AGRICULTURAL AND FOOD SCIENCE

Agricultural and Food Science (2025) 34: 38–50

https://doi.org/10.23986/afsci.145477

Comparison of the effects of bio-based and mineral fertiliser use on heavy metals dietary exposure in six European countries

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This study compared the effects of using bio-based fertilisers (BBFs) of high (BBF_H) or low (BBF_L) metal content with conventional mineral superphosphate (SP) fertilisation on dietary exposure of arsenic (As), cadmium (Cd), nickel (Ni) and lead (Pb), in Finland, Denmark, France, Germany, Hungary, and Spain. We estimated changes in the metal content of the following crops: wheat, barley, oat, rye, potato, carrot, sunflower seed, and maize following 100-year scenarios of fertilization with the different products. To estimate changes in chronic dietary exposure to metals via food, we used available national data of food consumption and metal content in target crops. Our results showed low changes to current chronic dietary exposure after using BBFs. Exceedance of the maximum allowed levels of Cd and Pb in food (EU 2023/915) was more rare with low-Cd BBF than with SP mineral fertilizer. Only the BBF_H slightly increased the dietary exposure to Cd, although similarly to SP. In conclusion, the studied BBFs did not increase dietary exposure to heavy metals, especially compared with the use of conventional SP mineral fertiliser.

Key words: recycling, health, risk, hazard, agriculture, contaminant

Introduction

Environmental impacts from production and excessive use of virgin synthetic and mineral fertilisers encompass greenhouse gas emissions during Haber-Bosch process (nitrogen-based fertilisers manufacturing), mining impacts (phosphorus extractions), and water systems eutrophication as a consequence of nitrogen and phosphorus overloading from agricultural soils (Guan et al. 2017, Hussain et al. 2021, Osorio-Tejada et al. 2022, Dias Blanco et al. 2023) social, and also a food sovereignty challenge for several countries. Due to the constantly growing global population, the food and feed production, and in turn, fertiliser needs are further rising (EC 2012, IPCC 2019). To minimize environmental impacts and enhance sustainability of food production, increasing circularity through the development of biobased fertilisers (BBFs) augments. BBFs can be produced from, e.g., treated sewage sludge, animal waste or food production by-products such as bones and feathers or crop residues.

Depending on the source and processing methods, BBFs may contain different ecotoxicological hazards (Chojnacka et al. 2020), which can be harmful to human health. For instance, dioxins, perfluorinated alkylated substances (PFAS), endocrine disrupting compounds, drug residues, antimicrobial resistant microbes, or heavy metals (HM) are some examples of these hazards. Though some HMs are essential for living organisms in trace levels, in general HMs are toxic, especially for humans. Cadmium (Cd), lead (Pb), inorganic arsenic (iAs) and nickel (Ni), targeted in this paper, are known to cause cancer, bone diseases or disorders in kidney, cardiovascular and neurological systems, and death (EFSA 2009a Cd, EFSA 2010 Pb, JECFA 2011 iAs, EFSA 2020 Ni, WHO 2007). In human body, the retention time of HM can range, depending on the organ, from days (Ni) to decades (Pb in bone, Cd in kidney) (e.g. see EFSA Journal reports for Cd (2012), Pb (2010) or Ni (2020) taking into account new occurrence data, the updated benchmark dose (BMD). The limits of HM content in fertilisers and food products are thus well regulated in Europe by, e.g., Commission Regulations (EU) 2023/915 and (EU) 2019/1009 as well as (EU) 2022/973, and at national level.

Although HMs are naturally present in Earth´s crust, human activities such as industry, urban waste, mining and agriculture contribute to HM pollution (Tchounwou et al. 2012, Saleh and Hassan 2022). Fertilisers containing HMs can increase the soil content of HM or enhance the mobility of existing HM reserves in soil. Metals enter in the food chain from soil through accumulation in crops´ tissues, however the type of fertiliser affects the crop metal content (Shumba et al. 2014). Shumba (2014) found that maize had the highest content of HMs (Cd, copper (Cu), Ni, and zinc [Zn]) after using sewage sludge-based fertiliser. Development of BBF processing and the assessment of HM bioaccumulation in environment and foods will minimize environmental and human health risks of using BBFs in agriculture.

Received 3 May 2024 / Accepted 7 February 2025
The Scientific Agricultural Society of Finland
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Cereals and vegetables are the main source of human dietary exposure to HMs, and for example for Finnish adults, their contribution was up to 30% of the total dietary exposure to the HM (Suomi et al. 2020, 2021). According to Khan et al. (2015) up to 90% of total HM human intake have crop origin. In addition, vegetarian, and cereal-based diet might grow as an environmentally friendly alternative to meat-based diet. Hence, HM exposure at population level might rise unless effective risk management strategies are implemented.

Toxicological dose-response studies determine adverse effects (response) caused at certain intake levels (dose). Based on these results, international food and health councils, like Food and Agriculture Organization of the United Nations (FAO), World Health Organization (WHO) or European Food Safety Authority (EFSA), develop health-based guidance values for risk assessment. Tolerable daily or weekly intake (TDI and TWI, respectively) values are maximum doses with which no adverse effect is likely over a lifetime exposure. Benchmark dose lower confidence limit (BMDL) values, on the other hand, are determined directly from the dose-response data and they still require a safety factor to arrive at the negligible risk dose.

The toxicity of the HM greatly depends on the metal, its species, and on the HM exposure frequency. In humans, the dietary characteristics, the gender and the age, play important role in HM toxicity (Suomi et al. 2021). For instance, HM toxicity in children is more severe compared to adults, due to organs formation and regeneration of new cells. In addition, children consume more food per bodyweight than adults do. In this sense, it is important to include age factor in risk assessments of HM dietary exposure.

