Estimation of heavy metals emissions in agricultural productions: The case of Italian products

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ARTICLE INFO

ABSTRACT

The agri-food sector is more complex than the industrial sector due to different climatic, geomorphologic and cultivation practice factors. Agriculture operations can significantly affect phenomena, such as erosion and leaching, and generate different types of emissions into the environment. These aspects should be considered when performing a Life Cycle Assessment (LCA) study on agri-food products, mainly because these elements are influenced by both natural and human factors that generate negative health and ecotoxicological impacts. The "Swiss Agricultural Life Cycle Assessment - heavy metals (SALCA-HM)" allows for calculating heavy metal emissions at the life cycle inventory level as part of LCA at a regional level. This study aims to customise the SALCA-HM for five Italian agricultural products (i.e., durum and common wheat, grapes, olives, and citrus fruits), using region- and crop-specific data. The results showed that, even though all the factors relating to cultivation techniques are constant, the use of site-specific data makes it possible to highlight the influence of the orographic characteristics of the territory. Therefore, the high variability of the results can be perceived as a strength of this regionalised approach, thus overcoming several limitations by using national average data instead. More effort is needed to enable greater data availability both for policy and the scientific community.

1. Introduction

Life Cycle Assessment (LCA) (compliance to ISO 14040:2006 and ISO 14044:2006) (ISO, 2006a; ISO, 2006b) allows for considering all inputs and outputs related to the whole life cycle of products, processes and services, taking into account geographical, temporal, and technological aspects. Even though LCA has been increasingly used to estimate the potential environmental impacts, several issues remain unsolved. One is the lack of secondary data to build appropriate Life Cycle Inventories (LCIs). Among the production activities that most need specific inventory data is the agri-food sector, where production is closely inter-linked with the biological characteristic of the farming system and, therefore, highly dependent on the specific conditions occurring in a given territory. It is, therefore, necessary to promote extensive data-collecting activities that can represent the geographical and

technological characteristics of regional agri-food productions (Notarnicola et al., 2022a; Mondello et al., 2022; Vono et al., 2022; Notarnicola et al., 2017; D'Eusanio et al., 2022). Indeed, the primary data collection is often integrated with secondary data for modelling the background data inventory taken from the scientific literature or commercial databases, such as GaBi (Kupfer et al., 2021) and Ecoinvent (FitzGerald and Sonderegger, 2022). Such data are often not generally applicable to all product systems, especially concerning the agri-food sector, which is characterised by the extreme complexity of several factors, such as geographical territoriality, climatic conditions, soil types and different cultivation practices. Indeed, these databases do not always provide fully representative data on the site-specificity of the analysed agri-food products (Notarnicola et al., 2017).

In this framework, developing specific agri-food databases that would consider the territorial peculiarities of the involved agri-food

supply chains becomes essential. Several countries have developed specific inventory databases for agri-food supply chains (e.g., Agribalyse; LCA Food DK; Agri footprint) (Notarnicola et al., 2022b) in order to improve the quality of the available data and to represent the peculiarities of their products as well as the processes of the supply chain in a better way (Morais et al., 2016). To date, the LCA studies that consider Italian agri-food product systems mainly have used secondary data from international databases (Notarnicola et al., 2017), such as Ecoinvent (FitzGerald and Sonderegger, 2022) or GaBi (Kupfer et al., 2021). To this end, "PRIN 2017 - Project of Relevant National Interest", entitled "Promoting Agri-Food Sustainability: Development of an Italian Life Cycle Inventory Database of Agri-Food Products (ILCIDAF)", aims at creating a regionalised Italian database for the agri-food products that would capture these peculiarities. The ILCIDAF database will be built for the wine, olive oil, citrus and wheat-derived product supply chains, with a system boundary starting from the agricultural phase to the product-use phase and the final waste disposal (end of life).

Agricultural operations significantly affect natural phenomena, such as erosion and leaching, and generate different kinds of emissions into the environment (Gulisano et al., 2018). The effects on the environment of the combustion of fuels, the application of plant protection products and fertilisers, the removal of grass tissue and various soil management techniques are widely studied by developing several estimation models. However, some critical aspects of estimating emissions from agricultural activities remain open. Indeed, when performing an LCA study on agri-food products, quantifying data related to heavy metals present many criticalities because these elements are affected by both natural and anthropic sources that generate serious negative health and eco-toxicological impacts (Filote et al., 2021). The "Swiss Agricultural Life Cycle Assessment - heavy metals (SALCA-HM)" model (Freiermuth, 2006), which is functional to the agri-food databases, is typically used for calculating heavy metals emissions (Notarnicola et al., 2022a) at the life cycle inventory level as part of LCA (Freiermuth, 2006). This model is limited to estimating the heavy metal flows into agriculture soil, groundwater, and surface waters, taking into account both natural phenomena (i.e., erosion and leaching) and anthropic products (e.g., seeds, fertilisers, plant-protection products, etc.) (Freiermuth, 2006).

Often, however, the application of the model is carried out using data not specific to the area under study, using Swiss agriculture data. Indeed, in many cases, the SALCA model has been implemented using national average data (e.g., Ecoinvent or World Food LCA), through which it is impossible to represent the area's specificities. This is a fundamental criterion for agricultural production, where the link with the territory where the agricultural activities are carried out is unavoidable.

In fact, heavy metals emissions are strongly influenced by geographical factors such as the orography of the territory, the type of soil, and exposure to atmospheric events that can increase or decrease the atmospheric deposition of these metals; so, referring to Italian agriculture, the link with the territory becomes even stronger, given the high heterogeneity of the Italian orography due to the complex geological and climatic evolution that this territory has undergone. From a purely geological point of view, the physical conformation of the peninsula has been determined by the collision of the African and Eurasian plates, generating, in particular, the Alpine and Apennine Mountain, which extend throughout the territory. Amplifying the territorial and landscape heterogeneity is also the great variety of lithotypes (Marchetti et al., 2017). The different altitudinal ranges (from 0 to over 4,800m), the presence of the sea along almost all of the country's boundaries and the great latitudinal extension strongly influence the climatic conditions of the Italian regions and cities (Fratianni et al., 2017). Based on the overview just described, it is essential to define an approach for estimating heavy metal emissions that considers this great variability in regions of Italy.

Therefore, the study aims to customise the SALCA-HM model (Freiermuth, 2006) for estimating emissions from Italian agricultural production processes, highlighting the use of region- and crop-specific data. In particular, the SALCA-HM model will be applied to the five most representative agri-food chains of the Italian food sector (i.e., durum and common wheat, olives, grapes, and citrus fruits) by describing how to identify the data and highlighting the variability of the results on a regional scale.

2. Material and methods

The following section provides i) a brief description of the widely used SALCA–HM model, ii) the procedure developed for selecting data for the Italian regions, and iii) the description of five agri-food products to which the methodology is applied.

2.1. Brief description of the SALCA-HM

The SALCA-HM model (Freiermuth, 2006) is applied to estimate heavy metal emissions in ILCIDAF agricultural datasets. The heavy metals considered by the SALCA model are Chromium (Cr), Cadmium (Cd), Lead (Pb), Nickel (Ni), Copper (Cu), Zinc (Zn) and Mercury (Hg) (Freiermuth, 2006). A simplified representation of the processes considered by the SALCA-HM model is summarised in Fig. 1.

The model allows for calculating three types of emissions: i) leaching of heavy metals to groundwater (M_{leach}), ii) run-offs of heavy metals into surface waters through erosion phenomena ($M_{erosion}$), and iii) emissions to the agricultural soil resulting from the mass balance of heavy metals (M_{soil}). These contributions are weighed for an allocation factor (A_i) that allows for evaluating only the direct contributions of agricultural activities. It is calculated as the share of agricultural inputs (seed, fertiliser, pesticides, and other substances reversed on soil) in the total inputs (agricultural input plus atmospheric deposition).

Indeed, the leaching phenomenon releases heavy metals into deep water, provided the soil is not drained. This parameter is calculated as average amounts of heavy metals leached ($m_{leach,i}$) for kilograms of soil. This factor depends on soil structure, crop planting, type and application rates of fertilisers and other factors such as irrigation and rain.

While the erosion phenomenon releases the heavy metal removed from soil that can be attributed to cultivation, it is the function of different factors that are affected by soil characteristics and human intervention. It is calculated following the USLE model (Freiermuth, 2006; Alewell et al., 2019), then multiplied for three factors that describe: i) the concentration of heavy metals in the soil ($C_{tot,i}$), ii) the distance from water sources ($f_{Erosion,i}$), and iii) the enrichment of heavy metals on the more easily eroded clay-humus complexes (a). In particular, the main factors that depend on geographic locations and that affect this phenomenon are:

- R (MJ mm h^{-1} ha^{-1} yr^{-1}) is the rainfall-runoff erosivity factor that captures the energy and amount of precipitation,
- k (Mg h MJ^{-1} mm^{-1}) is the soil erodibility factor that, accounting for the soil parameters, determines erosion potential,
- LS (dimensionless) is the slope length and steepness factor,
- C (dimensionless) is the land cover and management factor that describes the vegetation cover and management,
- P (dimensionless) is the soil conservation or prevention practices factor that delineates human management intervention,
- $C_{tot,i}$ (mg kg^{-1}) is the concentration of heavy metals in the soil, generally estimated through chemical soil analysis.

