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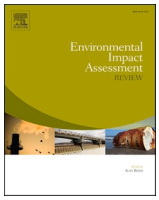

Application of North European characterisation factors, population density and distance-to-coast grid data for refreshing the Swedish human toxicity and ecotoxicity footprint analysis

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ABSTRACT

Here, we develop further the national chemical footprint assessment methods using Sweden as an example to enhance the precision of calculations. First, we integrate grid data on population density and distance-to-seacoast into the analytical framework to better match the European Pollutant Release and Transfer Register on the sub-compartment level with USEtox toxicity characterisation factors. Second, we use the latest USEtox 2.12 model version and its more punctual North European characterisation factors. Third, we conduct trend and geographic analysis and rank Swedish facilities in terms of toxicity potential. We show that total human toxicity potential in Sweden was smaller than previously estimated when using the North European USEtox landscape settings and sloped downwards over time. We confirm toxicity potential of major pollutants in previous research papers (Zn, Hg, Pb, Ni) and find that Hg's relative human toxicity potential in a longer period can be larger than previously estimated on shorter periods. Human toxicity is estimated to be mostly non-cancer type in Sweden. Results are largely invariant to the choice of air sub-compartments. Companies in the metals manufacturing sector are estimated to have the largest human toxicity potential in Sweden in the period between 2001 and 2017 and companies in the paper manufacturing industry have the largest ecotoxicity potential.

1. Introduction

The Protocol on Pollutant Release and Transfer Registers (PRTRs) have been binding its international parties since 2009, UNECE (2012). They have been established to offer a solid framework for enhancing public access to information on pollutant releases and for pursuing international cooperation in environmental impact assessment. PRTRs were constructed to keep track of environmental emissions across geographic regions and times, and are increasingly suggested as a fundamental data source for chemical footprint analysis, Sörme et al. (2016), DeVito et al. (2015), Leclerc et al. (2019).

Arvidsson et al. (2016) have shown that only a limited number of studies (< 5) have attempted to calculate indicators of national chemical footprints using the USEtox consensus model for toxicity impact assessment in life cycle assessment (LCA). According to our literature review this number is still below 10. Furthermore, there is no other study to our knowledge that has attempted to calculate indicators of national chemical footprints using the USEtox consensus model for toxicity impact assessment on the sub-compartment level of emissions for Sweden.

By using data from the European Pollutant Release and Transfer Register (*E*-PRTR) on emissions to air and water from Swedish point sources, and characterisation factors (CFs) from the USEtox 1.01 and 2.01 model, Sörme et al. (2016), Nordborg et al. (2017) suggested the aggregated impact potentials for human toxicity and ecotoxicity as key metrics to measure chemical footprints on the national level. Their first calculations showed that zinc contributed most to the impact potentials both for human toxicity (68%), and ecotoxicity (63%) in Sweden, in 2008, Sörme et al. (2016) The authors' literature review also revealed that ecotoxicity impacts of zinc on freshwater is well established, but there is no straightforward evidence yet on its adverse impacts on humans. For example, the World Health Organization (WHO) does not include zinc in its list of chemicals of major public health concern, but mentions, among others, cadmium, benzene, arsenic, lead and mercury, WHO (2010). Fosmire (1990) found manifestations of overt toxicity symptoms (nausea, vomiting, epigastric pain, lethargy, and fatigue) will

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occur with extremely high zinc intakes. Sanstead (1995) showed that risk of copper deficiency is increased when the molar ratio of zinc to copper (Zn:Cu) is high. Wallig and Keenan (2013) referred to the examination of chronically Zn-intoxicated chickens that showed feather pigmentation loss, feed refusal, weight loss, possibly associated with defective pancreatic acinar cell function. Decreased mineralization in bones have been reported in several species, particularly swelling of the epiphyseal region of the long bones in horses, leading to lameness. These effects are considered to be associated with Zn-induced Cu deficiency, rather than Zn itself, due to a metallothionein and copper interaction, leading to hypochromic anemia.

Bjørn et al. (2014) also identified zinc as a priority substance. They estimated the dilution needed to avoid ecosystem damage, based on CFs from USEtox 1.01. They found that zinc and copper were the substances with the largest contribution to ecotoxicity impact potential in Europe in 2004 (70%, and 30% respectively). Sala and Goralczyk (2013) also took advantage of the USEtox 1.01 characterisation factors and showed that zinc was also a major contributor, together with some other metals and pesticides in Europe.

