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Framework for estimating toxic releases from the application of manure on agricultural soil: National release inventories for heavy metals in 2000-2014

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Abstract

Livestock manure is commonly applied on agricultural land for its fertilising properties. However, the presence of toxic substances in animal manure such as pathogens, antibiotics and heavy metals, can result in damages to ecosystems and human health. To date, although relevant for policy-making, e.g. regulation framing, their releases to agricultural land have been incompletely and inconsistently quantified at global and national scales. Here, we thus developed a generic framework for estimating such releases based on the quantities of manure applied and concentrations of toxic substances. Applying this framework, we built a global release inventory for arsenic, cadmium, chromium, copper, mercury, nickel, lead and zinc differentiated into 215 countries and 15 years (period 2000-2014). Comparisons with more narrowly-focused inventories showed overall consistency in our inventory results, although a number of uncertainties and limitations were identified. In particular, the need for harmonising sampling and analytical methods for estimating heavy metal contents in manure and generating more country-differentiated data, especially for developing countries, should be prioritised by future research studies. Using life cycle impact assessment methods, it was additionally found that mercury, zinc and copper are the substances contributing the most to the toxic impacts on human health and freshwater ecosystems resulting from manure application to land. While countries such as China, India, Russia, Brazil and the United States of America contributed to half the heavy metal releases from manure application worldwide, the impact intensity per area of agricultural land was observed to be highest for island countries, the European Union and South-East Asia because of higher per-area applications of manure. These findings demonstrate the need to perform country-specific impact assessment to support policy-making regulating the concentrations of toxic substances such as heavy metals in utilised manure.

Keywords

Manure management, heavy metal concentration, inventory, toxicity, impact assessment, life cycle assessment

1. Introduction

Reusing animal manure as an organic crop fertiliser has been shown to enable a better use of nutrient sources and to help reduce pollution due to nitrogen leaching compared to utilisation of conventional mineral fertilisers (e.g. EC, 2012; Kramer et al., 2006). In addition, the application of animal manure on agricultural land has been reported to result in high crop productivity and soil fertility over longer time periods than synthetic fertilisers, thus making it a relevant alternative (Hepperly et al., 2009; Jensen, 2013; Redding et al., 2016; Russo and Taylor, 2010). However, animal manure contains traces of antibiotics, heavy metals and pathogens, which may damage ecosystems and human health (Eneji et al., 2003; Kumar et al., 2005, 2013; Millner, 2009; Nicholson et al., 2003). These potentially toxic substances may affect not only the plants through direct uptake (Kornegay et al., 1976; Tien et al., 2016; Zhou et al., 2005), but also the grazing cattle and humans by ingestion of contaminated food and water (Kumar et al., 2013). Manure treatment (e.g. anaerobic digester, chemical addition, thermal treatment) may help reduce the concentrations of antibiotics and pathogens to reach limits defined by authorities, e.g. the European Union (Bicudo and Goyal, 2003; Dolliver et al., 2008; EC, 2011). For many other pollutants (including heavy metals), some countries have implemented national regulations setting concentrations thresholds for composted waste (Cai et al., 2007; DüMV, 2012; MoE, 2006)

Estimating the current inputs of toxic substances to soil and their potential damage on the environment can aid define proper concentration thresholds and prioritise pollutants to address in policy-making. To date, the total releases of harmful substances resulting from the application of manure in a given country have only been quantified in few studies and for specific cases. Inventories of heavy metal releases have been built for the world as a whole in 1988 (Nriagu and Pacyna, 1988), for England and Wales in 2000 (Nicholson et al., 2003), for France in 2012 (Belon et al., 2012) and for China in 2005 (Luo et al., 2009). As an intermediate step to derive a global toxic release inventory (hence not disclosed), Cucurachi et al. (2014) quantified total heavy metal releases from manure application in Europe in 2010, using country-specific manure production statistics combined with average heavy metal contents for Europe. The European inventory was thus used as a basis to extrapolate to the global scale using different proxies, i.e. gross domestic product, carbon dioxide emissions or mercury emissions. Likewise, Sleeswijk et al. (2008) estimated heavy metal releases from manure application in the world in 2000 based on total livestock statistics, excretion rates and heavy metal concentrations reported for the Netherlands (detailed results not available either). Although highly relevant in environmental policy-making context, e.g. to aid frame existing or new regulations, none of these studies address a consistent national and temporal differentiation in their estimates of chemical releases from manure application in a global perspective.

In this study, we therefore aim to (i) develop a harmonised framework for estimating the total releases of toxic substances to agricultural soil resulting from the application of manure in a given country; (ii) apply this framework to derive national inventories of heavy metal releases in the world in the period 2000-2014 ; and (iii) quantify the impact of these releases on ecosystems and human health. The focus on heavy metals was motivated by their data availability and their environmental relevance as they range among the top contributors to damages on human health and freshwater ecosystems (Laurent et al., 2011).

2. Materials and methods

2.1. Release inventory framework

The release of a substance to agricultural soil is directly related to the quantity of applied manure and to the concentration of the substance in the manure. The latter mainly depends on the amount of substance present in the feed ingested by the animals, which is regulated for each livestock (EC, 2003, 2002). Furthermore, the form of the manure, either as a liquid or a solid waste, influences its composition and thus its content in toxic substances (Amlinger et al., 2004).

Following the method used by Cucurachi et al. (2014), we propose a general framework to estimate the input quantity $Q(S)_{l,t,c,y}$ of a substance S to the agricultural soil resulting from the application of manure of type t (solid or liquid) from the livestock l in the country c in year y – see Equation 1:

$$Q(S)_{l,t,c,y} = M_{l,c,y} \times P_{l,t,c,y} \times C(S)_{l,t,c,y} \qquad \text{(Equation 1)}$$

Where $M_{l,c,y}$ is the quantity of manure from the livestock l applied in the country c in year y (in kg N-content); $P_{l,t,c,y}$ is the proportion of manure managed through a solid or liquid system (type t) for the livestock l in the country c in year y (in %); $C(S)_{l,t,c,y}$ is the concentration of substance S in manure of type t from the livestock l in the country c in year y (in g/kg N-content).