Finally, the latter quantified emission is the amount of heavy metals released into the soil. It, resulting in positive or negative contributions, is calculated as a balance among the mass of heavy metals of agricultural activities in input and that loss through leaching, erosion, and plant harvesting. Only the amounts of heavy metal uptaken from products and by-products that leave the soil are accounted for the latter. All the equations used to estimate heavy metal emissions (Freiermuth, 2006)
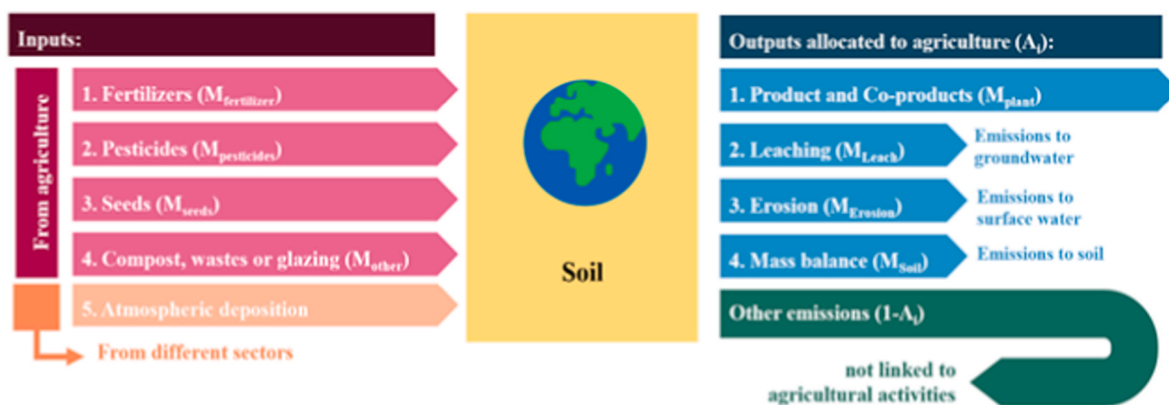
Fig. 1. A simplified representation of heavy metal flows through the soil.

are listed in the supplementary materials (SM).

As mentioned above, the factors used in the model are highly dependent on soil and climatic characteristics as well as on geographic, temporal, and technological features of agricultural activities. Their representativeness is crucial for consistently applying the model and, consequently, being correct and geographically representative of potential heavy metal emissions. In addition, agricultural operations depend on the type of crop and the orographic of the land (whether predominantly flat or hilly). The agricultural substances applied to the soil vary as the crop varies, but also over time and for a given area/region (Notarnicola et al., 2022a). Therefore, for the Italian case studies application, almost all the factors involved in the SALCA-HM model are specific for the regions and per crop type. Additional details are reported in the section below.

2.2. Selection of data for the Italian regions

Given the presence of complex orographic systems and the high variability of climate conditions in Italy (Khan and Chiti, 2022), to provide affordable data and create representative LCI for the Italian agri-food sector is necessary to evaluate the heavy metals emissions throughout agriculture activities using site-specific regional data. Therefore, the available heavy metals and soil properties databases have been searched to extract these data. Combining all these files allows for

identifying average values for region and cultivar. The methodology used in this study to extract and collect data and calculate heavy metal emissions could be entailed in five steps, summarised in Fig. 2.

2.2.1. Geospatial data on cropland types

Considering that the scope of the study is strictly connected to the ILCIDAF project, it was necessary to identify data representing the geographic locations of different croplands in the Italian regions to evaluate their average soil characteristics specific (when possible) for cultivars.

For this purpose, the database LUCAS (Land Use and Coverage Area frame Survey) topsoil (Orgiazzi et al., 2018) is used. It provides information on the spatial distribution of different crop types for different soil point data in Europe, representing one of the biggest integrated continental-scale soil inventories due to the various characteristics examined (Orgiazzi et al., 2018; Panagos et al., 2021). The collected data comprise stratified random samples, classified based on their theoretical geographic coordinates (latitude and longitude) (Ballabio et al., 2021). The database is downloaded in both shapefile and MS Excel format because the first contains only the LUCAS 2018 POINTID, while the second also includes information on soil type, the nomenclature of territorial units for statistics formats (NUTS3), main land cover class (e. g., forest land, cropland, etc.) and detailed land cover class description.

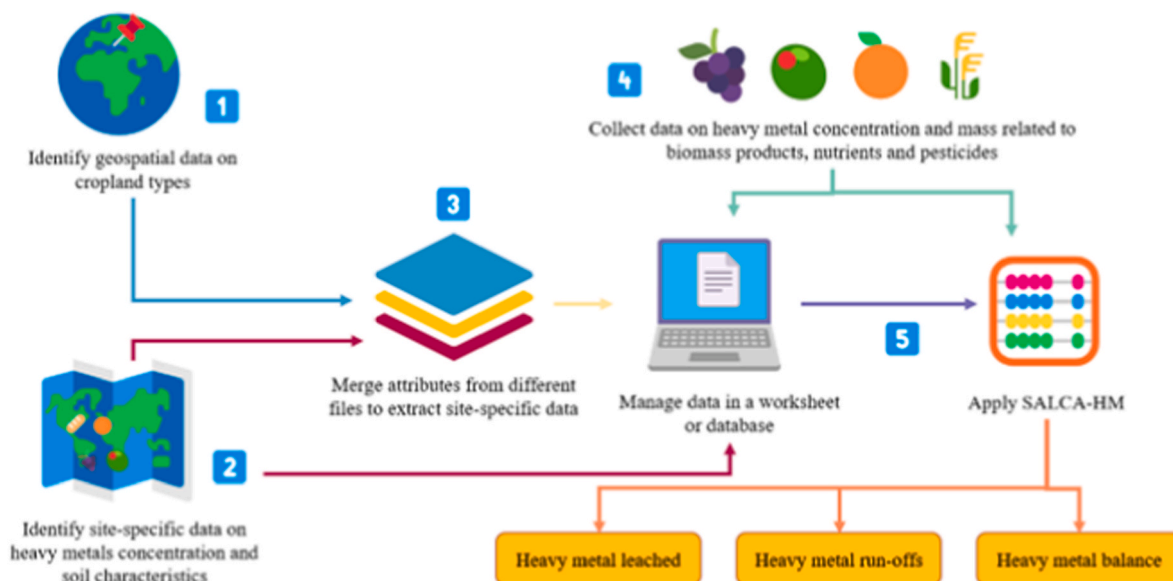Focusing on the cropland cover class, LUCAS hosted 32 sub-classes:

Fig. 2. Workflow of extracting site-specific data on heavy metals.

from cereal to fruit trees. For Italy, LUCAS includes 628 samples in the class cropland, as reported in Fig. 3.

In addition, LUCAS is also a fully accessible repository tool that contains data from the European Soil Data Centre (ESDAC) that is freely available (Panagos et al., 2022). ESDAC represents the focal point for soil data for supporting policy making and awareness raising for the European Union (EU), currently hosting 88 datasets, 6000 maps, 6 atlases, 500 scientific publications, and a copious amount of soil-related material (Panagos et al., 2022).

2.2.2. Heavy metals concentration and soil characteristics

To collect heavy metals data and soil properties, it is necessary to identify different databases that provide at least data at the regional level (e.g., raster maps). Therefore, the soil and heavy metal datasets developed by the European Commission (European Commission, 2022), JRC (Panagos et al., 2022), the European Environmental Agency (EEA) (EEA, 2020; EEA. CORINE, 2022), and the National Institute for

Environmental Protection and Research (ISPRA) (Italian, 2021) are investigated. From these datasets, it is possible to identify specific data (such as heavy metals soil concentration, leaching or atmospheric deposition) at the regional level and for crop types, then used to estimate heavy metals leaching and run-offs due to soil erosion processes.

In particular, concerning the erosion process, most of the data are extracted from the ESDAC database. As mentioned above, ESDAC contains datasets and maps related to heavy metals concentration in soil, and other soil properties developed complementary to the LUCAS database and Corine datasets (Panagos et al., 2022). Regarding heavy metals, it includes data on the concentration in the soil of the following elements: Arsenic (As), Cadmium (Cd), Cobalt (Co), Chromium (Cr), Copper (Cu), Iron (Fe), Mercury (Hg), Magnesium (Mg), Manganese (Mn), Nickel (Ni), Lead (Pb), and Antimony (Sb) (Panagos et al., 2021). Only the maps for six metals (i.e., Cd, Cu, Cr, Hg, Ni, and Pb) are downloaded for the ILCIDAF project in Raster format after compiling a request form on the ESDAC website. However, no data was found for Zn

Cropland sub-classes / regions	Abruzzo	Basilicata	Calabria	Campania	Emilia Romagna	Friuli Venezia Giulia	Latium	Liguria	Lombardy	Marche	Molise	Piedmont	Apulia	Sardinia	Sicily	Tuscany	Trentino South Tyrol	Umbria	Aosta valley	Veneto	Italy
Apple fruit				1	1									1			8				11
Barley					3	2			4	1		7		1	1	1				1	21
Cherry fruit													1			1				1	3
Clovers									2						1					1	4
Common wheat					7	3	1		5			6		1						5	28
Dry pulses	1	3			2	1				2	1				3	2		2			17
Durum wheat	1	3		2	4					6	8		12	1	14	3		1		1	56
Lucerne	1				20	3	3		7	3	1	1		1		2				6	48
Maize					1	10	4	1	28			16	2					1		21	84
Mix of cereals		5	1	2			5					2	11	2	6					1	35
Nurseries						2															2
Nuts trees	1			1			2						4								8
Oats			1	1			2						1		1	1					7
Olive groves	3	5	13	2	1		4	1			1		42	2	15	8		2			99
Oranges				1																	1
Other cereals					2					1											3
Other citrus fruit			1																		1
Other fresh vegetables	1			1			1		2					3	1	1				1	11
Other fruit trees and berries		1		3	1		1					3	2		3					1	15
Other Leguminous and mix	1		1	2			1		1				2		5	1		2			16
Pear fruit					3							1									4
Potatoes			1		1																2
Rice									3			4									7
Rye						1															1
Soya					4	2			1			1								13	21
Strawberries				1																	1
Sugar beet									1												1
Sunflower										2	1	1									4
Temporary grassland			2	3	1	1	3		2	1		4		1	1	1		4		2	26
Tomatoes	1		1	2					1		1	1								1	8
Triticale							1											1			2
Vineyards			2	2	7	3	1		5	1		9	3	2	12	11	10	1	1	11	81
Total	12	18	22	19	71	20	28	1	60	16	14	60	80	15	62	35	19	9	1	66	628

Fig. 3. Number of samples in LUCAS for regions and cropland sub-classes.

in the ESDAC database. In this case, the concentration of Zn in the soil at the regional level is extracted from the datasets proposed by the EEA on concentrations of heavy metals in European agricultural soils (EEA, 2020). They all represent the parameter called concentration of ith heavy metal in the soil ($C_{TOT,i}$) (Tóth et al., 2016). Instead, concerning the soil properties, the ESDAC database provides datasets on some parameters of erosion calculation, such as LS-Factor, C-factor, or R-Factor. Table 1 (section 2.2.5) summarises other details on sources used.