The human HM dietary exposure is a function of the hazard occurrence in food and of the amount and frequency of food consumed (Ranta et al. 2021). The cumulative effects from simultaneous exposure to several chemicals with the same mode of action in the body should also be addressed at a later point, but in a previous study the main component of the cumulative health effects of dietary heavy metals (from the whole diet) was found to be Pb (Suomi and Tuominen 2023). In addition to the way HMs from fertilisers are taken up by plants, the uptake of HMs from plants to cow meat and milk may be of importance.

This study is part of Lex4Bio project (Horizon 2020 grant agreement No 818309) whose main aim is to develop BBFs sustainable and safe to use. This paper is done in parallel with the work of Salo et al. 2025 (submitted). While Salo et al. focus on estimation of HM occurrence in crops in 100 years based on a bass mass model, this paper used these results on crop´s HM occurrence and is focussed on the assessment of HM content in BBFs and the associated risk to human health through using BBFs instead of mineral fertilizers. This study compared changes on current HM occurrence in food and HM dietary exposure after using BBFs against traditional mineral fertilisation.

The specific aims of this paper are the following:

a) To study current occurrence of Cd, Pb, iAs, and Ni in eight crops (wheat, rye, barley, maize, potato, carrot, and sunflower seeds) from the following five countries: Spain, Denmark, Germany, Hungary, and Finland.

b) To investigate the effects of BBFs and mineral superphosphate (SP) fertiliser use on the HM occurrence in the target crops and countries in a 100-year time scale.

c) To analyse Cd and Pb content in the studied crops against EC regulated maximum levels, after using BBFs and SP fertiliser.

d) To study changes in HM dietary exposure after using BBFs at different HM content and compared with SP mineral fertiliser. Dietary exposure of adults (18 to 64 years) and children (3 to 10 years, except Finland 3 to 6 years) from Finland, Denmark, Germany, France, Hungary and Spain was studied. Despite the lack of current occurrence data, a rough estimate was possible for France, which was not included in aim (a). Due to better data availability, HM exposure of Finnish adults was assessed with a more precise method than the other population groups.

Materials and methods

All data harmonization, analyses, and graphs used for this paper, explained below, have been done using RStudio (Posit team 2023).

Effects of fertilisation on the current heavy metal occurrence in European crops

To study the effects of mineral and BBFs fertilisation in HM occurrence in European crops, we first surveyed the current situation in five European countries participating in the Lex4Bio project (excluding France, see explanations in following subsection). Thereafter, the relative change from this current level induced by 100-years of fertilization with a BBF high or low in metals or conventional SP was calculated based on a mass balance model (reported separately by Salo et al. 2025). Finally, this proportional change in content was used to estimate the changes in current density distribution of HM occurrence data and to estimate changes in dietary exposure (explained in the second section of Methods).

Current occurrence of HM in European crops

To study the current HM occurrence values in European crops we used data (from 2000 to 2021) of Cd, Pb, iAs, and Ni content in wheat, oat, barley, rye, maize, sunflower seeds, potato, and carrot (hereinafter LexCrops) originated and analysed in Finland, Spain, Germany, Hungary, and Denmark. Finnish data from years 2000 to 2017 and 2020 were provided by the Finnish Cereal Committee (VYR) and previously collected for national risk assessment reported in Suomi et al. (2020, 2021). The main bulk of occurrence data (2017–2021) was requested from EFSA via Public Access to Documents (hereinafter, PAD-data) (regulation (EC) No 1049/2001). These data had been submitted to the EFSA Data Warehouse by the corresponding EU Member States in the context of a call for continuous collection of chemical contaminants occurrence data in food and feed. To clarify, the food samples in the entire dataset used in this study were taken and analysed in the context of national monitoring or, to a much lesser extent, research by national authorities. We requested from data providers the authorisation to scientifically use their data included in the PAD-dataset (request PAD 2022/144). Spain, Germany, Denmark, Hungary, and Finland granted the permission, and only those results were used in analysis of occurrence data, although codifying countries´ names for the sake of confidentiality. Sample-level results were available at the EFSA food coding system FoodEx2 levels L1 to L3, mainly at the more detailed levels.

HM content analyses were done in accredited laboratories at each country. In total, the dataset used comprised 10882 samples of Pb, 9105 of Cd, 6012 of total As, 441 of iAs, and 5715 of Ni (see Table 1s in Supplementary material). Analysis methods and number of hazard-food pairs analysed varied among countries and years (Table 1s). However, these variabilities did not affect our statistical analyses cause the aim of this project was not to compare occurrence data geographically or chronologically.

Arsenic was mostly analysed as total arsenic instead of inorganic and organic arsenic independently. We estimated missing measurements of inorganic arsenic as the 70% of the total arsenic reported, which is the factor also used by EFSA (EFSA 2009a) for plant-based foods.

In addition to numerical occurrence data, a part of the data was left-censored, which means that their HM content was lower than limit of quantification (LOQ) (range summarised in Table 1s) and/or limit of observation (LOD) (values not shown). A traditional way of handling left-censored data is using lower bound (LB) and/or upper bound (UB) scenarios, where values below the limit of reporting (LOD or LOQ) are included as zero values (LB) or as values equal to the limit of reporting (UB).

After data cleaning (see Data Cleaning and uncertainties, in Supplementary material), the lower bound values were calculated. The mean and range of contents for each food-hazard pair in each target country are summarized in Table 2s, the results are shown only for cases where the number of samples was eight or higher.

The current distribution of HM occurrence in LexCrops was considered the starting point for analyses of BBFs and SP effects on content, explained in the following sections. However, due to the lack of individual-level food consumption data for target population, except for Finnish adults, we were unable to use the current HM occurrence data in the analyses of dietary exposure changes in other cases than for Finnish adults. HM occurrence data was used in BIKE model to estimate the HM exposure of Finnish adults (explained below in this article).