2.2. Data collection

2.2.1. Manure applied to soil $M_{c,y,l}$

Quantities of manure applied to soil, expressed as kg of nitrogen content (kg N-content), can be retrieved from the statistic division of the Food and Agriculture Organization of the United Nations (FAOSTAT) for 215 countries across the world from 2000 to 2014 (FAOSTAT, 2015a). The quantity of nitrogen in manure applied to soil is calculated by FAOSTAT following the 2006 Intergovernmental Panel on Climate Change guidelines for estimating nitrous oxide emissions from nitrogen present in the manure added to agricultural soils by farmers (FAOSTAT, 2015a; IPCC, 2006). It is expressed as the amount of nitrogen excreted by livestock, net of the nitrogen losses due to manure management systems, plus the nitrogen contribution from bedding materials when present. The amount of nitrogen excreted is obtained by multiplying the number of livestock heads in a country by typical animal masses and by nitrogen excretion coefficients specific to each livestock. Thus, FAOSTAT (2015a) assumes that the totality of livestock manure is used for crop fertilisation.

The applied manure data set is differentiated into 16 livestock species: buffaloes, cattle (dairy, non-dairy), sheep, goats, swine (market, breeding), chickens (layers, broilers), turkeys, horses, donkeys, mules, camels, ducks, and llamas (FAOSTAT, 2015a). Manure from horses, camels, lamas, mules, donkeys and buffaloes represented only 0.4% of the total manure applied in Europe, and 7.2% in the world (FAOSTAT, 2015a). Because their manure is poorly used and seldom analysed (see Section 3.1), these livestock were considered as cattle (non-dairy) when building the inventory of heavy metal releases. For the same reason, turkey was treated as broiler poultry.

2.2.2. Manure management system $P_{l,t,c,y}$

Livestock excreta can be collected as solid manure (i.e. scraped from the floor with beddings) or liquid slurry (i.e. flushed out of enclosed areas). The choice of the manure management system depends on many factors, including the type of livestock, the size of the farm, the management costs and the environmental and regulatory policies (BIOFerm, 2009; Westerman and Bicudo, 2005). The proportion of cattle (dairy and non-dairy), swine and poultry kept on either solid or liquid manure management systems was compiled for 17 European countries from a questionnaire addressed to experts across the United Nations Economic Commission for Europe (UNECE) in 2003 – see Table B1 (Kuczyński et al., 2005). For a specific livestock, the proportion of animals kept on a liquid manure management system is considered equivalent to the proportion of excreta collected as liquid slurry, and likewise for solid manure management systems and solid manure.

The analysis of the proportion of animals kept on a liquid manure management system (Table B1) shows clear differences between the types of livestock. While most of the swine are kept on liquid manure management system, nearly all layer chickens are kept on solid manure management systems. These proportions also vary considerably between EU countries, for example ranging for non-dairy and dairy cattle from 0-3% in Hungary to 100-100% in the Netherlands (Table B1). For poultry, slurry was assumed to have the same heavy metal content as solid manure as very little data exist on heavy metal concentrations in poultry slurry. This assumption should be acceptable because liquid manure management system is rarely used for poultry (see Table B1). For countries for which no information exist about the type of manure management systems, the geometric means of the available data were used as approximations. This assumption is deemed of little influence on the total releases as the metal concentrations show little variations across solid and liquid manure for a given livestock (see Section 3.1). Further work is required to refine this assumption and extend data of manure management systems to all countries in the world.

2.2.3. Heavy metal content in manure $C(HM)_{l,t,c,y}$

Heavy metals are commonly added to animal feeds for health and welfare reasons, and a large part of the consumed heavy metals may then be excreted and end up in manure (Faridullah et al., 2014; Nicholson et al., 2003). In intense pig farming, the amounts of copper and zinc eliminated through the animal manure can thus correspond to 72-80% and 92-96% of the amount ingested, respectively (Mantovi et al., 2003). Most studies investigating heavy metal contents in manure report concentrations in milligram of heavy metal per kilogram of dry matter (mg/kg.dm) (Amlinger et al., 2004; Møller et al., 2007). To obtain the heavy metal content $C(HM)_{l,t,c,y}$ in g/kg N-content, information about the dry matter content and the nitrogen content in manure are necessary, as shown in Equation 2.

$$C(HM)_{l,t,c,y} = [HM]_{l,t,c,y} \times \%DM_{l,t,c,y} \times \frac{1}{[N]_{l,t,c,y}} \qquad \text{(Equation 2)}$$

Where $[HM]_{l,t,c,y}$ is the concentration of heavy metal *HM* in manure of type *t* from the livestock *l* in the country *c* in year *y* (in g/t.dm); $\%DM_{l,t,c,y}$ is the dry matter content in manure of type *t* from the livestock *l* in the country *c* in year *y* (in %); and $[N]_{l,t,c,y}$ is the nitrogen content in manure of type *t* from the livestock *l* in the country *c* in year *y* (in kg-N/tonne fresh matter).

The organic matter and nitrogen contents in manure strongly depend on its treatment, making difficult any time or country differentiation (Amlinger et al., 2004; Faridullah et al., 2014; Westerman and Bicudo, 2005). Typical dry matter content and nitrogen content on a fresh weight basis in solid manures and slurries from cattle, pig, sheep, duck, layer, broiler and turkey have been retrieved from Chambers et al. (2001). Albeit old, these data are assumed to be valid for the purpose of the current study. Further studies to determine dry matter contents and nitrogen contents in manure are recommended to improve the accuracy of these estimates.