Considering the leaching process, it was necessary to identify the amount of heavy metal leaching for each region and crop ($m_{Leach,i}$). The datasets proposed by the EEA are used for this purpose, as made for Zn concentration in soil (EEA, 2020). This data collection involves information on current and critical metal concentrations in topsoil, metal inputs to and outputs from soils (i.e., uptake, accumulation, and leaching) and the impacts of exceeding critical metal inputs. However, the leached metals included in this dataset are Cd, Cu, Pb and Zn, and no other datasets at the European level have been identified on it. Thus, the missing metals are assumed from (Freiermuth, 2006; Wolfensberger and Dinkel, 1997), in which no Ni data is available.

The atmospheric deposition of heavy metals represents the balance of anthropogenic emissions and secondary emissions (wind re-suspension of dust particles containing heavy metals) received from activities inside and outside the country or transferred to other countries (Ilyin et al., 2021). Due to missing data on atmospheric deposition temporal and geographic representation of the actual Italian regional situation (Ilyin et al., 2021; EMEP, 2021), the heavy metal deposition amounts are assumed to be the heavy metal emissions estimated by ISPRA at regional scales for 2019 (Italian, 2021) and totally deposited on regional soil. This assumption allows for accounting that heavy metal emissions have been strongly reduced since 1990 in all the Europe and that Italy is characterised by a very diversified emission context at the territorial level (e.g., Lombardy originates the largest shares of emissions of all metals except for arsenic, deriving for the most part from Puglia).

2.2.3. Merging attributes and extracting site-specific data

The datasets downloaded from ESDAC and EEA and the LUCAS soil data points vector are read through QGIS software to collect site-specific data. QGIS is an Open Source Geographic Information System that provides common GIS functions and features such as data capture, advanced GIS analysis, map presentations, atlases and reports, and supporting raster and vector data formats (QGIS v3.28, 2021). The software allows for extracting and merging attributes using the following functions:

1. for raster files (ESDAC maps), the "*sample raster values*" function is used to extract for any point the attributes contained by the raster pixel value at the location of the point reported in LUCAS,
2. for vector files, the "*join attributes by location*" function is used to extract the average values for data that intersect, are contained, or touch the location of the point reported in LUCAS.

The resulting attributes are extracted and copied into an MS Excel worksheet, in which any point is associated with the different classes reported on LUCAS (MS Excel file). Then, using pivot tables, it was possible to calculate quickly the average values divided for regions and crops, excluding all the Italian data on, e.g. forest or artificial land.

2.2.4. Collecting heavy metals and mass data on biomass, nutrient and pesticides

Although all previously mentioned data have been collected accounting for region soil characteristics and cropland management, the concentration of heavy metals in biomass products, nutrients and pesticides is based on (Freiermuth, 2006; Nemecek et al., 2019; Koch and Salou, 2016). Despite this, it was necessary to collect some data from the literature to account for all the agri-food products included in the ILCIDAF project. In particular:

- the concentration of heavy metals in olives is taken from (Luka and Akun, 2019), and missing data for Hg and Zn are assumed as grapes values in Agribalyse (Koch and Salou, 2016),
- the concentration of heavy metals in citrus fruit is taken from (Özcan et al., 2012), and missing data for Hg and Cd is taken from (Khudair, 2021),
- considering no mercury is reported for mineral fertiliser in (Freiermuth, 2006), the mercury concentration in nutrient NPK is taken from the analysis conducted by the Washington State Department of Agriculture (WSDA) (METALS, 2018),
- while no additional data are added to compensate for insufficient data on heavy metals for pesticides, only limited data on Cu and Zn concentration are included (Freiermuth, 2006).

Indeed, the kilograms of inputs and outputs through the system boundaries (soil) must be collected for crops, respecting yield production and admitted nutrients and pesticides for hectares of cultivated land. Other information on collecting these inventory data in practices for Italian agri-food products is reported in section 2.3.

Table 1
Summary of data collection for heavy metals emissions calculation.

Data types	Unit	Data specific for	Sources
Geospatial data			
Cropland point	-	Latitude and longitude for regional and crops points	Orgiazzi et al. (2018)
Erosion			
LS-factor	–	Regional and crops	(Panagos et al., 2015a, 2022)
C-factor	–	Regional and crops	(Panagos et al., 2015b, 2022)
k and k stoniness factor	[(t•h)/ (MJ•mm)]	Regional and crops	(Panagos et al., 2014, 2022)
$C_{tot,i}$	[mg/kg] [(1)]	Regional and crops for metals	(Panagos et al., 2022; EEA, 2020; Tóth et al., 2016) [c]
R factor	[(MJ•mm)/ (ha•h•yr)]	Regional and crops level	Panagos et al. (2015c)
P-factor	–	Regional and crops level	(Panagos et al., 2020, 2022)
$f_{Erosion,i}$	–	Default value (0.20)	(Freiermuth, 2006; Koch and Salou, 2016)
A	–	Default value (1.86)	Prasuhn (2006)
Leaching			
$m_{leach,i}$	[kg/(ha•yr)]	Metals for regions and crops	(Freiermuth, 2006; EEA, 2020) [d]
Plant and agro actives			
$c_{Plant,i}$, $c_{seeds,i}$	[mg/kg] [a]	Metals for crops, seeds	(Freiermuth, 2006; Koch and Salou, 2016; Özcan et al., 2012)
$c_{Fertiliser,i}$	[mg/kg] [a]	Metals for fertiliser	(Freiermuth, 2006; Koch and Salou, 2016)
$c_{Pesticides,i}$	[mg/kg] [b]	Metals for pesticides	(Freiermuth, 2006; Koch and Salou, 2016)
F	–	Percentage of active substance transported from pruning and harvesting (5%)	(Freiermuth, 2006; Koch and Salou, 2016)
$m_{Deposition}$		Metals emitted for regions	Italian (2021)

[a] Concentration of heavy metals for dry matter.
[b] Concentration of heavy metals for active substances.
[c] Concentration of Zn into the soil from EEA.
[d] Concentration of leached heavy metals (Cr, Cu) from SALCA and the others from EEA report.

2.2.5. Data management and application of the SALCA method

All the equations of the method SALCA for Italian agri-food products are currently implemented as a set of almost fully automated MS Excel spreadsheets, in which all the data collected are organised in tables for each parameter. All the data types and sources are summarised in Table 1.

Indeed, manual operations are needed for entering the inventory data that affects heavy metals in input (i.e., mineral and organic fertiliser, pesticides, seeds, and eventually, other biomass distributed on soil) and in output (i.e., biomass such as wood pruned and agri-food products). At the same time, the other operation is strictly linked to selecting cropland sub-classes from lists. This action allows the selection of different parameters specific for crops to be applied to all regions; thus, the heavy metals leaching, run-off for erosion and emitted in the soil are automatically calculated. So far, the calculations are applied to Italian crops (i.e., olives, citrus, grapes, common and durum wheat), showing easy applicability and saving time for future applications. In addition, in case of missing specific regional data for some crops, data could be integrated with regional average data without crop detail or national average data for crops.

2.3. Cases studies application

The above methodology is applied to five Italian agricultural products (durum and common wheat, grapes, olives, and citrus) to observe the variability of heavy metal emissions generated by crops and regions' soil characteristics. The following data are required for calculating heavy metals: seeds, fertilisers, and pesticides as input to soil and production yields as output from the soil.

The datasets for each crop are elaborated by combining statistical and secondary data provided by: i) the Italian statistical database (ISTAT, 2021), ii) Integrated Production Regulations (IPR) (MASAF, 2022), and iii) a technical handbook of agriculture (Ribaudo, 2017).

In particular, the data obtained from the ISTAT database includes the cultivated land and the amount of produced agricultural products between 2015 and 2020. These data on productivity allow for considering medium-long-term values for reducing the influence (negative or positive) caused by the yearly fluctuation. Instead, IPR recommendations are used to calculate the quantity of fertiliser applied per hectare. For the study, only mineral fertilisers are accounted for analysis. Instead, from Ribaudo (2017), data on pesticides are extracted for each crop. However, considering that differences exist between crop yields resulting from ISTAT and that reported in (MASAF, 2022; Ribaudo, 2017), fertilisers and pesticides are linearly scaled to account for that.

These data have been first calculated as "one cultivated hectare during one season" for permanent fruit (i.e., olive tree, vineyards, and citrus tree) and "one cultivated hectare during one growing season" for annual crops (durum and common wheat grain). Then, the functional unit "1 kg of product" is used to show the analysis results.

Relative to some SALCA-HM factors, the following assumptions are made: i) the erodibility factor is assumed to be equal to k-stoniness specific for regions and crops, ii) the cover management factor is assumed to include all the reductions made from tillage activities specific for crops and regions, iii) all factors are assumed specific for regions and crops, and iv) when data missing, the gaps are covered using national average data for the specific crop. A limitation of this approach is the case of oranges. In fact, for these crops, specific data are available for only one region that also coincides with the national average, as this is obtained from a single value (see Fig. 2).