Calculation of HM content change factor induced by fertilisers use

To assess the effects of BBFs on the crop HM content and further on current dietary exposure, we conducted 100-year mass balance modelling for several crops in conditions of Spain, Denmark, Hungary, Finland, Germany and France (Salo et al. 2025). To compare with BBFs effects, we also used typical mineral superphosphate fertiliser

(SP) of each LexCountry. Results of SP uses were compared with lower (L) and higher (H) HM concentrated BBFs. In addition, we used a very high Cd-concentrated BBF_E, close to the EC maximum level, just to see the effect on the Cd content in wheat (Table 3s).

Results and details of mass-balance model will be reported in a separate paper (Salo et al. 2025). In brief, the metal inputs to and losses from a representative cropland in each country were estimated using best available published literature values or values calculated from experimental data of the Lex4Bio project. The input values for the model included total metal content in soil, aerial deposition, annual runoff and erosion, average liming and phosphorous fertilisation rates, metal contents in lime and fertilisers, average yield level of the target crops, soil-to-plant transfer factor and soil/water partition coefficient. For mineral fertilizer, typical HM contents in superphosphate of each country were used, whereas for BBFs the calculations were carried out with 2–4 varying contents adopted from screening of European BBF products under the Lex4Bio project. Wheat, barley, oat, rye, maize, and sunflower seed were modelled as monocultures, whereas potato and carrot were rotated with barley. The model produced annual estimates of HM mass balance, annual HM loss through leaching and erosion, HM content in runoff water, and soil, and yield uptake and content at current state (S0) and after 100-years (S100). The relative change in HM content (S_{ch}) with time (i.e., S100/S0) was considered the change factor induced by the BBF. S_{ch} (results in Table 3s) was further multiplied by current HM occurrence and dietary exposure data to estimate changes in content and foodborne exposure respectively in LexCountry´s crops and population.

Effects of BBFs and SP in current HM occurrence data in European crops

The relative content change factor S_{ch} estimated with HM mass-balance model was multiplied by current occurrence HM data at lower bound (explained in Current occurrence of HM in European crops). To visualize the effects of BBFs and SP in HM occurrence, we plotted the current density curve (Before in Fig. 1s) overlapping with density curves obtained after using S_{ch} for BBF_H or BBF_L (Fig. 1s).

We analysed the number of cases in which the current contents of Cd and Pb (C in Table 3) exceeded the European Commission maximum levels in foods (EC_ML) regulated in (EU) 2023/915, pooling data of all countries. After multiplying current HM content values by S_{ch} at each fertiliser use scenario, we repeated calculations of the number of cases exceeding Cd and Pb EC_ML after using BBFs (H, L in Table 3) and SP, followed by estimations of the relative proportion (in %) of cases after BBFs and SP uses out of the current situation (Table 3).

Effects of fertilization on current dietary exposure to heavy metals in European population

Current dietary exposure assessments

Due to the different source and quality of food consumption data, estimations of HM dietary exposure through food products derived from LexCrops (hereinafter Lexfood) were different for Finnish population compared of that for the rest of the cases. In addition, we estimated separately HM exposure for Finnish adults using BIKE model (explained in Finnish adults´ dietary exposure assessment before and after BBFs, BIKE model).

Dietary exposure is compared with health-based guidance values summarized in Table 1. Exposure below TWI or TDI value is of negligible risk. For Pb, the margin of exposure between the BMDL value and total exposure should be at minimum 10 for negligible risk, and for iAs, at least 1000 due to its carcinogenic properties.

Table 1. Toxicological reference values used in this paper. T(W/D)I is Tolerable (weekly or daily respectively) intake, and BMDL(i) is the benchmark dose at lower confidence limit for a (1 or 0.5% respectively) of cases.

Heavy metal	Toxicological reference value and its type	Reference
Cd	TWI=2.5 $\mu g\ kg^{-1}$ bw $week^{-1}$	EFSA (2009)
Pb	$BMDL_{01}$= 0.5 $\mu g\ kg^{-1}$ bw d^{-1}	EFSA (2010)
Ni	TDI = 13.5 $\mu g\ kg^{-1}$ bw d^{-1}	EFSA (2020)
iAs	$BMDL_{0.5}$=3.0 $\mu g\ kg^{-1}$ bw d^{-1}	FAO/WHO (2011)

In all exposure assessments, we assumed that neither the food consumption behaviour of the population groups nor the contents in other components of the diet than Lexfoods would change over the studied period.

For Spain, Germany, France, Hungary, and Denmark we used the total dietary exposure reported by EFSA (EFSA 2009a&b, 2012, 2020, and 2021 for Cd, Pb, Ni and iAs respectively, see Table 2a) in adults (18 to 64 years) and children (3 to 10 years) based on general food categories instead of ingredient levels. We first estimated the contribution of Lexfood categories to the total dietary exposure, considering that all ingredients of studied food category contributed equally to the 100% of the EFSA reported total dietary exposure. To estimate the contribution of Lexfoods to the total dietary exposure we first collected consumption information of food containing Lexfood as ingredients, from EFSA Food Consumption Database (URL https://www.efsa.europa.eu/en/data-report/food-consumption-data). EFSA Food Consumption database contains data on nationally conducted surveys, uploaded to EFSA by the national health authorities, the data producers of surveys. Only the statistical indicators at age- and gender- groups are available as open access, and these were used by us. We selected food consumption values from the same surveys that EFSA used in the dietary exposure reports for each HM. We selected food categories from L1 to L5 in the FoodEx2 coding system, out of which L1 is the roughest and L5 the most detailed categorization. From L1 we selected the following food groups: a) grains and grain-based products, containing wheat, oat, barley, rye and maize; b) starchy roots or tubers and products thereof, sugar plants, containing potato; c) vegetables and vegetable products, including carrot; and d) legumes, nuts, oilseeds and spices, which included sunflower seeds. Finally, we estimated the content of LexCrop in L5 categories using recipes found in internet and choosing those mixed foods either with more than 60% of LexCrop or with high consumption in corresponding age- or country-groups. Finally, the relative Lexfood out of the total food consumed was multiplied by the total HM dietary exposure to estimate the current Lexfood dietary exposure at each age- and country-group.

