574; 0.0671)	0.0522 (0.0471; 0.0573)	0.0615 (0.0555; 0.0675)	<.0001
Cypermethrin	0.0745 (0.0734; 0.0755)	0.0826 (0.0746; 0.0907)	0.0605 (0.052; 0.069)	0.0572 (0.0473; 0.0671)	<.0001
Cyprodinil	0.0693 (0.0685; 0.0701)	0.066 (0.0599; 0.0721)	0.0603 (0.0539; 0.0668)	0.0641 (0.0566; 0.0717)	0.02
Difenoconazole	0.0165 (0.0163; 0.0167)	0.0176 (0.0162; 0.0189)	0.0164 (0.0151; 0.0178)	0.0155 (0.0139; 0.0171)	0.26
Dimethoate	0.0032 (0.0032; 0.0032)	0.0033 (0.003; 0.0035)	0.0029 (0.0026; 0.0032)	0.0029 (0.0026; 0.0033)	0.08
Fenhexamid	0.0904 (0.089; 0.0918)	0.072 (0.0614; 0.0825)	0.0806 (0.0694; 0.0917)	0.078 (0.065; 0.091)	0.0006
Glyphosate	0.0035 (0.0034; 0.0035)	0.0041 (0.0037; 0.0045)	0.005 (0.0046; 0.0054)	0.0069 (0.0064; 0.0074)	<.0001
Imazalil	0.7518 (0.742; 0.7616)	0.5871 (0.5117; 0.6624)	0.5908 (0.5112; 0.6704)	0.8144 (0.7213; 0.9074)	<.0001
Imidacloprid	0.0783 (0.0775; 0.0791)	0.0825 (0.0764; 0.0886)	0.0785 (0.0721; 0.0849)	0.0926 (0.0851; 0.1001)	0.001
Iprodione	0.1296 (0.128; 0.1312)	0.1173 (0.105; 0.1297)	0.1133 (0.1002; 0.1263)	0.1286 (0.1134; 0.1439)	0.02
Malathion	0.0003 (0.0003; 0.0003)	0.0003 (0.0002; 0.0003)	0.0003 (0.0002; 0.0003)	0.0003 (0.0003; 0.0004)	0.16
Methamidophos	0.0003 (0.0003; 0.0003)	0.0002 (0.0002; 0.0002)	0.0002 (0.0001; 0.0002)	0.0002 (0.0002; 0.0002)	<.0001
Profenofos	0 (0.0000; 0.0000)	0 (0.0000; 0.0000)	0 (0.0000; 0.0000)	0 (0; 0)	<.0001
Pyrethrins	0.0021 (0.0021; 0.0021)	0.0024 (0.0022; 0.0025)	0.0025 (0.0024; 0.0027)	0.0027 (0.0025; 0.0029)	<.0001
Spinosad	0.136 (0.1342; 0.1378)	0.2393 (0.2255; 0.2531)	0.2151 (0.2005; 0.2296)	0.2716 (0.2546; 0.2886)	<.0001
Tebuconazole	0.0317 (0.0313; 0.0321)	0.0285 (0.0253; 0.0316)	0.0281 (0.0248; 0.0315)	0.0316 (0.0277; 0.0355)	0.04
Thiabendazole	0.2698 (0.2666; 0.273)	0.2174 (0.1928; 0.2421)	0.226 (0.1999; 0.252)	0.3228 (0.2923; 0.3532)	<.0001
lambda-Cyhalothrin	0.0097 (0.0096; 0.0098)	0.0097 (0.0089; 0.0104)	0.0086 (0.0078; 0.0094)	0.01 (0.009; 0.0109)	0.05
Aggregated indicator	0.1953 (0.1938; 0.1967)	0.2262 (0.2148; 0.2375)	0.2030 (0.1911; 0.2150)	0.2487 (0.2347; 0.2627)	<.0001

[1]Values are adjusted means (95%CI) derived from ANCOVA models adjusted for age, sex and energy intake using the residual method under the lower-bound hypothesis.

Supplementary Figure 1. Flow-chart of participants of the study



Org-FFQ, organic semi-quantitative food frequency questionnaire