In order to show the variability of results among regions, statistical analyses are applied to each crop. In particular, the MS Excel function "descriptive statistical analysis" is applied to calculate the mean, standard error, median, standard deviation, sample variance, range, minimum and maximum values, sum, count and confidence level (95%). To evaluate the variability magnitude of results for HM and type of crop, a variability index (VI) is calculated as in equation (1).

$$i, n = \frac{i, n}{^\wedge}$$ Eq. 1

where:

- StD is the standard deviations calculated for i-th crop product and n-th heavy metals;
- Mean is the mean values calculated for i-th crop product and n-th heavy metals.

Furthermore, a sensitivity analysis is conducted to highlight the factors' variability at the regional level, maintaining constant input and outputs for all scenarios. More details are reported in section 3.2.

3. Results and discussion

This section presents the calculated heavy metal emissions for the five studied crops, showing first the contribution of each heavy metal to the global masses emitted for leaching, run-offs and balance to the soil, then the variability of data at the regional level for each crop. The quantitative results of applying the SALCA model to the Italian regions are presented in SM. Furthermore, the last section illustrates the challenge and limits of this study.

3.1. Variability for regions

3.1.1. Durum wheat

Fig. 4 shows the estimated contributions of each heavy metal eroded, leached and emitted to the soil per 1 kg of durum wheat produced in 17 Italian regions (excluding Aosta valley, Trentino South Tyrol and Liguria). As it is possible to observe, the results show that the regional scale variability is significant. Full data in MS (Tables 1,2, 3).

Focusing on total eroded heavy metals, the highest quantity of HM per kg of durum wheat occurs in Marche (0.406 g), Tuscany (0.334 g), and Calabria (0.306 g). Among these regions, Zn represents the highest contribution with an average percentage value of 29.7%, and in 8 regions, its contribution is over 30%. At the same time, the second highest contribution (27.6%) is generated by Cr (in 10 regions, this value ranges from 25.6% to 39.2%). Then, Ni, Cu, and Pb contribute respectively with 20.5%, 16.3% and 5.9%, while Cd and Hg represent the lowest mass contribution.

For leached heavy metals, the highest value occurs in Campania (0.02 g), followed, as for erosion, by Tuscany (0.017 g) and Calabria (0.015 g). The largest contribution to this process is Cr, with an average percentage of 52.4%. Also, in this case, it is highly variable among regions. In fact, the range varies from a maximum of 86.9% for Sicily to a minimum of 26.3% for Veneto. In general, for 12 regions, the percentage contribution is over 40.0%, while in 5 regions, it is over 60.0%. Cu and Zn also contribute significantly and with a very similar average percentage contribution of 21.3% and 21.0%, respectively. For Cu, 11 regions have a value greater than 20.5%, while for Zn, the regions rise to 12. Even the range of values is comparable (6.4%–33.2% for Cu and 5.4%–35.6% for Zn). Lead contributes, on average, 4.6%, while Cd and Hg have a contribution below the percentage value. Considering the soil balance of heavy metals, common wheat and durum wheat are globally negative in all regions and for almost all metals.

Fig. 5 shows the results of the descriptive analysis for the 17 Italian regions mentioned above, focusing on the three contributions (erosion, leaching and soil). Greater variability is obtained for Hg emitted to soil and leached with a percentage value of 785.9% and 141.3%, respectively. This high variability is due to the modelling approach used for the durum wheat, which, given their seasonality, differs from the other crops under study. Indeed, the quantity of seeds (with their heavy metal content) is provided as input, while straw is provided as output in addition to wheat. The highest yield variability for regions strongly
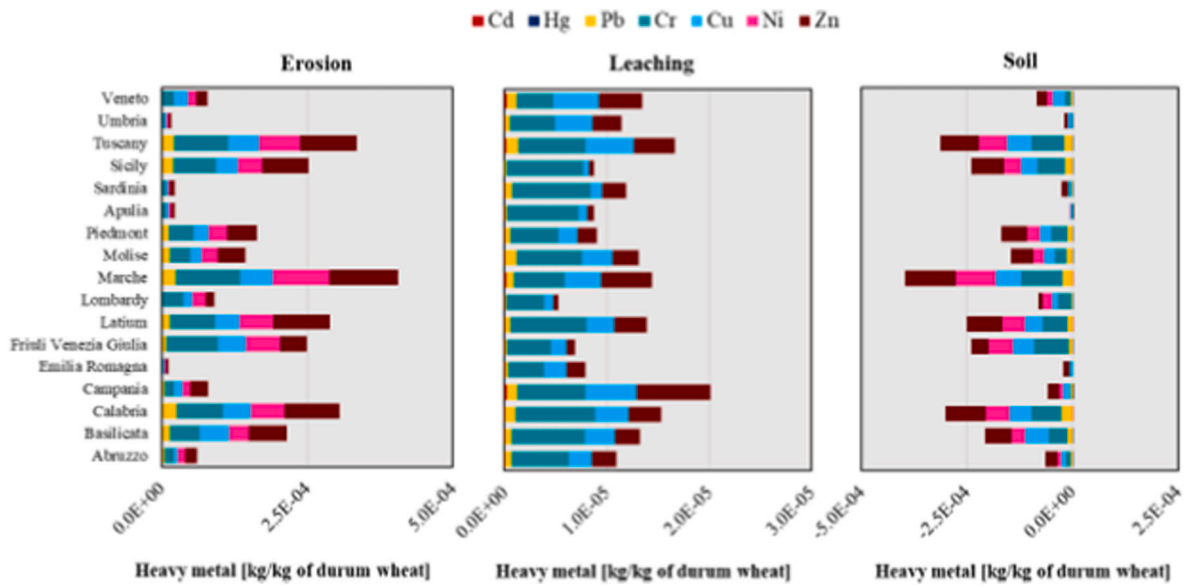
Fig. 4. Contribution analysis of heavy metals emissions for 1 kg of durum wheat among 17 Italian regions.
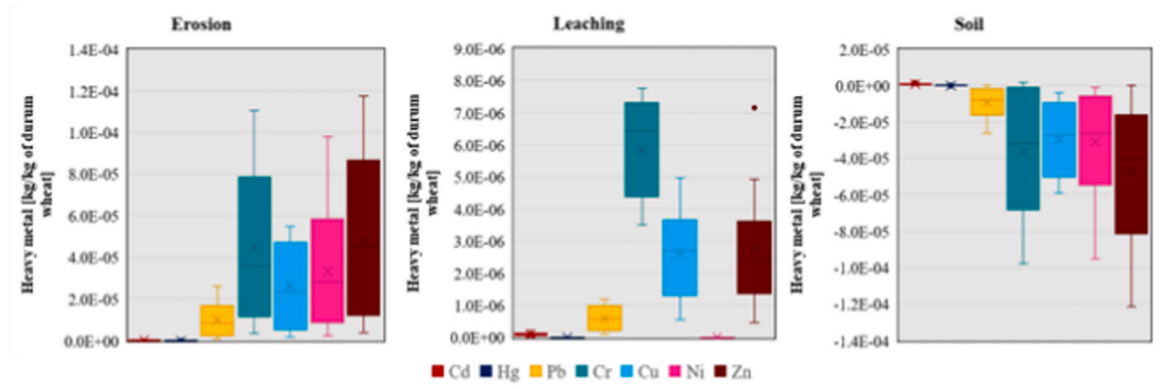

Fig. 5. Box Plot of heavy metals emissions for 1 kg of durum wheat among 17 Italian regions.
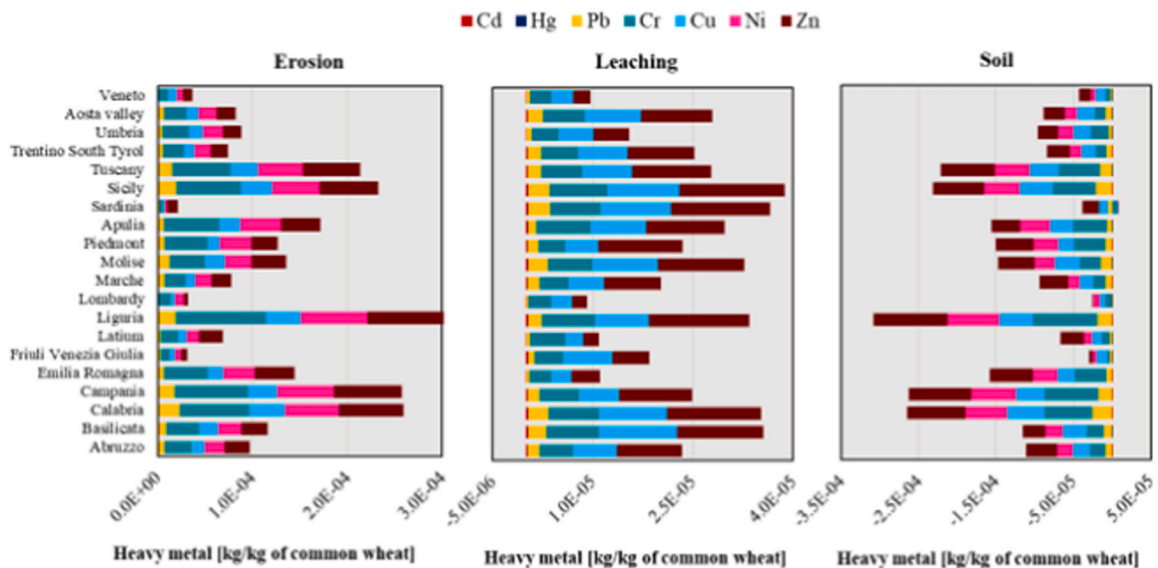

Fig. 6. Contribution analysis of heavy metals emissions for 1 kg of common wheat among 20 Italian regions.

affects the soil balance results and the allocation factor, A_i. The latter factor for mercury strongly varies for regional scale ranging from 2% in Lombardy to 54% in Marche, while Ai has a lower variability for the other metals.