With respect to heavy metal concentrations, the Assessment and Reduction of Heavy Metal Input into Agro-ecosystems (AROMIS) database contains information about the concentrations of cadmium, chromium, copper, nickel, lead and zinc in animal manure. These data were compiled from 32 reports and scientific articles published prior to 2003 for 10 European countries – see Table B2 (KTBL, 2005). In spite of its relative comprehensiveness, the AROMIS database does not document arsenic and mercury concentrations, does not provide any data related to non-European countries and does not include data more recent than 2003. New European regulations concerning heavy metal contents in animal feeding stuffs were implemented in 2002 and 2003 (EC, 2003, 2002), and may have resulted in changes in heavy metal concentrations in manure. The AROMIS database includes only 1 study carried out after these regulations from 2002 – 2003, and could therefore be assumed outdated. To address these gaps, an additional literature review was conducted, using Web of Science database (Thomson Reuters, 2017) and Google Scholar (https://scholar.google.com), with the keywords 'heavy metal', 'content', 'concentration', 'manure'.

2.3. Impact assessment

To evaluate the toxicity-related environmental impacts caused by the releases of heavy metals to agricultural soil, the USEtox 2.02 model was used (Hauschild et al., 2008; Rosenbaum et al., 2008). It is a scientific consensus-based model for characterising impacts of chemicals on human health (termed 'human toxicity', differentiated between cancer and non-cancer effects) and freshwater ecosystems (termed 'freshwater ecotoxicity'), and it is typically used in life cycle impact assessment (Hauschild et al., 2013, 2008; Rosenbaum et al., 2008).

The model enables the calculation of substance-specific characterisation factors (CF), which are used to assess their potential environmental impacts. These CF are the product of three factors, which describe the transport and distribution of the substance in the different environmental compartments (i.e. fate factor), the increase in the amount of substance transferred to living organisms (i.e. exposure factor), and the resulting probability of adverse effects in the organisms (i.e. effect factor) (Rosenbaum et al., 2008). In USEtox 2.02, CF are available for the ionic forms of the eight heavy metals considered in this study, with arsenic being differentiated between arsenic III and V, and chromium being differentiated between chromium III and VI. Chromium is primarily emitted as chromium III and, while chromium VI is highly toxic, it is likely reduced to chromium III in soil (EFSA, 2009). The CF for chromium III was therefore selected for the purpose of this study. As emission distribution of arsenic species remain unclear, the average of the CF for arsenic III and arsenic V was used to characterise arsenic releases (Fantke, 2016).

Characterised impact scores were calculated by combining release data with CF for each heavy metal, thus leading to impact results for human toxicity (cancer and non-cancer), expressed in number of disease cases, and freshwater ecotoxicity, expressed as potentially affected fraction of species integrated over time and volume (PAF.m^3.day). Within an impact category (i.e. human toxicity or freshwater ecotoxicity), the impact scores of all metals can be compared against each other to identify the highest contributor, and they can be aggregated to obtain a total impact score from the application of manure to agricultural soil (only covering impacts from heavy metals). Assuming an equal weighting factor of 1, cancer and non-cancer effects can be aggregated together, thus yielding a single impact indicator result for human toxicity (Rosenbaum et al., 2008).

To facilitate the interpretation of the results across countries, “impact intensities” can be calculated. These are defined as the toxicity impact scores divided by the area of cultivated agricultural land, thus reflecting the magnitude of the toxic impacts stemming from a unit of agricultural land on which manure is assumed to be applied. Cultivated agricultural land was defined as the sum of arable land, permanent crops, and permanent cultivated meadows and pastures (data retrieved from FAOSTAT (2015b)).

3. Results and discussion

The results of the study and their analyses are presented in Sections 3.1-3.4, each addressing a key aspect. The literature review of heavy metal concentrations in manure is addressed in Section 3.1 as it reports an up-to-date attempt at consolidating different data sources to obtain consistent concentration estimates. The resulting inventory of heavy metal releases is documented in Section 3.2 and it is later compared with alternative literature sources for validation purposes in Section 3.3. Finally, Section 3.4 describes the use of the inventory for toxicity impact assessment of manure application (focus on heavy metals).

3.1. Review of heavy metal concentrations in manure

In addition to the AROMIS database (see Table B2), the literature review has led to identifying 17 scientific articles, covering arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn) in solid manure and/or slurry from cattle, swine, goat, sheep and poultry, in European countries (7 studies) and non-European countries (10 studies). The compilation of heavy metal concentrations is available in Table B3. As most available data applies to Europe, heavy metals concentrations reported in European countries are analysed separately from non-European countries in the subsequent sections.

3.1.1. European countries

For a given livestock and heavy metal, ranges of concentrations reported for each individual country in the AROMIS database overlap each other. It means that there appear to be no significant difference in the heavy metal content in manure among European countries (see Table B2). Furthermore, for a given country and heavy metal, concentrations reported in more recent scientific articles fall within the ranges of those in the AROMIS database. These results hence suggest that there is no significant change over time for European countries and that the concentration values from the AROMIS database can be considered valid for the period 2000-2014 (see Tables B2 and B3). Considering the lack of significant time or country differentiation, the high influence of living conditions and measuring methods on the results (see Section 3.1.3) and the low amount of studies available per country, it is therefore assumed that heavy metal concentrations are homogeneous in Europe for the whole period 2000-2014 of the inventory.

For cadmium, chromium, copper, nickel, lead and zinc, the AROMIS project reported the European means of concentrations weighted by the number of samples in each study to correct for potential biases due to individual sampling and analytical errors (KTBL, 2005). Because the articles from the literature review rarely document the number of analysed samples and have concentrations similar to the ones reported in the AROMIS database (see Table B2 and B3), the weighted means were directly used in the inventory. For heavy metals and livestock not included in the AROMIS project (i.e. arsenic and mercury; duck, sheep and goat), geometric means were calculated from the data retrieved for Europe from the literature review (see data in Table B3). The resulting heavy metal concentrations representative for Europe are reported in Table 1.

As illustrated in Table 1, which highlights in bold the highest metal concentrations for each heavy metal, copper and zinc show the highest concentrations in swine manure. Manure from sheep and goat present the highest concentrations of arsenic, chromium, mercury, nickel and lead due to maximal values reported by Amlinger et al. (2004). Heavy metal contents in manure from such livestock have rarely been quantified and additional analytical studies should be conducted to reduce the uncertainty associated with these values. It was noted that for most livestock, heavy metal concentrations in manure do not necessarily respect the thresholds defined by national legislations, e.g. in Denmark (MoE, 2006) and Germany (DüMV, 2012) (data not shown here).