For Finnish adults (25 to 64 years) and Finnish children (3 to 6 years) we used dietary exposure values calculated in the national risk assessments Suomi et al. (2020) and (2015) respectively, the main results of which were published in Suomi et al. (2018) and (2021). In the national risk assessments, the food consumption data were collected in surveys FinDiet 2007 and FinDiet 2012 (adults, whole country) and DIPP (children, regional). For the current study, we used the older 2007 consumption data to estimate the exposure from other food sources, as it was entirely calculated to ingredients. For comparison, Table 2b also includes the corresponding EFSA estimates for Finland with these food consumption data. EU-level data used by EFSA usually gives a more conservative estimate than national data, and the difference in iAs results in Table 2b is caused by the uncertainty from using conversion factors (total As to iAs) in the national assessment.

Table 2a. Current chronic dietary exposure to HM from the whole diet according to EFSA's reports (EFSA 2012 Cd; EFSA 2012 Pb; EFSA 2021 iAs; EFSA 2020 Ni). Values are lower bound estimates (<LOQ = 0) of the mean exposure. If several estimates were available from a country, the most recent is shown. In units, bw means body weight. Missing value is marked NA.

Hazard (unit)	Country	Adults (18–64 yr)	Children (3–10 yr)
Cd (µg kg^{-1} bw $week^{-1}$)	DE	1.21	2.63
	DK	1.30	2.86
	ES	1.74	3.52
	FR	1.54	3.32
	HU	1.62	NA
Pb (µg kg^{-1} bw day^{-1})	DE	0.42	0.65
	DK	0.50	0.92
	ES	0.51	0.78
	FR	0.39	0.76
	HU	0.34	NA
iAs (µg kg^{-1} bw day^{-1})	DE	0.04	0.10
	DK	0.05	0.09
	ES	0.04	0.11
	FR	0.06	0.12
	HU	0.03	NA
Ni (µg kg^{-1} bw day^{-1})	DE	3.34	7.13
	DK	3.18	6.11
	ES	2.01	7.12
	FR	3.02	7.13
	HU	3.06	NA

Table 2b. Current lower bound chronic dietary exposure to HM from the whole diet in Finland according to national risk assessments (Suomi et al. 2015, Suomi et al. 2020) and EFSA's reports. National risk assessment on adults (Suomi et al. 2020) assessed dietary exposure based on two FinDiet survey datasets from different years (in column FinDiet-year), the results of which are presented on separate rows. Children's exposure was calculated separately for 3 and 6 years olds.

Hazard (unit)	Adults (25–64 yr)	Children (3 and 6 yr)	FinDiet year	Source
Cd (µg kg^{-1} bw $week^{-1}$)	1.22	3.20	2007	EFSA 2012
	0.98 (CI 0.95-1.07)	2.45 (3 years)	2007	National
	1.12 (CI 1.02-1.17)	2.17 (6 years)	2012	National
Pb (µg kg^{-1} bw d^{-1})	0.47	1.00	2007	EFSA 2012
	0.19 (CI 0.17-0.21)	0.39 (3 years)	2007	National
	0.17 (CI 0.15-0.18)	0.32 (6 years)	2012	National
iAs (µg kg^{-1} bw d^{-1})	0.06	0.12	2012	EFSA 2021
	0.17 (CI 0.16-0.18)	0.19 (3 years)	2007	National
	0.18 (CI 0.17-0.19)	0.17 (6 years)	2012	National
Ni (µg kg^{-1} bw d^{-1})	3.34	6.40	2012	EFSA 2020
	2.43 (2.21 – 2.71)	not included	2007	National
	2.53 (2.31 – 2.77)	not included	2012	National

Effects of BBFs and SP on current dietary exposure on European population

Like we did with occurrence data, S_{ch} was multiplied by the current dietary exposure through LexCrops at each BBFs and SP fertilisation. Since the food consumption was assumed to remain constant, the effect on the exposure would be equal to the effect on the content.

We summed results of all Lexfood dietary exposure at each country and age group separately for each fertilizer type used in order to estimate the total Lexfood dietary exposure before ($TLexE_B$) and after ($TLexE_{A(i)}$) treatments. The background exposure from foods and drinks other than Lexfoods (bE) was estimated subtracting $TLexE_B$ from the total dietary exposure (TE) (values from Table 2). bE was assumed to remain constant after treatments. Changes in chronic dietary exposure (DE_{ch}) after using SP or different BBFs were the sum of total Lexfood exposure (fertiliser dependent) and background exposure (fertiliser independent) relative to TE, the total dietary exposure (for better visualization of calculations, see Equation:

$$DE_{ch} = \left(\frac{\left(TLexE_{A(i)} + bE\right) * 100}{TE}\right) - 100$$

According to Equation the use of target fertilisers may increase (positive results), decrease (negative results) or have no effect on (zero results) the current total dietary exposure to HMs (see Table 4).

Finnish adults´ dietary exposure assessment before and after BBFs, BIKE model

Finnish adults' current dietary exposure assessment through LexCrops

The dietary exposure for Finnish adults, 18 to 64 years old, through LexCrops was carried out using a Bayesian model (BIKE) developed at the Finnish Food Authority (Ranta et al. 2021). The model is fully described in (Ranta et al. 2021) and available as online application (Ranta et al. 2023). In brief, using content, information on the presence of target hazards in food, the amount and frequency of food consumed per body weight, and the tolerable maximum intake for each hazard (see Table 1), BIKE simulates acute or chronic dietary exposure, and the inference of variability and uncertainties. For chemical hazards like HM, the current version of BIKE estimates the chronic exposure, which is the most relevant for these chemicals. The content data for BIKE needs to be at sample level and the food consumption data at individual level. We used data of HM collected in Finland, explained in subsection Current occurrence of HM in European crops. (See current occurrence values of HM in Finnish wheat as an example of BIKE outcome in Figure 2).