The lowest variability in all phenomena is obtained for leached Cr (27.0%), while the other metals vary from 53.3% to 92.4%.

3.1.2. Common wheat

Tables 4, 5 and 6 in the SM show the results of heavy metal emissions per kg of harvested common wheat in 20 Italian regions for erosion, leaching and soil balance processes. The results show a high variability on a regional scale. Considering the erosion phenomenon, Liguria is the region for which the highest amount of total heavy metals per kg of harvested common wheat is obtained (0.311 g), followed by Calabria (0.259 g) and Campania (0.258 g). Fig. 6 graphically represents the quantities of eroded, leached, and to soil heavy metal for each region for common wheat production. The metal that contributes most to this process is Cr (30.3%–40.0% in 8 regions) and Zn (greater than 25.4% in 13 regions). It is followed by Ni (22.5% on average), Cu (14.0% on average) and Pb (6.9% on average). Cd and Hg contribute an average percentage of less than 1%. Considering the leaching phenomenon, the region with the highest amount of total heavy metals leached in Sicily, with a value of 0.039 g per 1 kg of wheat, followed by Sardinia (0.037 g) and Basilicata and Calabria (0.035 g). The highest contributions for this process are attributed to Zn (average value of 38.2% and in 9 regions with a value greater than 40%), Cu (average value of 28.9% and in 11 regions with a contribution between 28.1% and 39.7%) and Cr (average value of slightly less than 25.2% and in 6 regions with a contribution greater than 25%). They are followed by Pb and Cd (with an average value of 6.5% and 1.2%, respectively); while the contribution of Hg is less than 1%. On the contrary, the heavy metal balance in the soil is negative overall in all regions and for almost all metals. The highest value is obtained for Lombardy, while the lowest value (highest in absolute value) is in Liguria and Calabria.

Analysing the individual heavy metals, Fig. 7 shows the results of the descriptive analysis of heavy metal emissions per kg of wheat for all Italian regions considering the three contributions (erosion, leaching and soil). High variability of the results is obtained. The greatest variability is observed for mercury emitted in soil, with a ratio of standard deviation to mean of about −719.8% of the mean value, followed by eroded Hg (98.2%). For common wheat, the results lead to observations similar to those described above for durum wheat. The lowest variability is recorded for leached heavy metals, such as Cr (30.9%), Cd (38.1%), and Cu (39.9%). All other parameters vary between 45.8% and 80.6%.

3.1.3. Grapes

Fig. 8 and Table 3 (SM) show the mass of heavy metal emissions per kg of grapes for all 20 Italian regions regarding erosion, leaching and soil. The analysis demonstrated that for those deriving from soil erosion due to run-off, the Umbria region is achieving the highest values (0.48 g of heavy metals/kg of grapes), followed by Piedmont (0.4386 g of heavy metals/kg of grapes). The region of Apulia seems to achieve the lowest values for all heavy. On the other hand, the highest values for erosion (SM) can be attributed to Umbria (Cr, Cu, Ni), Piedmont (Cd, Pb, Zn), and Marche (Hg). In terms of the heavy metals themselves, Cr reaches the highest values (26.4% of the total), followed by Zn (26.3%), whilst the lowest values refer to Hg and Cd (less than 0.1%).

When it comes to leaching (SM), it is the region of Trentino South Tyrol that reports the highest emissions (0.021 g of heavy metals/kg of grapes), followed by Piedmont (0.011 g of heavy metals/kg of grapes). Furthermore, the lowest values of heavy metals emissions refer to Sicily (Cd, Pb, Cu, Zn), Friuli Venezia Giulia (Hg) and Lombardy (Cr), whilst the highest to Trentino South Tyrol (Cd, Cr, Cu), Marche (Hg), Piedmont (Zn), and Veneto (Pb). In terms of the heavy metals themselves, Cr reaches the highest values (40.3% of the total), followed by Zn (29.3%), whilst the lowest values refer to Hg (less than 0.1%) and Cd (1.2%); Ni emissions are zero due to missing data.

Finally, focusing on the mass balance of the soil, most of the results appear to be negative. It means that the heavy metals introduced on the soil as input (i.e., fertilisers, pesticides) are less than that lost as output through harvesting processes of plants as well as leaching and erosion processes attributed to agriculture activities. Indeed, concerning soil, the region of Apulia reports the highest emissions (−0.0045 g of heavy metals/kg of grapes). Furthermore, it is Piedmont to receive the lowest scores Cd, Pb, Zn), Umbria (Cr; Ni), Marche (Hg) and Trentino South Tyrol (Cu), whilst it is Apulia to receive the highest scores (Cr, Cu, Ni, Zn), Emilia Romagna (Cd, Pb) and Friuli Venezia Giulia (Hg). In terms of the heavy metals themselves, Cr reaches the highest values (28.2% of the total), followed by Zn (26.8%), whilst the lowest values refer to Hg (less than 0.1%) and Cd (−0.3%).

Furthermore, Fig. 9 and SM show the mass of heavy metals emissions per kg of grapes amongst the 20 Italian regions for erosion, leaching and soil, demonstrating that such emissions varied significantly across regions. In particular, the highest variability for erosion and leaching is observed for Hg, in which the standard deviation is *circa* 147% of the mean value (erosion) and 104% (for leaching), whilst for the soil, it is related to Cd reaching *circa* 265%.

3.1.4. Olives

Fig. 10 reports the quantities of heavy metals emitted to rivers, groundwater, and soil, referring to one kg of olives. The heavy metal results refer to 19 Italian regions (Aosta Valley is excluded from the sample due to a lack of statistical data), showing high variability from region to region through the three phenomena.

In particular, concerning the erosion phenomena, the analysis shows that the highest amounts of heavy metals are emitted in Tuscany (0.93 g
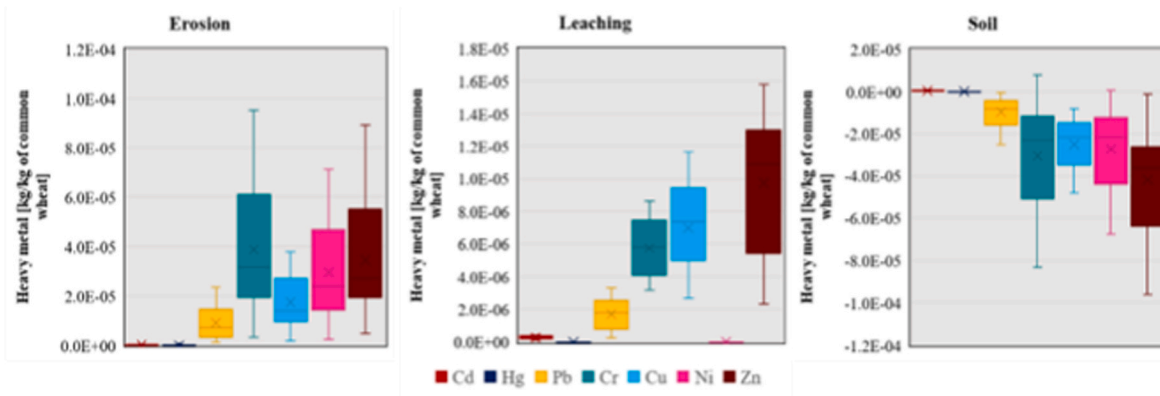

Fig. 7. Box Plot of heavy metals emissions for 1 kg of common wheat among 20 Italian regions.
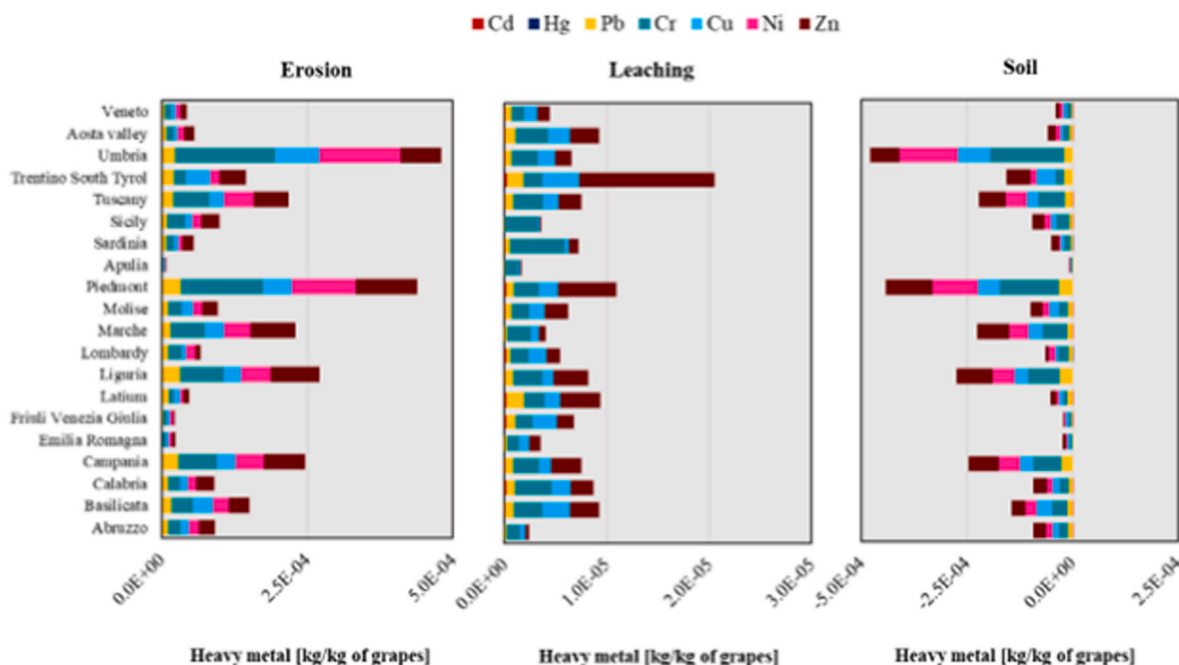
Fig. 8. Contribution analysis of heavy metals emissions for 1 kg of grapes among 20 Italian regions.
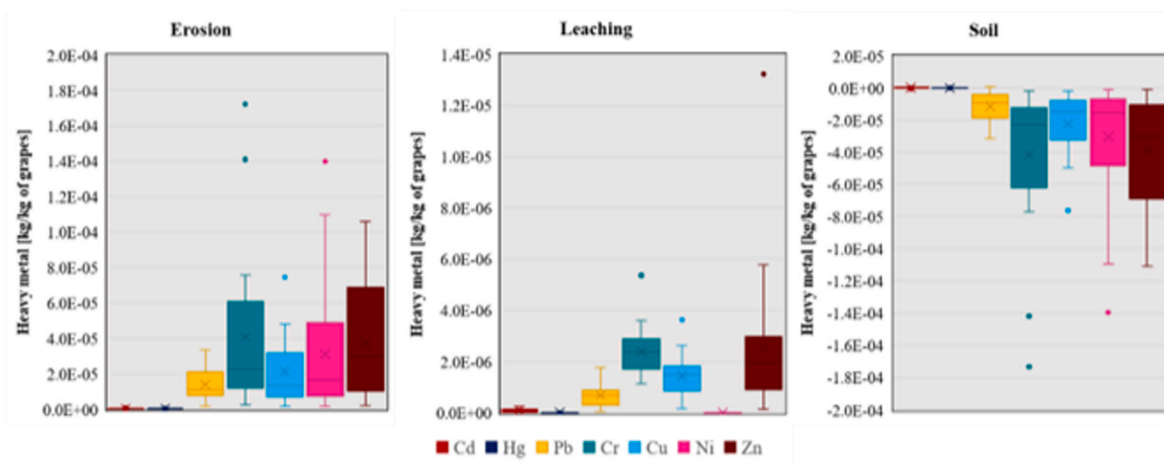