Table 1. Heavy metal contents in manure $C(HM)_{l,t}$ by livestock and manure type retained for European countries

Livestock	Manure type	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
		(in g/kg N-content)							
Dairy cattle	**Solid**	3.46E-2	1.25E-2	3.13E-1	9.58E-1	5.00E-3	1.83E-1	1.58E-1	4.96E+0
	Liquid	1.44E-2	8.00E-3	1.38E-1	8.40E-1	1.80E-3	1.24E-1	1.12E-1	4.14E+0
Non-dairy cattle	**Solid**	3.46E-2	1.25E-2	3.13E-1	9.58E-1	5.00E-3	1.83E-1	1.58E-1	4.96E+0
	Liquid	1.88E-2	1.04E-2	1.80E-1	1.10E+0	2.35E-3	1.62E-1	1.46E-1	5.40E+0
Swine	**Solid**	3.46E-2	1.79E-2	5.00E-1	**8.46E+0**	1.07E-3	2.96E-1	1.29E-1	**3.31E+1**
	Liquid	9.60E-3	3.00E-3	9.40E-2	1.93E+0	6.00E-4	1.20E-1	3.00E-2	9.34E+0
Sheep & goat		**6.75E-2**	1.41E-2	**8.10E-1**	1.36E+0	**7.08E-3**	**4.35E-1**	**4.07E-1**	6.16E+0
Layer chicken		9.56E-3	7.50E-3	1.22E-1	1.16E+0	9.38E-4	1.31E-1	6.00E-2	8.83E+0
Broiler chicken		1.02E-2	8.00E-3	4.00E-1	1.78E+0	1.00E-3	1.24E-1	7.40E-2	7.06E+0
Duck		1.95E-2	**2.25E-2**	2.78E-1	2.84E+0	2.11E-3	3.39E-1	1.97E-1	1.54E+1
Standard deviation		1.70E-2	5.19E-3	1.98E-1	2.09E+0	2.06E-3	1.02E-1	9.68E-2	7.98E+0

3.1.2. Non-European countries

As shown in Table B3, heavy metal contents in manure could be retrieved from 10 publications covering 8 non-European countries (i.e. China, Pakistan, Turkey, Nigeria, Japan, Malaysia, Thailand and Brazil). This underlines the lack of data and the need for further research in this area. To build interim inventories at national and global scales, extrapolations and assumptions are therefore necessary. Concentrations in non-European countries were compared to the average values retrieved for Europe as part of the literature review, and the heavy metal contents were observed to be generally higher for non-European countries than for European countries (values higher than 1 in Figure 1). For copper and zinc, concentrations however remain in the same ranges as in Europe or end up lower than the means calculated in Table 1 (e.g. Adesoye et al. (2014), Faridullah et al. (2014)). Higher heavy metal contents in manure in non-European countries may be due to less restrictive regulations concerning animal feed, or to higher background concentrations in the environment caused by other sources of pollution, e.g. intense traffic and industrial activities (Wang et al., 2013).

Large data gaps exist with respect to non-European country coverage (only 8 represented countries) and large discrepancies are observed in the estimation of the heavy metal contents in manure, with differences of more than one order of magnitude for some metals within a same country, e.g. Pakistan and China for mercury (see Figure 1). These shortcomings, along with the lack of harmonised heavy metal extraction methods (see Section 3.1.3), led to privilege the use of the means established for Europe, as reported in Table 1. These are recommended for use until more consistent sets of data become available, although studies specifically addressing developing countries should carefully check for possible underestimations when using these proxies.