Taylor et al. (2020) used the Canadian National Pollutant Release Inventory (NPRI) to calculate ecotoxicity potential with the USEtox model in Nova Scotia for 2015. The authors found that the highest priority chemicals identified using the NPRI's quantity-based approach differed markedly from those identified using the toxicity-based approach. Toxicity-based analysis showed that copper (51.06%) composed the largest share of ecotoxicity potential for freshwater, followed by manganese (29.13%), vanadium (9.08%), zinc (4.59%), and cadmium (1.86%). Aluminum had the highest ecotoxicity potential for releases to both rural and urban air.

Persson et al. (2019) studied emissions from Swedish consumption and found that the potential impact on human health of emissions of hazardous chemicals was highest in Sweden, followed by China, Germany and Russia. Hence, increased precision of Swedish national chemical footprint analysis conveys globally important messages. Ecotoxicity potential was calculated to be the highest in the same country group as the human toxicity, although Germany has replaced China as the second largest after Sweden, and Denmark was in third place.

The president of the current European Commission declared in her political manifesto that*'Europe needs to move towards a zero-pollution ambition' and she 'will put forward a cross-cutting strategy to protect citizens' health from environmental degradation and pollution, addressing air and water quality, hazardous chemicals, industrial emissions, pesticides and endocrine disrupters.'*

The *E*-PRTR data can help measure and monitor progress both towards specific Sustainable Development Goals (SDG) and multitudes of them (OECD, 2017; OECD, 2019). First and foremost, the identification of SDG Target 12.4 on environmentally sound management of chemicals as the most closely aligned target with PRTR data. Also, targeting SDG6 ('Ensure availability and sustainable management of water and sanitation for all') or Goal 3 ('Ensure healthy lives and promote well-being for all at all ages') are all influenced by pollutants released into water, air and soil, which are registered in the E-PRTR. Target 6.3 ("By 2030, improve water quality by reducing pollution, eliminating dumping, and minimizing release of hazardous chemicals and materials, halving the proportion of untreated waste water and substantially increasing recycling and safe reuse globally') is another specific example of relevant SDG targets. E-PRTR data can also support measure progress towards Target 3.9 ('By 2030, substantially reduce the number of deaths and illnesses from hazardous chemicals and air, water, and soil pollution and contamination.'). Although the E-PRTR does not register deaths or illnesses resulting from hazardous chemicals, pollution and contamination, it can provide plenty of indicators to measure the related risk exposure by matching emission quantity and toxicity information on the substance level.

The key innovation in our research is that we develop a novel methodology to analyse industrial pollution at the subcompartment level. In particular, to increase the precision of earlier calculations we integrate population grid and distance-to-sea-coast data into the analytical framework. Hence, we better connect pollution data with the USEtox model structure at the sub-compartment level. We draw distinctions between rural air and urban air sub-compartments, and between freshwater and seawater, while Sörme et al. (2016) and Nordborg et al. (2017) used default USEtox characterisation factors in their papers, and assumed that all water emissions were freshwater emissions, and all air emissions were rural air emissions. We also differentiated between cancer and non-cancer characterisation factors to calculate which of the two drives human toxicity impact potentials.

The ultimate goal of this study is to support the development of a more precise national chemical footprint assessment, as this is the cornerstone of sustainable environmental management. Therefore, we somewhat retailored the methodology of Sörme et al. (2016) to better match the *E*-PRTR and USEtox model and increase the precision of national chemical footprint calculation results. The advantage of our approach is that the proposed new methodology can be further extended to other EU Member States in the *E*-PRTR database. Furthermore, our paper assesses the trends and geographic spread of human toxicity and ecotoxicity, which was investigated in earlier papers for shorter periods. Such information is important from a chemical management perspective, and could be used by decision makers for regulatory purposes.