The food consumption data of Finnish adults were collected in FinDiet 2017 survey done as part of FinHealth 2017 project. The data were received from THL Biobank (research number THLBB2022_20). The details of the FinDiet survey and its data collection are described in Kaartinen et al. (2018) report. The survey contained information of two 24-h recall interviews from 1593 persons out of whom 1157 were aged 18 to 64, target group for this paper. This selected group was 53% female with body weight range from 39 to 150 kg. The mean weight of females was 72.6 kg and that of males 86.3 kg.

The chosen model settings in BIKE were the following: independent days for Consumption model, no analyses of variability in consumption frequencies between users, and no correlation model of consumption frequencies. We chose sigma uniform priors for variances, best suited to scarce data, and used 10000 Markov Chain Monte Carlo (MCMC) iterations. BIKE gives results both as posterior predictive distribution estimates (Table 4s), which combine uncertainty and variability from the data and the model, as well as distribution curves also presenting separately the variability of the estimate (Table 5).

To estimate the changes on the current (2017) dietary exposure of Finnish adults using BBFs or SP fertilisation (Table 6), assuming that food consumption did not change with time, we first estimated the exposure with the current contents and then estimated with the same settings the after-BBF exposures by using S_{ch} as a multiplication factor for each sample and using the resulting data as the input for the contents in a new estimate.

In the detailed exposure assessment with BIKE, we only assessed exposure from Lexfoods (Table 5). To estimate the effect on the total dietary exposure, we used the previous estimate of total dietary exposure of Finnish adults calculated with FinDiet 2007 data (Suomi et al. 2020), the raw data of which were available at detailed ingredient level and thus the contribution of Lexfoods and the rest of the diet were easy to separate. The background exposure from other than Lexfoods was thus assumed to be the same as the lower bound median estimate for these foods in (Suomi et al. 2020), since a more recent national estimate on the other sources of exposure was not available (Table 7).

Results

Effects of BBFs and SP on crop HM occurrence after 100-years of fertilizer use

Analyses on current content of HMs showed large variability in the means and minimum-maximum range values within studied crops and countries (Fig. 1). On average, Ni and Cd were accumulated the most in sunflower seeds, largest Pb contents were in wheat and maize, and iAs seemed to be the less concentrated compound in all analysed crops. The use of BBFs or SP exhibited a minor effect on the density distribution curves of LexCrops HM occurrence (Fig. 1).

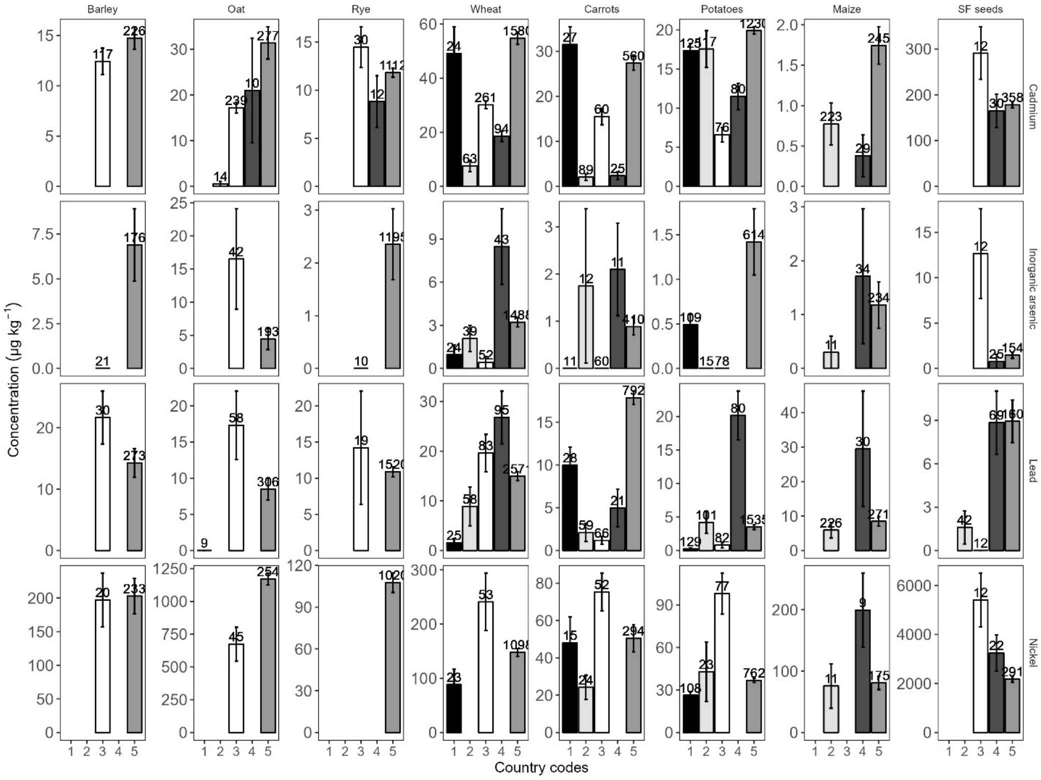

Fig. 1. Mean (bars), standard error SE (vertical line) and number of samples (on the top of bars) of lower bound HM content values in crops in different countries. Countries, identified by numbers to pseudonymise data for the sake of confidentiality, are shown as separate bars. Identity of the crops is marked at the top of the graph ("SF seeds" = sunflower seeds) and the heavy metal studied is at the right axis of the graph. Only cases with more than 8 samples are included in the graph.

The Cd and Pb content values rarely exceeded the maximum levels of Commission Regulation (EU) 2023/915 (EC_ML), in both, before and after scenarios, using BBFs and SP (Table 3). The number of cases exceeding EC_ML after using BBF_L (L in Table 3) relative to the cases under current conditions (C, in Table 4) decreased in the majority of LexCrops. This means that the use of BBF_L would decrease the portion of produced crops that are unfit for human consumption due to Cd or Pb levels. The exceptions were Cd in rye as well as Pb in barley and oat, for which there was no change from the current situation (see Table 3). The use of BBF_H seems to increase the number of cases where Cd exceeds EC_ML (H in Table 3) relative to the current scenario, except for sunflower seeds that was constant. Thus, use of BBF with high Cd levels would lead to more sample lots being unfit for human consumption than use of SP fertilisation. However, BBF_H reduced or unchanged the number of samples exceeding Pb EC_ML (Table 3). BBF_E use in wheat increased in the number of cases exceeding Cd EC_ML by 310% (data not shown).