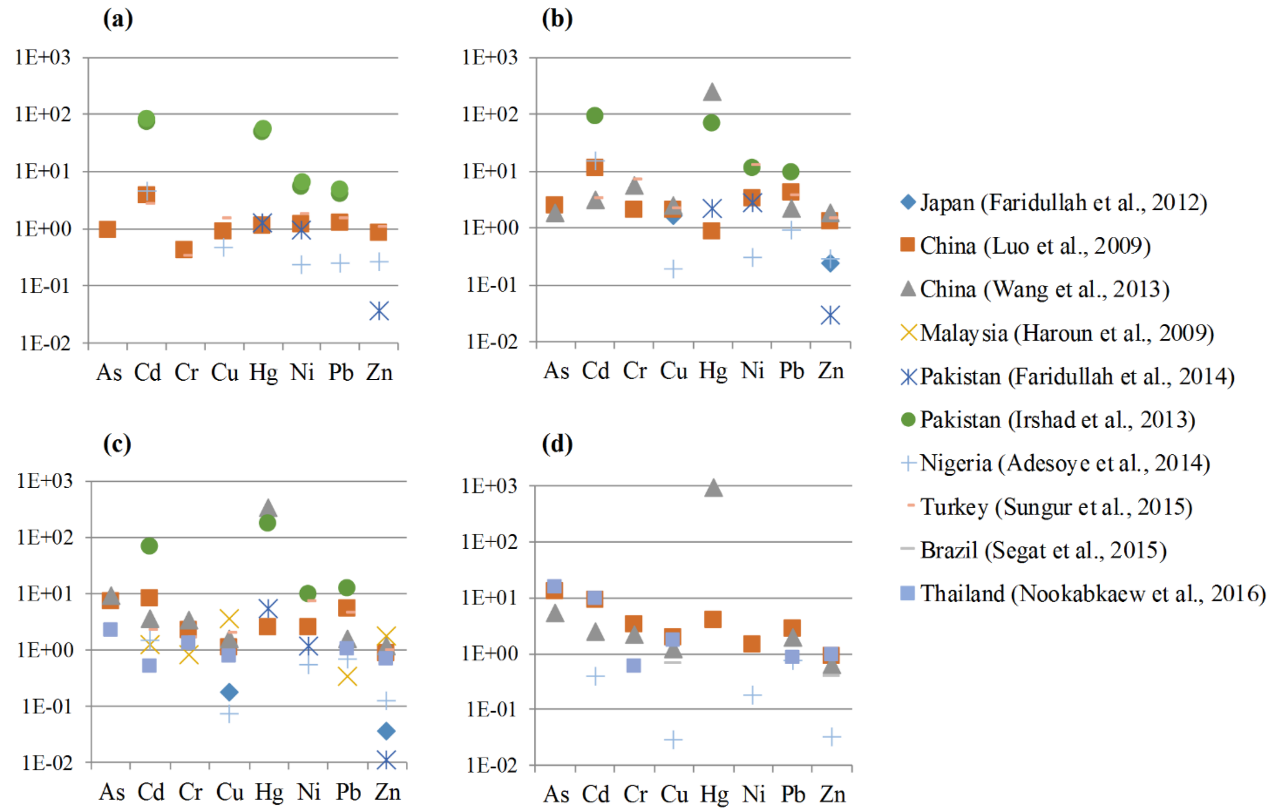

Figure 1. Heavy metal concentrations of manure reported for non-European countries normalised by the average concentrations in Europe for (a) sheep and goat, (b) poultry, (c) cattle, and (d) swine. Average concentrations in Europe are reported in Table 1. Note logarithmic scale on y-axis.

3.1.3. Sampling and analytical methods

Several publications have highlighted the influence of different parameters on the measured heavy metal content in manure (e.g. Eckel et al. (2005), Faridullah et al. (2012)). They have shown that, irrespective of the livestock, the heavy metal contents in animal manure vary in a broad range across studies as it highly depends on the animal diet, age and living conditions (Bolan et al., 2004). Manure treatments such as composting or ashing also influence its organic matter content and heavy metal concentrations (Amlinger et al., 2004; Faridullah et al., 2014; Gul et al., 2015; Hsu and Lo, 2001; Lv et al., 2016).

Regarding the sampling and analytical methods, unrepresentative sampling due to manure inhomogeneity may result in up to 50% error in measures of heavy metal concentrations (Amlinger et al., 2004; Eckel et al., 2005; Gonçalves Júnior et al., 2007). In addition, the metal extraction and determination methods (including type of extractant, pre-treatment of the sample, digestion) differ across and within countries (Amlinger et al., 2004). The choice of the extractant determines which chemical forms of the metals are measured, such as the exchangeable fraction, the organically complexed form, and the residual part (Eneji et al., 2003; Faridullah et al., 2012; Irshad et al., 2013). In an effort to align the studies, the commonly used *aqua-regia* extraction method, which determines the pseudo-total concentration of metals, could be used as a basis for further harmonisation (Amlinger et al., 2004; ISO, 1995). More time-consuming sequential extraction procedures can complement this approach and help characterise each chemical fraction of metals, thus providing more relevant information when it comes to assessing the quantities of heavy metal bioavailable to plants or susceptible to leak into water bodies.

Therefore, the potential variations in heavy metal concentrations reported across countries may reflect the livestock living conditions, but also result from differences in manure treatment or sampling and analytical methods. The paucity of available data confirms that consistent country and time differentiation of heavy metal concentrations in manure is currently not possible. This calls for future studies reporting on the content of contaminants in manure to transparently document the techniques used and strive for harmonisation with the *aqua-regia* extraction method, possibly complemented with sequential extraction procedures. In the current study, provided the limited amount of data available (see Section 3.1.1-3.1.2), no categorisation of the reported heavy metal content according to the extraction techniques used was attempted.

3.2. Inventory results

The framework defined in Equation 1 was applied to develop a global inventory of heavy metal releases (arsenic, cadmium, chromium, copper, mercury, nickel, lead and zinc) differentiated into 215 countries and 15 years, i.e. from 2000 to 2014. The detailed inventories at those scales are available in Appendix A.

3.2.1. Influence of livestock differentiation

For each heavy metal, the substance concentration in manure only varies within one order of magnitude across the different livestock and types of manure (liquid or solid state), with a maximal standard deviation of 7.98 for zinc (see Table 1). The contribution of a livestock to the releases of a heavy metal in a given country is therefore typically driven by its contribution to the quantities of applied manure, as illustrated in Figure 2, where similar livestock distributions were noted for manure application and nickel releases in Belgium in 2012.

At national level, releases of heavy metals with higher standard deviations are typically driven by the contribution of the livestock with the highest concentration in manure. In such cases, the distribution of livestock in manure application may therefore depart significantly from that in specific metal releases. For example, the contribution of swine manure to the total releases of copper in Denmark in 2012 (77%) was higher than its contribution to the quantity of applied manure (59%) since swine manure – liquid or solid – has among the highest copper concentrations (Figure 2).

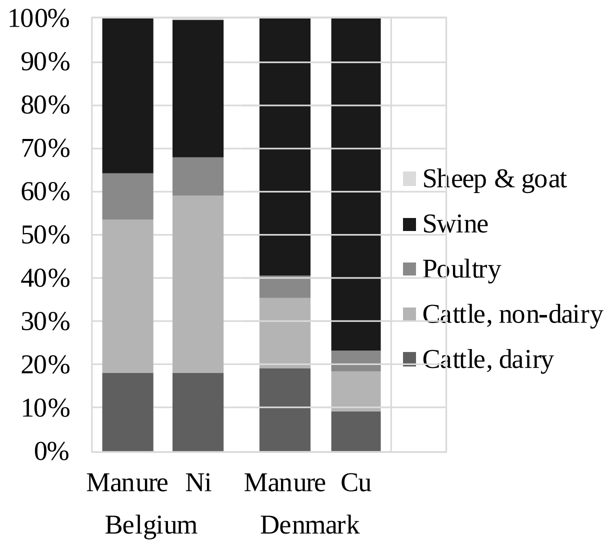

Figure 2. Contribution of livestock to the applied manure and the resulting releases in 2012.