2. Materials and methods

We applied the same general method described in Sörme et al. (2016), Nordborg et al. (2017) and calculated national impact potentials for human toxicity and ecotoxicity associated with emissions from point sources in Sweden. The emissions (E) of substances (i) in the E-PRTR have been multiplied by their USEtox 2.12 characterisation factors (CFs) and aggregated across all substances and release media (j), Eq. (1) as suggested in the USEtox Manual, Fantke et al. (2015). Taylor et al. (2020) stressed the importance of the differentiation of sub-compartments. Hence, to further increase the precision of earlier calculations by Nordborg et al. (2017), we draw distinction between different subtypes (k) of compartments (e.g. release media into which the pollutants are released). In case of air emissions urban air and rural air were differentiated in accordance with the USEtox 2.12 grouping of sub-compartments. In the same vein, freshwater and seawater emissions were also differentiated. In their recent papers Sörme et al. (2016) and Nordborg et al. (2017) assumed that (i) all water emissions were freshwater emissions and (ii) all air emissions were rural air emissions.

$$Impact\ Potential = \sum_{ijk} E_{ijk} \times CF_{ijk} \quad (1)$$

We used the GISCO population database from EUROSTAT (2020) to draw distinction between point sources of emission in urban and rural areas. Definitions of urban areas are often different across countries and are based mostly on administrative or legal concepts of cities that do not necessarily reflect the functional forms of cities, Dijkstra et al. (2019). For the distinction, areas where population density was below 150 inhabitants per km^2 was classified as rural in our study, and above 150 inhabitants per km^2 as urban. In the harmonised European definition of cities and rural areas Dijkstra and Poelman (2014) outlines other criteria (e.g. total population) to capture true urban areas, but in our case population density is the key factor, as this measures exactly human exposure to the toxicity at the emission source points.

To differentiate between seawater and freshwater emissions we identified seawater emissions as those pollutant releases into water in the *E*-PRTR database, where the distance of emission point sources was less than 500 m from the seacoast. The data on'Distance to the Nearest Coast' was also retrieved from the EUROSTATS GISCO database EUROSTAT (2020). Coordinates have been rounded to the 1st decimal (roughly 10 km precision) to match pollutant release data with geographic information on population density and distance-to-seacoast.

Impact potentials are measured in Comparative Toxic Units for human health (CTUh) and ecotoxicity (CTUe), respectively. It should be noted that CTUh and CTUe values can not be directly compared, as they are measured on different scales and in different units.

CFs were obtained from the USEtox 2.12 model, downloaded from the USEtox website (www.usetox.org). Here, both recommended and indicative CFs were used in the calculations, but similarly to Sörme et al. (2016), Nordborg et al. (2017) we note that indicative CFs are associated with considerable uncertainties, primarily related to input uncertainties, Rosenbaum et al. (2008). USEtox is a model based on scientific consensus providing midpoint and endpoint characterisation factors for human toxicological and freshwater ecotoxicological impacts of chemical emissions in life cycle assessment, developed under the auspices of the United Nations Environment Program (UNEP) and the Society for Environmental Toxicology and Chemistry, (SETAC) Life Cycle Initiative. The updated USEtox 2.12 model version was released in 2019. This is the latest corrective release version of USEtox. Important new features of the latest USEtox model versions include human exposure to pesticide residues in crops; an indoor air compartment for human exposure through inhalation, and improved fate and effect modeling of metals.

For comparability, we followed the pioneering works of Sörme et al. (2016) and Nordborg et al. (2017) in many ways. We also used 2008 as a basis year, and analysed pollutant releases to air and water from Swedish point sources, as reported to the *E*-PRTR and as published on the website of the European Environmental Agency in March 2020. Substances covered the same substances as in Nordborg et al. (2017) some of which were not characterized in Sörme et al. (2016) due to lack of CFs in USEtox 1.01. Also, the highest CFs, e.g. the CFs of the most toxic types were applied whenever the E-PRTR does not provide information on the chemical types of a compound. This assumption is relevant for Cr and As. Also, AOX (Halogenated Organic Compounds) were assumed to be represented by 1,4 di-chlorobenzene, NMVOC by Benzene and PAH (Polyaromatic Hydrocarbons) by Benzo-(a)pyrene. These were chosen as a conservative approach, because they have high CFs and are representative for the group.

3. Results

Tables 1 and 2 present results of earlier studies by Sörme et al. (2016), Nordborg et al. (2017) and updated calculations of this paper with the following modified assumptions.

1. The latest, 2.12 version of the USEtox model was also used, beyond 2.01 and 1.01 (from the third column of Tables 1 and 2).
2. We fine-tuned the results by introducing sub-compartments

Table 1
The substances with largest contribution to human toxicity (CTUh), emitted from Swedish point sources to air and water in 2008 and on average in the period 2001–2017, characterized with USEtox 1.01 and 2.01 and 2.12. Only the largest contributions are shown, consisting of the ten substances with largest impact potentials in USEtox 1.01 and 2.01, respectively. Assumptions made in characterization are given in the table footnotes, and follow Sörme et al. (2016).