Table 3. Commission Regulation (EU) 2023/915 maximum values (EC_ML) for Cd and Pb and samples in the content dataset exceeding the maximum value. C is number of samples exceeding EC_ML in the dataset used, and effect of mineral (SP) and BBFs (L and H) fertilisers is shown relative to C.

Hazard	Type (N)	C	EC_ML (µg kg^{-1})	SP/C (%)	L/C (%)	H/C (%)
Cadmium	Barley (360)	11	50	80	70	180
	Oat (548)	7	100	90	90	170
	Rye (1169)	20	50	100	100	140
	Wheat (2022)	151	100	90	80	150
	Maize (497)	0	100	nd	nd	nd
	SF seeds (389)	5	500	40	40	100
	Carrots (761)	11	100	90	40	150
	Potatoes (1628)	5	100	100	80	140
Lead	Barley (316)	2	200	100	100	100
	Oat (385)	1	200	100	100	100
	Rye (1562)	1	200	nd	nd	nd
	Wheat (2832)	36	200	90	90	90
	Maize (528)	13	100	70	70	70
	SF seeds (271)	0	900	nd	nd	nd
	Carrots (966)	5	100	80	80	80
	Potatoes (1927)	12	100	90	90	90

nd = non-detected values over EC_ML

Effects of BBFs and SP on dietary exposure in LexCountries

In Table 4 are summarised results of the change in total dietary exposure after BBFs or SP fertilisation in all studied LexCountries (Table 4a) and in Finland separately (in Table 4b), since the sources of data were different.

Use of high-Cd-concentrated BBF_H increased the dietary exposure to Cd in adults and children of all countries. Similar effects were seen after using SP mineral fertiliser except for population from country C. However, the use of low-Cd-concentrated BBF_L, decreased marginally (less than 1.5%) the dietary exposure in all populations (Table 4a and b).

Regarding to the rest of heavy metals, the use of BBFs and SP unlikely affect the dietary exposure. Exposure to iAs slightly decreased in all cases, and exposure to Pb and Ni exposure decreased in all population of three and two out of six countries after using BBF_H and SP, including Ni for Finnish adults after using BBF_H (Table 4b).

Table 4a. Changes in dietary exposure in five LexCountries (a–e) through target crops (LexCrops) relative to the total dietary exposure of heavy metals, at each age- and country-groups (N of LexCrops in paranthesis) after using mineral (SP) and low (L) or high (H) metal-contained BBFs. Units in %. Countries were identified by letters to pseudonymise data for the sake of confidentiality.

Hazard	Adults (18–65 yr)				Children (3–10 yr)			
	Country (N)	SP	BBF$_L$	BBF$_H$	Country (N)	SP	BBF$_L$	BBF$_H$
Cd	a (5)	0.79	-1	0.87	a (5)	0.76	-0.98	0.84
	b (7)	2.29	-0.65	3.1	b (6)	3.13	-0.88	4.23
	c (8)	-0.43	-0.68	0.98	c (7)	-0.74	-1.18	1.67
	d (7)	3.53	-1.39	1.74	d (5)	3.54	-1.32	1.78
	e (8)	6.28	-0.36	8	NA	NA	NA	NA
Pb	a (5)	0.56	0.56	0.5	a (5)	0.54	0.54	0.49
	b (7)	0.19	0.19	0.1	b (6)	0.26	0.27	0.14
	c (8)	-0.6	-0.63	-0.58	c (7)	-1.04	-1.1	-1.01
	d (7)	-1.16	-1.22	-1.16	d (5)	-1.11	-1.17	-1.11
	e (8)	-0.53	-0.53	-0.71	NA	NA	NA	NA
Ni	a (8)	-0.69	-0.78	-0.25	a (6)	-0.74	-0.84	-0.27
	b (7)	-2.2	-2.6	-0.93	b (7)	-3.08	-3.65	-1.33
	c (8)	-0.69	-0.7	-0.4	c (8)	-1.25	-1.27	-0.72
	d (7)	1.96	2.05	1.58	d (7)	2.23	2.32	1.8
	e (8)	1.15	1.27	0.08	NA	NA	NA	NA
iAs a (8)-1.48			-1.55	-1.48	a (6)	-1.57	-1.66	-1.57
	b (7)	-0.11	-0.25	-0.07	b (7)	-0.18	-0.36	-0.13
	c (8)	-0.95	-0.95	-0.95	c (8)	-1.72	-1.72	-1.68
	d (7)	-2.77	-2.85	-2.77	d (7)	-3.16	-3.22	-3.16
	e (8)	-1.29	-1.29	-1.1	NA	NA	NA	NA

*BBF$_H$ used for iAs was different than the used for the rest of heavy metals; NA = missing data

Table 4b. Changes in dietary exposure in Finland through target crops (LexCrops) relative to the total dietary exposure of heavy metals at each age group after using mineral (SP) and low (L) or high (H) metal-contained BBFs. Units in %. The method for calculating the changes is the same than in 4a. For the adults, the raw data on dietary exposure were based on FinDiet 2007 survey and ref. (Suomi et al. 2020), and thus a different dataset from the one used in the detailed assessment with BIKE.