A strong linear correlation between the national releases of a given heavy metal and the total quantity of applied manure is observed across countries, with correlation coefficients r-squared (i.e. r2) ranging from 0.953 to 0.994 (Table 2). The inclusion of country-specific heavy metal concentrations is expected to reduce the quality of this correlation because the heavy metal contents in manure are likely to be higher in non-European countries like China (see Section 3.1.2). However, in the absence of data allowing for country differentiation of metal concentrations in manure, national heavy metal releases can be extrapolated from the total application of manure (i.e. regardless of the livestock distribution) with the metal-specific proxies presented in Table 2.

Table 2. Proxies for extrapolating heavy metal releases from the total application of manure

	Proxy (kg/kg N-content)	**$R2^a$**
As	2.46E-05	0.973
Cd	1.04E-05	0.994
Cr	2.97E-04	0.992
Cu	1.81E-03	0.967
Hg	2.81E-06	0.953
Ni	1.87E-04	0.988
Pb	1.32E-04	0.974
Zn	8.56E-03	0.970

[a]Linear regression on 3225 points

3.2.2. Geographical variations

In 2013, the total quantities of applied manure per country ranged from 8.26E+03 kg N-content (Tokelau) to 5.52E+09 kg N-content (China). As a result, the total per-country releases varied by up to 6 orders of magnitude for a given heavy metal. In 2014, the top 5 countries in the world with respect to heavy metal releases from manure application were China (e.g. 22% of global Ni releases), India (9% for Ni), the United States of America (7% for Ni), Brazil (6% for Ni) and the Russian Federation (4% for Ni). In comparison, the 28 European Union members represented 18% of the global releases of nickel. The actual contributions of countries like China or India are likely to be more important as the heavy metal content in manure is expected to be higher in non-European countries – see Section 3.1.2.

3.2.3. Temporal variations

The quantity of applied manure by livestock is the only time-dependent parameter in the developed inventory. The correlation between heavy metal releases and manure inputs (see Section 3.2.1) therefore entails that, in a given country, heavy metal releases follow a temporal evolution that is very similar to the one of the total quantity of applied manure. Using the country classification defined by the United Nations to differentiate developed economies, economies in transition and developing countries (UN, 2014), distinct trends could be observed for each group of countries –

see the example of zinc in Figure 3. In developing economies (red lines), total releases have generally increased by a factor of 1-2.5 between 2000 and 2014 (exception of Sao Tome and Principe and Rwanda). In contrast, heavy metal releases have generally decreased by a factor ranging from 1 to 2 in developed countries (blue lines). No general trend could be observed for economies in transition (in yellow).

At the global scale, there has been a linear increase of soil-borne releases for all heavy metals between 2000 and 2014, with R-squared values higher than 0.959 (thick black curve in Figure 3). Each year, the releases of heavy metals were thus calculated to increase by 5.57 t/yr (As), 23.9 t/yr (Cd), 80.8 t/yr (Cr), 479 t/yr (Cu), 0.58 t/yr (Hg), 46.6 t/yr (Ni), 31.0 t/yr (Pb) and 2.27 kt/yr (Ni). These values are believed to be sufficiently representative at global scale to be used as basis to develop forecast heavy metal release inventories.

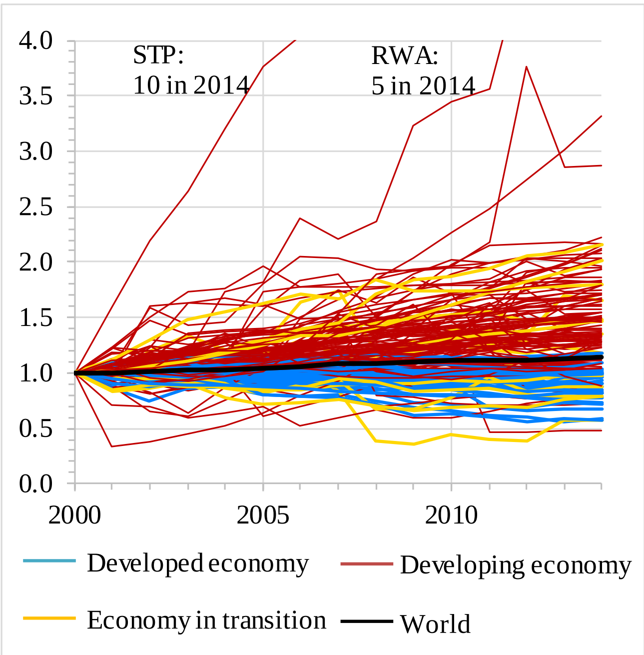

Figure 3. Temporal evolution of zinc releases by country indexed on values in 2000. STP: Sao Tome and Principe, RWA: Rwanda

3.3. Validation of the framework

The globally-differentiated inventory described in Section 3.2 demonstrates the operationalisation of the proposed framework although several uncertainties and limitations can be noted given the current level of data availability. Taking the different terms of Equation 2, the quantity of applied manure was found to be the most influential parameter to the determination of heavy metal releases (see Section 3.2.1) and may be overestimated since it does not consider other utilisations of manure than for fertilising purposes (FAOSTAT, 2015a). Although it was evaluated to be less influential, the proportion of animals kept on liquid or solid manure management system was averaged for all non-European countries based on European data, which may not be representative. Finally, the discrepancies observed in the measuring and analytical methods used to determine heavy metal concentrations and the lack of sufficient number of country-specific data on the heavy metal, organic matter and nitrogen contents in manure led to calculating concentration means representative for all years and countries. As indicated in Section 3.1.2, these can be questioned as concentrations are very likely underestimated for developing countries due to lower regulations. Further studies addressing these shortcomings and providing harmonised and country-specific data are therefore recommended to improve the overall quality of the inventory.

To test the precision of the inventory results, comparisons were performed with previous studies having disclosed national or regional inventories of heavy metals, viz. releases resulting from manure application in England and Wales, in France, in China and in the whole world – see Table 3.