Fig. 9. Box Plot of heavy metals emissions for 1 kg of grapes among 20 Italian regions.

of heavy metals/kg of olives), followed by Calabria and Piedmont (respectively 0.54 and 0.50 g of heavy metals/kg of olives). The region with the lowest overall eroded metals is Liguria, with only 0.004 g of heavy metals/kg of olives. This result is associated with the small amounts of eroded soil affected, in this case, by support practices and cover management factors that are smaller than in other regions. In addition, the relatively low allocation factor (influenced by small yield production and nutrient used per hectare compared to atmospheric deposition) contributes to that result. Instead, for the other regions, the heavy metal run-offs from erosion phenomena are less than 0.45 g (Marche). On average, the largest contribution to erosion phenomena for olives is made by Cr, Zn and Ni, which account respectively for about 31.0%, 27.2% and 20.7% of emissions. While Pb and Cu emissions contribute 11.1% and 9.9%, Cd and Hg account for a significantly lower share (less than 0.1%) of total eroded heavy metals.

Focusing on the leaching phenomena, the highest emissions to the groundwater occur in Piedmont and Trentino South Tyrol, with 0.055 and 0.050 g for a kg of olives, while in the other regions, the heavy metal leached is less than 0.039 g for a kg of olives (Calabria). Considering that

concentrations of heavy metals leached are almost homogeneous among regions for metals, the high variability is strongly affected by the allocation factor, for which the highest values are calculated for Piedmont and Trentino South Tyrol in almost all metals. On average, the largest contribution to leach phenomena for olives is made by Cr and Zn, representing about 46.0% and 30.4% of heavy metals leached in almost all the regions. Accounting for the high variability of results, an exception to these metals exists for Friuli Venezia Giulia, for which Cu contributes 31.9% of its total heavy metals leached.

Focusing on the balance of heavy metals in the soil, results are globally negative in all regions and for almost all metals. However, exceptions are Cd for 3 regions (Apulia, Sicily, and Molise) and Hg for 8. Although their contributions increase the heavy metal globally added to the soil, their low values do not change the globally negative effects generated in regions soil (e.g., the balance of heavy metals for Apulia is −0.02 g/kg of olives). Therefore, the highest heavy metals reduction is generated in Tuscany with −0.96 g/kg of olive, while the other regions are affected by a reduction lower than −0.56 g/kg of olive. In this case, the highest amounts of heavy metal reductions on average are Cr
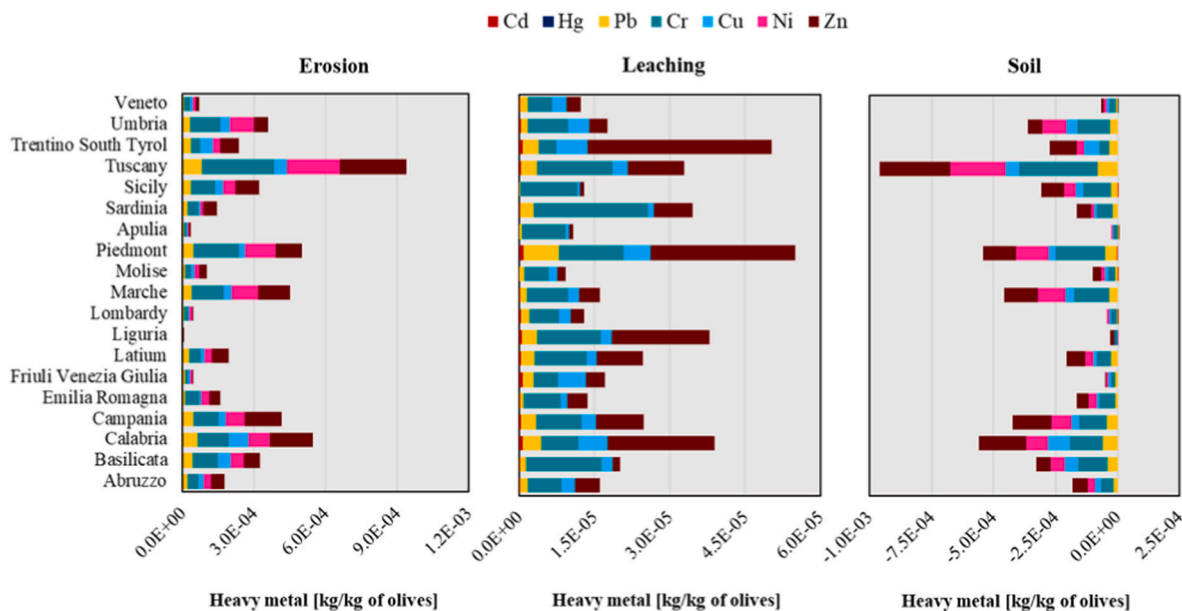
Fig. 10. Contribution analysis of heavy metals emissions for 1 kg of olives among 19 Italian regions.

(36.6%) and Zn (25.3%) since these heavy metals losses for leaching and erosion phenomena are more than that introduced in input with fertilisers and pesticides. To this balance, it also contributes the heavy metals adsorbed by harvested products, representing about 16% of global heavy metals reversed on soil for agricultural activities, ranging from 1 to 181%.

Focusing on the individual heavy metals, Fig. 11 shows the variability of heavy metal emissions per kg of olives among the 19 Italian regions for the three phenomena. As mentioned above, the heavy metal emissions vary significantly across regions for olive groves. In particular, the highest variability is observed for Cd emitted in the soil, in which the standard deviation is about 225.5% of the mean value, followed by Hg (196.8%) emitted in the soil. Indeed, the lowest variability is shown in Cr leached (49.8%). All the other resulting heavy metal emissions varied from 59.2% (Cu leached) to 133.5% (Hg run-offs). Other statistical data for olives are reported in Table 4 (SM).

3.1.5. Oranges

Fig. 12 shows the quantities of heavy metals emitted to rivers, groundwater and soil, referring to one kg of oranges. The data refer to 11 Italian regions with statistically significant oranges production. The three emission compartments are analysed separately (Fig. 12 and SM

Table 5). From the analysis, it emerges that, for those deriving from soil erosion as a result of run-off, the Liguria region has the highest values (0.11 g of heavy metals/kg of oranges), followed by Latium and Campania (respectively 0.0922 g of heavy metals/kg of oranges and 0.0583 g of heavy metals/kg of oranges). The region with the lowest overall emissions is Apulia, with only 0.0209 g of heavy metals/kg of oranges. Based on productivity data extrapolated from official statistical sources, this region has the highest production yields per hectare, contributing to lower emission levels per unit of mass. In general, the largest contribution of heavy metals generated by the orange production process to the river is made by Cr, Zn and Ni, which all account for over 80% of total emissions on average for all regions. Pb and Cu emissions provide minor contributions (together, less than 20%), while Cd and Hg account for a significantly lower share of heavy metals emitted to the river (together, less than 1%).