Hazard	Adults (25–64 yr)				Children (3–6 yr)			
	LexCrops	SP	BBF$_H$	BBF$_H$*	LexCrops	SP	BBF$_L$	BBF$_H$*
Cd	6	0.12	-0.5	15.62	5	0.13	-0.51	16.19
Pb	6	0.42	0.42	0.48	5	0.61	0.61	0.73
Ni	6	-0.52	-0.52	0.99	NA	NA	NA	NA
iAs	4	-0.06	-0.06	-0.04	3	-0.19	-0.2	-0.12

*BBF$_H$ used for iAs was different than the used for the rest of heavy metals; NA = missing data

Dietary exposure in Finnish adults through Lexfoods (results from BIKE)

The mean values, including uncertainties and variabilities of the most probable posterior predictive distribution of LexCrop dietary exposure in Finnish adults were the following: Cd= 0.41 (µg kg^{-1} bw per week), Pb = 0.05 (µg kg^{-1} bw per day), Ni = 0.70 (µg kg^{-1} bw per day), and iAs= 0.02 (µg kg^{-1} bw per day) (Table 4s). Table 5 summarizes the estimated 50% and 95% quantiles (P50 and P95 respectively) of Finnish adults exposure to HM through Lexfoods, and with FinDiet 2017 consumption data.

Table 5. Chronic dietary exposure of Finnish adults (18 to 64 years) from Lexfoods with the current content data and FinDiet 2017 consumption. Median (P50) and 95th percentile (P95) of the exposure is shown in µg kg^{-1} body weight per day except for Cd that is in µg kg^{-1} body weight per week.

Hazard	P50 (CI 90%)	P95 (CI 90%)
Cd	0.41 (0.37–0.44)	0.70 (0.62–0.83)
Pb	0.04 (0.03–0.09)	0.08 (0.06–0.22)
Ni	0.64 (0.52–2.49)	1.21 (0.96–5.24)
iAs	0.02 (0.06–0.16)	0.09 (0.02–0.63)

The contribution of each LexCrop to these total values (Fig. 2) indicated that wheat consumption is the main source for Cd, Pb and Ni exposure, while oat is the main source for inorganic arsenic among LexCrops.

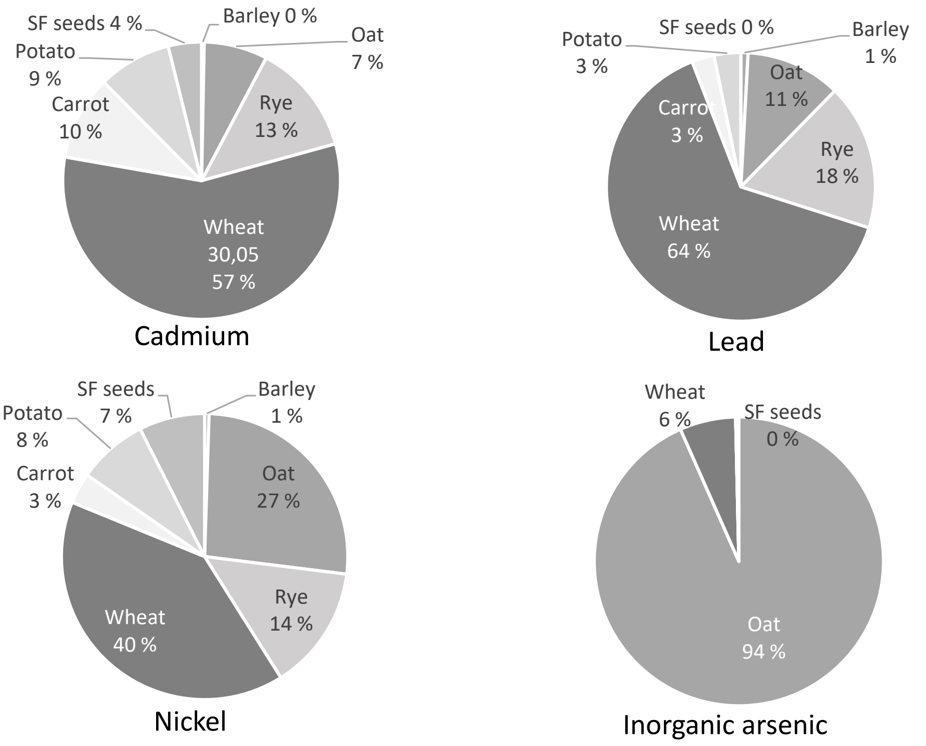

Fig. 2. Contribution of each LexCrop to the dietary exposure from all Lexfoods (Finnish adults, 18 to 64 years)

Changes in total dietary exposure in Finnish adults

Table 6 indicates the dietary exposure (at P50 and P95 quantiles respectively) estimated after using mineral (SP), low (L) and high (H) metal-concentrated BBFs relative to those estimated for current (C) situation (in Table 5). Relative changes seen in both quantiles are very similar to each other. In general, dietary exposure to Cd seems to increase over the current values after using BBF$_H$, while the rest of the cases likely unchanged or slightly decreased, especially for exposure to iAs after using BBF$_L$ and SP.

Table 6. Dietary exposure after using mineral (SP) and low (L) and high (H) metal-containing BBFs relative to current dietary exposure (C) (in %) to metals estimated at the 50th (P50) and 95th (P95) percentile for Finnish adults. Values of C are shown in Table 5.

Hazard	P50			P95		
	SP/C	H/C	L/C	SP/C	H/C	L/C
Cd	100	123	100	102	120	102
Pb	100	99	100	101	101	101
Ni	96	105	96	97	102	97
iAs	84	96	84	80	78	80

Compared with the current situation, the total dietary exposure (P95) for Finnish adults to Cd was estimated to slightly increase only after using the higher Cd concentrated BBF_H. In the rest of the cases, neither mineral (SP) fertiliser most used in Finland, nor studied BBFs modify the current HM chronic dietary exposure (Table 7)

Table 7. Total dietary exposure (P50) to HM for Finnish adults, assuming that the HM exposure from the rest of the diet is equal to the contribution of other foods than Lexfoods estimated with FinDiet 2007 data in (Suomi et al. 2020).