Nicholson et al. (2003) estimated the heavy metal releases in England and Wales resulting from manure application based on livestock numbers, excreta production quantities, and average heavy metal concentrations in manure. Heavy metal releases calculated in our study for the United Kingdom are found to be 1.1 to 7.1 times higher than in Nicholson et al. (2003) – see Table 3. The discrepancies may be explained by the different geographical scope, i.e. Scotland and Northern Ireland being included in our study and not in Nicholson et al. (2003), and by the generally lower manure concentrations in Nicholson et al. (2003). For example, the chromium concentrations used by Nicholson et al. (2003) are ca. 50% smaller than the European geometric means calculated in the AROMIS project (Table B2).

While Belon et al. (2012) generally used the same methodology as Nicholson et al. (2003), the study also accounted for the type of effluents and the time spent by animals inside and outside the farm buildings. The authors estimated heavy metal releases, which are 1.4 to 3.2 times higher than the values from our inventory (see Table 3). The concentration data used by Belon et al. (2012) could only be retrieved for swine, and are 1.1 to 1.9 times higher than those used in our inventory for this livestock. The absence of further documentation precluded any further analysis of the discrepancies, including the possible influence of the time spent inside and outside the farm on the heavy metal concentration of manure.

When calculating the heavy metal inputs to agricultural soil due to the application of manure in China, Luo et al. (2009) applied the same method than Nicholson et al. (2003). However, Luo et al. (2009) used concentrations higher than those applied in our inventory (see Figure 1), thus resulting in systematically higher release estimates. As discussed in Section 3.1.2, there is a likelihood that estimates for non-European countries are underestimated in our study, thus calling for developing approaches to derive more consistent release inventories for these countries.

At global scale, Nriagu and Pacyna (1988) built an inventory based on the combination of annual global discharge to soil of animal waste and manure with ranges of heavy metal concentrations. They disregarded any differentiation of livestock. Heavy metal releases calculated in our inventory are observed to fall within the ranges found by Nriagu and Pacyna (1988) for cadmium, copper, mercury and zinc, while they are up to two times lower than the minimum of the ranges for arsenic, nickel and chromium (Table 3). The higher releases obtained by Nriagu and Pacyna (1988) are likely explained by the use of concentrations ranges higher than those in our study and the lack of livestock differentiation.

Table 3. Comparison of heavy metal inventory results with retrieved literature sources.

Region	Year	Reference	Unit	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
United Kingdom	2000	This study	t	18	6.5	203	909	2.1	119	97	4200
England & Wales	2000	Nicholson et al., 2003	t	16	4.2	36	643	0.3	53	48	1858

Ratio				1.1	1.6	5.6	1.4	7.1	2.2	2.0	2.2
France	2012	This study	t	24	10	256	1199	3	156	120	5840
France	2012	Belon et al., 2012	t	78	13	371	2578	6	368	307	11795
Ratio				0.3	0.7	0.7	0.5	0.5	0.4	0.4	0.5
China	2005	This study	t	127	54	1522	10057	14	1011	689	47812
China	2005	Luo et al., 2009	t	1412	778	6113	49229	23	2643	2594	95668
Ratio				0.1	0.1	0.2	0.2	0.6	0.4	0.3	0.5
World	1988[a]	This study	kt	0.5	0.2	6.1	35.1	0.1	3.8	2.8	163.9
World	1988	Nriagu and Pacyna, 1988	kt	1.2-4.4	0.2-1.2	10-60	14-80	0-0.2	3-36	3.2-20	150-320
Ratio[b]				0.2	0.3	0.2	0.7	0.6	0.2	0.2	0.7

[a] Backcasting using linear extrapolations from our inventory and the mean increase rates estimated for each metal in Section 3.2.3.

[b] Based on the median of the range calculated by Nriagu and Pacyna (1988)

In light of these comparisons, the developed inventory therefore appears consistent with the available literature since our estimations of heavy metal releases remain in the same order of magnitude as those reported in other individual studies (except for arsenic and cadmium in China; see Table 3). The four comparisons additionally reflect the importance of country-specific heavy metal concentrations in addition to the quantities of applied manure.

3.4. Impact assessment

3.4.1. Heavy metal contribution

As illustrated in Figure 4, mercury (52%), zinc (87%) and copper (94%) are the most contributing substances for human toxicity (cancer effects), human toxicity (non-cancer effects) and freshwater ecotoxicity impacts, respectively. These metals should therefore be addressed in priority through regulations limiting their concentrations in feedstuff and/or manure applied to land.

These results however are associated with some uncertainties because they are based on averaged quantities of heavy metals, the chemical forms of which are not specified. The use of sequential extraction procedures could help mitigate those uncertainties and estimate heavy metal release inventories which are consistent with the life cycle impact assessment method (see Section 3.1.3).

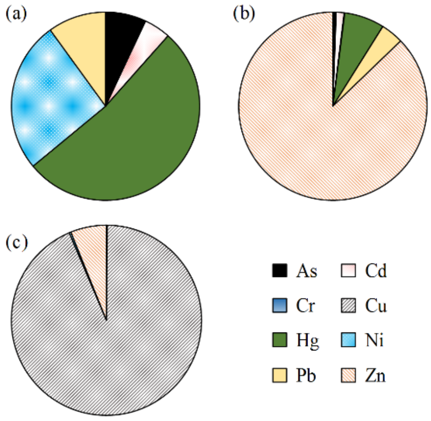

Figure 4. Heavy metal contribution to (a) human toxicity (cancer effects), (b) human toxicity (non-cancer effects) and (c) freshwater ecotoxicity resulting from manure application in the world in 2013.