A different picture is represented when heavy metal leaching is analysed (Fig. 12 and SM). In this case, Latium is the region where the highest emissions occur (0.0050 g of heavy metals/kg of oranges), followed by Liguria (0.0048 g of heavy metals/kg of oranges) and Abruzzo (0.0038 g of heavy metals/kg of oranges). Instead, the lowest emissions are recorded in Basilicata and Apulia (0.0013 and 0.0015 g of heavy metals/kg of oranges, respectively), while in the other regions, total
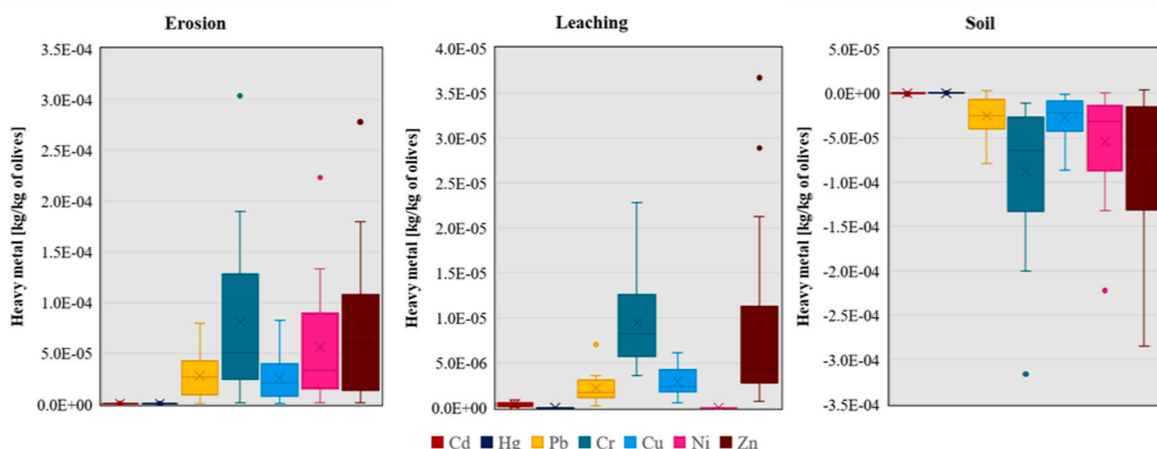

Fig. 11. Box Plot of heavy metals emissions for 1 kg of olives among 19 Italian regions.
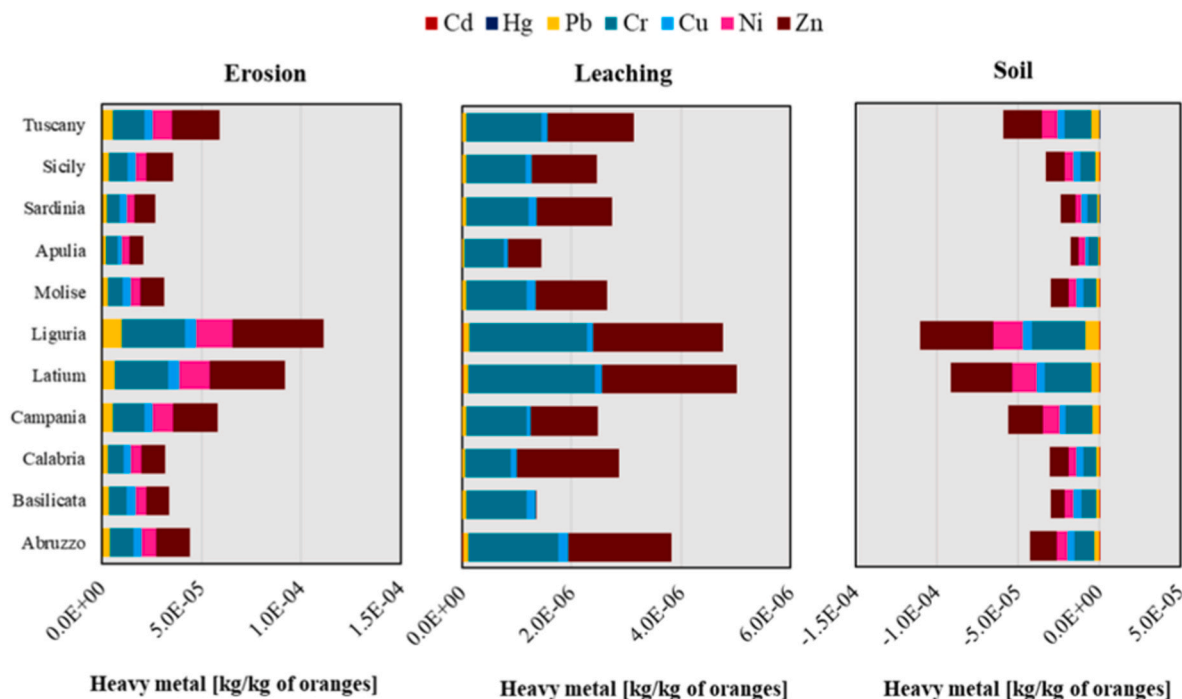
Fig. 12. Contribution analysis of heavy metals emissions for 1 kg of oranges among 11 Italian regions.

emissions are around 0.0020 g of heavy metals/kg of oranges. Cr and Zn are the most emitted metals, reaching more than 92% (together) of the total emissions, while the sum of Pb and Cu account for just 7% of the total. Cd and Hg emissions are almost insignificant.

Focusing on the mass balance of the soil, except for Cd, the results are always negative. It means that the heavy metals introduced on the soil as input (i.e., fertilisers, pesticides) are less than that loss as output through harvesting processes of plants (i.e., oranges collected) as well as leaching and erosion processes attributed to agriculture activities.

In order to assess how regionalised data could affect the heavy metal results, the results of descriptive statistical analysis applied to heavy metals for oranges are reported in Fig. 13 and SM (Table 5), showing the graphical representation of the distribution of data and the tabular statistical data calculated, respectively. The analysis highlights high variability for oranges produced in different regions for the three phenomena and the seven heavy metals. In particular, by calculating the coefficient of variation (standard deviation/mean), it emerges that in the case of erosion emissions, the variability is rather high for all metals

(e.g., 70.8% for Hg, 63.2% for Zn and 63.2% for Cd). Very high variability values are also found in emissions by leaching. In particular, Hg has a variability of 73.6% and Zn of 49.9%. The highest values, however, are found in the ground-level balance of heavy metals, where Hg has a variability of 139.9%, Pb 75.9% and Zn 69.6%.

3.2. Sensitivity analysis

As highlighted above, each crop is characterised by specific cultivation processes, varying from fertilisation processes to tillage and management of soil. Although these data influence heavy metal emissions, showing high variability except in some cases (e.g., Zn and Cr are generally the highest emissions contributions in leaching and erosions phenomena, while Hg and Cd are the lowest), the site-specific data extracted for each region and crop strongly affect their breadth. A sensitivity analysis is carried out to show the influences of orographic systems and the factor variation for crops, varying the SALCA factors for the 5 studied crops and the region's average profiles, accounting for the
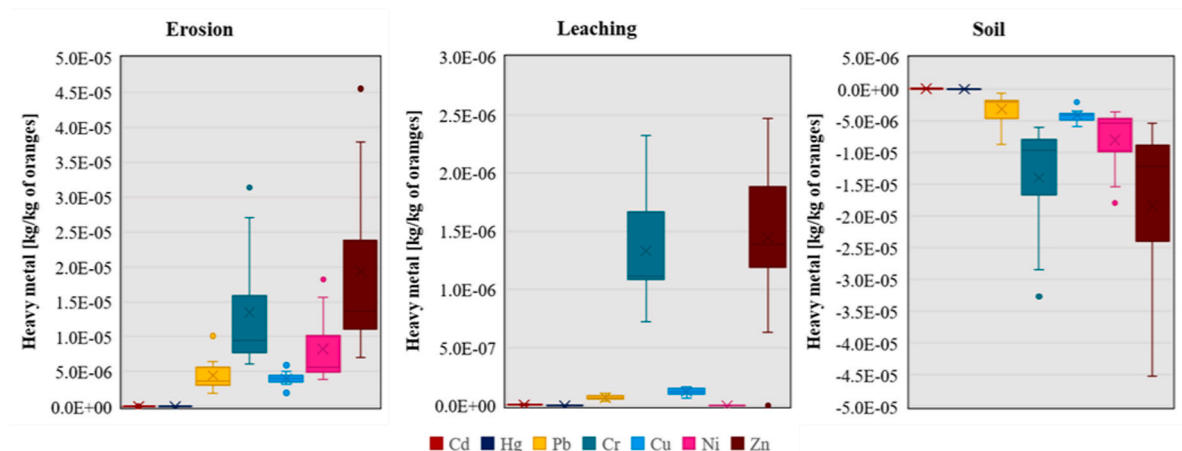

Fig. 13. Box Plot of heavy metals emissions for 1 kg of oranges among 11 Italian regions.

same data in input and output for each region. In particular, a generic agricultural product is considered as output (4000 kg/ha), and generic N fertiliser (120 kg/ha), generic P fertiliser (50 kg/ha), generic K fertiliser (120 kg/ha), and copper pesticides (4 kg/ha) as input. All the concentrations of HM are taken from (Koch and Salou, 2016). The results are reported in Fig. 14 for 1 ha, divided into six scenarios: 1) Durum wheat, 2) Common wheat, 3) Grapes, 4) Olives, 5) Oranges, and 6) Regional average. In this Figure, the minimum value is represented by the first line, whilst the maximum value by the last line. The lower quartile refers to the start of the box, whilst the upper quartile to the end of the box. The median corresponds to the line inside the box. Finally, the external points (outliers) are anormal values of the distribution. Instead, all the results of the descriptive analysis are reported in SM.

As expected, heavy metal emissions vary from scenario to scenario. In particular, the highest mean variabilities are observed for grapes and average regional scenarios for almost heavy metals. While the lowest variabilities are generated in citrus scenarios, for which only one sample point exists in the LUCAS database, generating a small variability in emissions. In addition, for the wheat scenarios, the highest variability of heavy metals is generated for the erosion phenomenon of durum wheat (81.2%). This analysis strongly highlights the importance of site-specific data to obtain more representative inventory data in LCA, increasing data quality, especially for the agri-food sector.

3.3. Challenges and limits

The results provide an opportunity to reflect on the data, generally used for estimating heavy metal emissions in LCI of agri-food products. The proposed approach provides novel insights into the quantification of heavy metals loss for agricultural activities. The estimations of this study allow for considering the limits of data and the usefulness of the existing models. However, the methodological and data gaps and uncertainties are acknowledged. Heavy metal concentration in agri-food products, fertilisers, and pesticides represents one of the main issues. It is widely known that heavy metal concentration data reporting on agri-food products is still limited in the literature.