	C	SP	L	H
Cd (µg kg bw^{-1} $week^{-1}$)	0.83	0.83	0.82	0.92
Pb (µg kg bw^{-1} d^{-1})	0.14	0.14	0.14	0.14
Ni (µg kg bw^{-1} d^{-1})	2.79	2.77	2.76	2.8
iAs (µg kg bw^{-1} d^{-1})	0.11	0.11	0.11	0.11

Discussion

In this study we compared how the use of different fertilisers may affect the HM dietary exposure in adults and children of six European countries. The aim was to survey a lower (L) and higher (H) metal-contained biobased fertilisers (BBFs), commercialized in Spain, Germany, Hungary, Denmark, France, and Finland. We aimed to compare results from BBFs (common for all countries) against the typical mineral superphosphate (SP) fertiliser in each studied country, to have more real cases in mapping scenarios. BBF_H used to study inorganic arsenic (iAs) was different than that used for the rest of studied heavy metals, i.e., cadmium (Cd), lead (Pb) and nickel (Ni).

The occurrence of HMs showed large variability in the distribution within studied crops and countries (Fig. 1), which was expected, since different crops and even cultivars, environment and soil characteristics affect HM accumulation in plants (e.g. Suomi et al. 2015 and Salo et al. 2018). The effect of the different fertilisers on the mean content of different crops is shown in Supplementary data (Table 3s). The effect of the different fertilisers on the portion of produced crops exceeding the legislative maximum values for Cd or Pb (Table 3) could not be studied at country level, as fortunately the main part of the produce sampled in official controls is fit for human consumption.

Whether the exposure leads to risk (of harmful health effect) or not depends on the magnitude of the exposure in relation to the toxicological reference value of the chemical. Heavy metal dietary exposure is based not only on the content of the target chemical in studied food, but also on the consumption frequency, and on the amount of food consumed expressed by the body weight. The frequency of consuming cereal and crop-based food may increase in response to healthier and more environmentally friendly dietary choices. The effects of using BBFs on HM dietary exposure in dairy or meat products were not the scope of this study, but they may increase the HM dietary exposure. Hence, reducing the HM content BBFs will reduce the HM content in crops, and hence food products will be more secure to consume.

In general, the use of BBFs referred in this paper seems to be safe from the perspective of HM content in crops, since BBFs use is unlikely to affect the total dietary exposure to Pb, iAs, Ni, and, in the case of BBF_L, also to Cd. The use of BBF_H was estimated to increase the Cd exposure to all studied cases, but specially of Finnish children, which is likely due to the large contribution of wheat, oat, rye and barley on the total exposure of this age group: cereals and cereal products together contributed ca. 30% to the total dietary exposure assessed in Suomi et al. (2015 and 2018). BBF-H increased the cadmium content in cereals by ca. 20%. The rough estimates of other countries give an approximate effect of the BBFs, thus with more detailed consumption data, the estimate could have been more exact.

Conclusion

We studied the current occurrence of Cd, Pb, iAs, and Ni in wheat, rye, barley, oats, maize, potato, carrot and sunflower seeds from five European countries. The mean contents were presented in Figure 1, which shows clear differences between the countries due to, among other things, variance in the soil characteristics, HM contents due to fertilization or environmental pollution, and the variability between cultivars to take up HMs from the soil.

The effects of mineral superphosphate as well as biobased fertilisers with high (H) or low (L) HM content on the HM contents in crops in a 100-year time scale was estimated, and also the effect on exceedance of the EC regulated maximum levels of Cd and Pb in crops was assessed. In general, the low-HM BBF decreased the HM contents taken up by crops compared with SP fertilization, and this would lead to a lesser portion of crops being rejected due to exceedance of the Cd maximum levels. Correspondingly, the use of high-HM BBF would increase the amount of crops unfit for human consumption due to too high Cd content (Table 3). For Pb, the BBFs and the SP fertilization did not differ in their effect on rejected sample lots.

The dietary exposure of adults and children of six European countries was assessed with the estimated changes in the HM contents in crops in a 100-year time scale assuming that both the consumption habits and the HM exposure from other foods would remain constant over the time. The HM exposure through the studied crops (Tables 4a and 4b) was estimated to mainly decrease the total dietary exposure slightly after use of BBF_L compared with SP, although there were differences between the countries. The effect on the total dietary exposure was, however, low even for BBF_H except for Finnish children.

For Finnish adults, the estimated dietary HM exposure with FinDiet 2017 food consumption data (Table 7) was, at population median level, only one-third of the tolerable weekly intake of Cd and less than one-quarter of the tolerable daily intake of Ni, which means that neither Cd nor Ni are a health risk for the median of adult population. The dietary exposure to Pb and iAs exceeds, even at the population median level, the exposure at which the health risk would be negligible. This is in line with the previous national assessments (e.g. Suomi et al. 2021), based on which the dietary Pb exposure is estimated to cause a burden of disease of 570 DALY annually (Suomi et al. 2019). However, BBFs were not estimated to increase the dietary exposure to Pb or iAs.

In conclusion, it seems that the use of well processed and low HM-concentrated biobased fertiliser is safe from the point of view that the dietary exposure to HM did not increase.

Acknowledgements

This project received funding from the European Union’s Horizon 2020 research and innovation programme under grant agreement No 818309 (LEX4BIO). The results reported in this paper reflect only the authors’ views, and the European Commission is not responsible for any use that may be made of the information it contains.

The authors thank the participants of FinHealth 2017 for their participation in the dietary recall and in the THL Biobank (www.thl.fi/biobank). This article used the data from THL Biobank (study number THLBB2022_20).

The data providers of Denmark (Danish Veterinary and Food Administration), Germany (Federal Office of Consumer Protection and Food Safety BVL), Hungary (National Food Chain Safety Office Nébih), Spain (AESAN) and Finland (Finnish Food Authority, Customs Laboratory, Finnish Cereal Committee) are gratefully acknowledged for the data from EFSA Call for continuous collection of chemical contaminants occurrence data in food and feed 2000–2021 as well as national monitoring data from Finland.

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