3.4.2. Impact intensity

Impact intensities for human toxicity and freshwater ecotoxicity are linearly correlated with the quantity of manure applied per area of agricultural land, with r-squared values of 0.993 and 0.994, respectively. As observed in Figure 5 for human toxicity, European countries and South-East Asian countries are regions with large impact intensities due to intensive manure application per agricultural area. The geometric mean of the human toxicity impact intensities reached 5.11E-01 cases/km^2 in the European Union in 2013 while it was equal to 1.45E-01 cases/ km^2 for the world. Although they were among the top countries in terms of absolute human toxicity impact score (see Appendix A), India, Russia, Brazil and the United States had human toxicity impact intensities close to the global mean, with values ranging between 9.21E-02 and 1.72E-01 cases/km^2 in 2013. Because of the aforementioned linear correlation, a similar pattern is observed for freshwater ecotoxicity impact intensities. Studies aiming at refining data such as heavy metal concentrations and quantities of applied manure should therefore focus on these regions to enable the development of more accurate assessments and help implement consistent regulation frameworks aiming at reducing these impacts.

Albeit not visible in Figure 5, many of the top-ranking countries with respect to their toxicity impact intensities were observed to be small islands and territories (15 out of the top 20). In particular, Singapore ranked first with a human toxicity impact intensity of 8.50E+01 cases/km^2 due to large inputs of manure in comparison to the available area of agricultural land. The second country (Saint Kitts and Nevis) obtained an impact intensity 10 times lower, i.e. 8.27E+01 cases/km^2, which still was 57 times higher than the global mean. Such outlying results are interpreted as misreporting of manure application data. FAOSTAT indeed reports that 43% of the total manure applied in Singapore in 2013 originated from swine (FAOSTAT, 2015a) while pig-farming was phased out by the government in the mid-80s (Chien-Fang and Savage, 2015). In addition, the potential imports of manure and the usage of manure for other purposes than crop fertilisation (e.g. heating) are not included in the data reported by FAOSTAT (2015a). Results should therefore be used with caution in countries where applying manure to agricultural soil is not the main practice (see also Section 3.3).

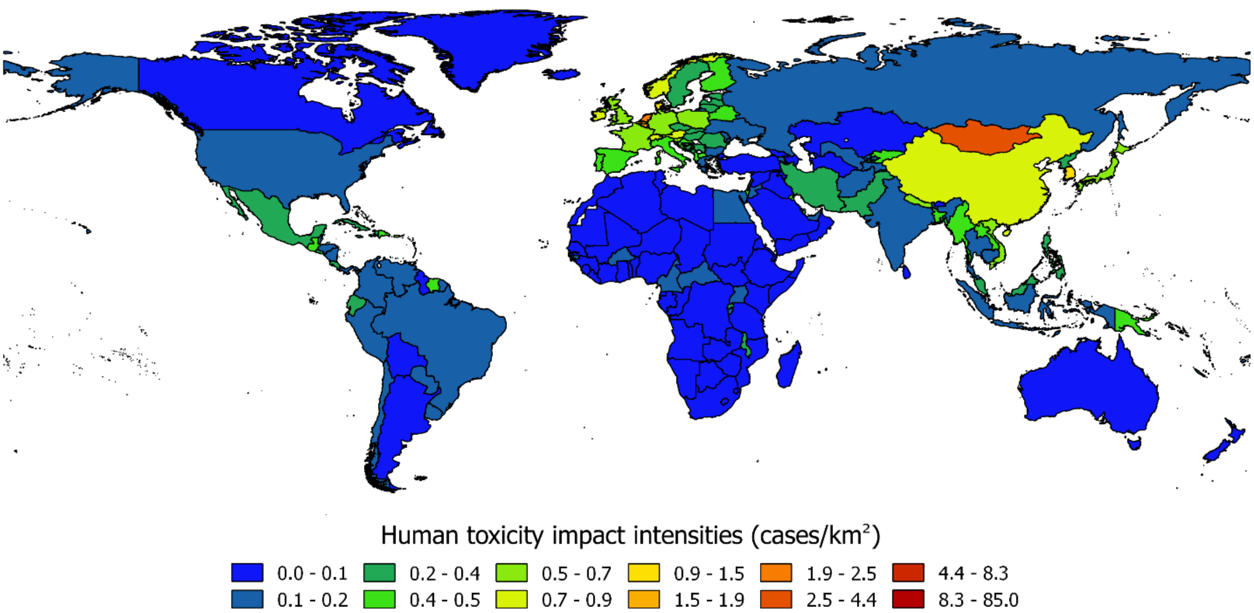

Figure 5. Map of human toxicity impact intensities (aggregated cancer and non-cancer effects) in 2013, created with QGIS 2.18.0 (QGIS Development Team, 2016).

4. Conclusions and recommendations

A framework was developed for estimating soil-borne releases of potentially toxic substances from the application of manure on agricultural land. When applied to eight heavy metals typically present in manure, it allowed calculations of national inventories for 215 countries over 2000-2014. Although the results showed good consistency with previous inventories performed for single countries or the entire world, several points were identified as requiring further research. In particular, additional studies are required to get country-specific and harmonised data on heavy metal contents in manure.

The characterisation of impacts on human health and freshwater ecosystems resulting from manure application evidenced the contribution of mercury, copper and zinc, as well as important impact intensities in Europe and South-East Asia. Policy-making addressing manure management should therefore target these specific metals and regions for framing regulations on heavy metal contents in feedstuff and manure. In a broader perspective, these findings and recommendations also demonstrate the need and relevance of such country- and time-differentiated global inventory of toxic releases that can be used for example with life cycle impact assessment to support effective policy-making.

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Appendix A. Supplementary Results (electronic file)

Contains inventories of the releases of arsenic, cadmium, chromium, copper, mercury, nickel, lead and zinc to agricultural soil resulting from the application of manure for 215 countries between 2000 and 2014. Inventory results by country, year and livestock, and aggregated totals by country and year are both available. Impact scores for human toxicity (aggregated cancer and non-cancer effects) and freshwater ecotoxicity by country in 2013 are also presented.

Appendix B. Supplementary Materials (electronic file)

Table B1 displays the proportion of cattle (dairy and non-dairy), swine and layer chicken kept on a liquid manure management system in 17 European countries.

Table B2 is a compilation of heavy metal concentrations extracted from the AROMIS project, indicating calculated ranges and geometric means of data for each metal, country and livestock.

Table B3 presents the heavy metal concentrations retrieved from the literature review.

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