For this reason, many existing LCA databases refer to Swiss data provided by (Freiermuth, 2006) instead of primary data extracted from laboratory tests. In addition, the continuous updating of the approved inputs list in agriculture (e.g., fertilisers, pesticides etc.) and the simultaneous release of new input on the market do not provide clear and up-to-date information on the content of heavy metals in agricultural inputs. The labelling regulations for such agricultural inputs should be revised to include a requirement to declare the heavy metal content.

In addition, the data reported on SALCA is limited due to a need for geo-referenced data on crop composition and soil properties. Using geo-referenced data for soil properties increases the geographical, temporal, and technological representativeness of agri-foods datasets, especially
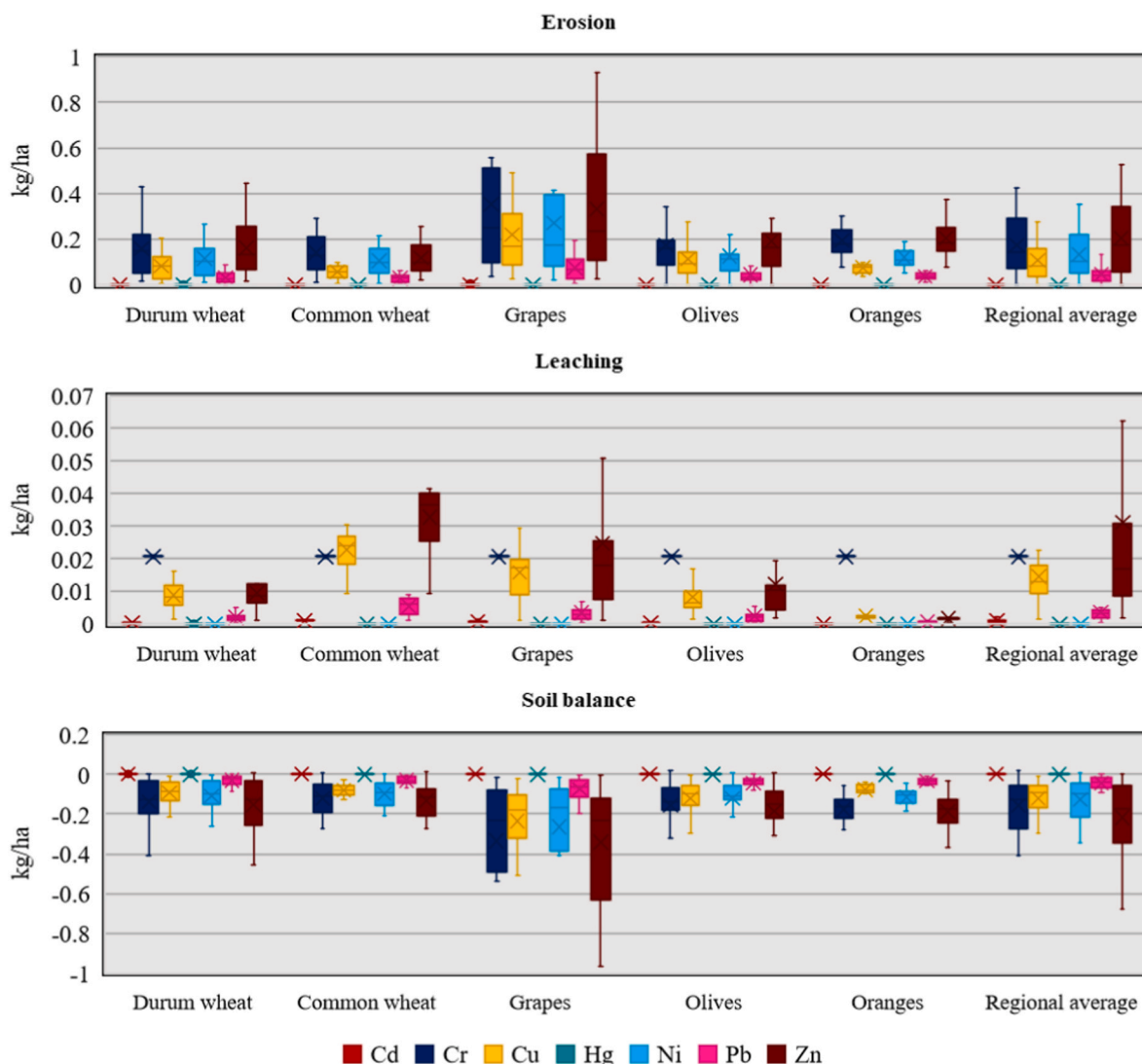

Fig. 14. Box plot of heavy metals emissions for 1 ha for production of generic product among 20 Italian regions and different crops factors.

for Italian case studies. Indeed, applying the SALCA model using specific Italian factors for representing leaching and erosion processes allows for considering Italy's complex orographic systems (Khan and Chiti, 2022) and reducing the uncertainty in LCIs.

In fact, extracting data for specific arable land and permanent crops (using LUCAS soil point) allows selecting average regional data, excluding the effects generated on soil from artificial land or forest. For example, as highlighted by (Panagos et al., 2015b), the C-factor could be totally different from forest to arable lands. The mean C-factor in Italy is estimated to be 0.13, with an extremely high variability; forests have the lowest mean C-factor (0.0013), and vineyards and olive lands are the highest (0.3454 and 0.2163, respectively). Contrarily, the mean LS-factor for Italy is 3.63 ± 4.86; forests have the highest mean LS-factor (6.64), and Arable land and Permanent crops the lowest (0.99 and 1.80, respectively) (Panagos et al., 2015a). Similar trends have also been noticed for the other factors (see (Panagos et al., 2014; Panagos et al., 2020)).

However, as mentioned above and reported in Fig. 3, although LUCAS includes 628 samples in the class cropland for Italy, for some crops (such as orange and citrus) and some Italian regions (such as Aosta valley and Liguria), this database does not provide the sufficient sample of points to provide reliable average site-specific data.

Another limitation relates to the atmospheric deposition, assumed for this approach equal to the heavy metal emissions (Italian, 2021). Unfortunately, as mentioned above, no spatial data distribution was found for Italy for this parameter. The only data available refer to the heavy metal depositions reported by Nicholson et al. (2003) and EMEP (EMEP, 2021). However, the HM atmospheric deposition reported in the first document (Nicholson et al., 2003), although it seems the most coherent dataset regarding parameters and the number of heavy metals evaluated, refers to one rural area in 1997. These data cannot be accounted for in the study's aim since heavy metal emissions have been strongly reduced since 1990 in all the Europe, affecting atmospheric deposition and making the data not representative of the current situation. Indeed, the report proposed by EMEP calculated the atmospheric deposition of heavy metals among Europe countries. The report is limited to three heavy metals (Cd, Pb, and Hg) and provides only data at the national level. However, considering that Italy is characterised by a very diversified emission context at the territorial level (e.g., Lombardy originates the largest shares of emissions of all metals except for arsenic, deriving for the most part from Apulia), also these data could not be accounted representative for each Italian region. In addition, from the data reported in the EMEP report, it should be noted that the emission sources of Italy contribute from 58% to 81% of total depositions, and the other part comes from other countries. Instead, heavy metals emitted from Italy are 1.21–1.47 times that of deposition. Despite this, no homogenous trend was observed among heavy metal depositions and emissions to estimate an eventual scaling factor.

4. Conclusions

The implementation of regionalised data in the SALCA model was aimed at bridging the existing gap regarding the use of heavy metal emission models suitable for representing the diversity of the Italian territory. Following the objectives of the ILCIDAF project, this study aimed to define a generalisable approach for estimating heavy metal emissions using site-specific and regionalised data. The model was then applied by using specific data for the agricultural productions of the four supply chains under study in the ILCIDAF project: the cereal, wine, olive oil and oranges supply chains.

Using site- and crop-specific data in the SALCA model made it possible to assess heavy metal emissions caused by erosion, leaching and heavy metal balance in the soil. However, the most important aspect highlighted by the study was the variability of the data, which represents the prowess of a regionalised approach to characterising the diversity of the Italian territory through the SALCA model.

All in all, even though the sensitivity analysis, where all the factors relating to the cultivation technique were kept constant, the use of site-specific data makes it possible to highlight the influence of the orographic characteristics of the territory. Therefore, the high variability in the results is to be considered the strong point of this regionalised approach, which makes it possible to overcome some limits that the use of national average data would instead accentuate. Undoubtedly, further efforts are needed by the scientific community and public decision-makers to enable greater data availability.

The continuation of the research will aim to enrich the database used for the regionalised SALCA model, in particular by integrating data on most of the crops grown in Italy, in order to make the model useable by the entire national scientific community.

CRediT authorship contribution statement

Bruno Notarnicola: Funding acquisition; Resources; Supervision; Validation; Visualization; Writing - review & editing; Francesco Astuto: Investigation; Methodology; Preliminary data extraction; Visualization; Roles/Writing - original draft; Writing - review & editing; Rosa Di Capua: Visualization; Roles/Writing - original draft; Writing - review & editing; Teresa Maria Gulotta: Methodology; Data extraction; Software; Visualization; Roles/Writing - original draft; Writing - review & editing; Giovanni Mondello: Methodology; Validation; Roles/Writing - original draft; Writing - review & editing; Giuseppe Saija: Funding acquisition; Resources; Supervision; Validation; Visualization; Writing - review & editing; Ioannis Arzoumanidis: Visualization; Roles/Writing - original draft; Writing - review & editing; Manuela D'Eusanio: Visualization; Roles/Writing - original draft; Writing - review & editing; Luigia Petti: Funding acquisition; Resources; Supervision; Validation; Visualization; Writing - review & editing; Giacomo Falcone: Investigation; Methodology; Preliminary data extraction; Visualization; Roles/Writing - original draft; Writing - review & editing; Antonio Fazari: Visualization; Roles/Writing - original draft; Writing - review & editing; Alfio Strano: Funding acquisition; Resources; Supervision; Validation; Visualization; Writing - review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgements

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cesys.2023.100122.

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