Authors	Sörme et al. (2016)	Nordborg et al. (2017)	This study (2021)	This study (2021)	This study (2021)	This study (2021)
USEtox - region	na	na	default	default	North Europe	North Europe
USEtox version	1.01	2.01	2.12	2.12	2.12	2.12
USEtox - Subcompartments	R.A. F.W.	R.A. F.W.	R.A. F.W.	R.A. U.A. F.W. S.W.	R.A. U.A. F.W. S.W.	R.A. U.A. F.W. S.W.
Year of observations	2008	2008	2008	2008	2008	2001-2017
PollutantName			**Impact potential (CTUh)**			
Zinc and compounds (as Zn) [a]	4.90E+02	1.60E+02	1.48E+02	1.47E+02	1.37E+01	1.09E+01
Mercury and compounds (as Hg) [b]	9.20E+01	1.40E+02	8.91E+01	8.91E+01	7.92E+00	1.46E+01
Lead and compounds (as Pb) [c]	2.80E+01	4.70E+01	4.85E+01	4.85E+01	4.21E+00	2.69E+00
Chromium and compounds (as Cr) [d]	5.40E+01	4.60E+01	1.76E+01	1.61E+01	4.12E-01	1.35E+00
Arsenic and compounds (as As) [e]	4.80E+01	4.50E+01	4.09E+01	3.96E+01	1.40E+00	1.31E+00
Polycyclic aromatic hydrocarbons (PAHs) [f]	2.60E-01	3.20E+01	3.83E+00	3.83E+00	5.84E-02	3.33E-02
Cadmium and compounds (as Cd) [g]	3.10E+00	6.90E+00	5.67E+00	5.60E+00	4.31E-01	4.09E-01
Non-methane volatile organic compounds (NMVOC) [h]	3.60E+00	2.50E+00	2.20E+00	3.39E+00	1.03E+00	1.14E+00
Nickel and compounds (as Ni) [i]	4.10E-01	1.20E+00	9.96E-01	9.80E-01	4.51E-02	4.58E-02
Fluoranthene	3.20E-02	7.60E-01	6.77E-04	6.77E-04	3.36E-06	1.91E-06
Halogenated organic compounds (as AOX) [j]	2.40E-01	2.10E-01	2.26E-02	1.90E-02	1.47E-03	1.94E-03
Total	**7.20E+02**	**4.82E+02**	**3.57E+02**	**3.54E+02**	**2.92E+01**	**3.25E+01**
			Impact potential (in % of the total)			
Zinc and compounds (as Zn) [a]	68.08%	33.20%	41.42%	41.45%	46.84%	33.54%
Mercury and compounds (as Hg) [b]	12.78%	29.05%	24.97%	25.16%	27.10%	44.81%
Lead and compounds (as Pb) [c]	3.89%	9.75%	13.58%	13.69%	14.39%	8.26%
Chromium and compounds (as Cr) [d]	7.50%	9.55%	4.93%	4.54%	1.41%	4.15%
Arsenic and compounds (as As) [e]	6.67%	9.34%	11.47%	11.20%	4.78%	4.04%
Polycyclic aromatic hydrocarbons (PAHs) [f]	0.04%	6.64%	1.07%	1.08%	0.20%	0.10%
Cadmium and compounds (as Cd) [g]	0.43%	1.43%	1.59%	1.58%	1.47%	1.26%
Non-methane volatile organic compounds (NMVOC) [h]	0.50%	0.52%	0.62%	0.96%	3.52%	3.50%
Nickel and compounds (as Ni) [i]	0.06%	0.25%	0.28%	0.28%	0.15%	0.14%
Fluoranthene	0.00%	0.16%	0.00%	0.00%	0.00%	0.00%
Halogenated organic compounds (as AOX) [j]	0.03%	0.04%	0.01%	0.01%	0.01%	0.01%

Notes: R.A. = Rural Air, U.A. = Urban Air, F.W.= Fresh Water, S.W.= Sea Water, [a] As Zn(II). [b] As Hg(II). [c] As Pb(II). [d] As Cr(VI).[e] As As(V). [f] As benzo[a]pyrene. [g] As Cd (II). [h] As benzene. [i] As Ni(II). [j] As 1,4-dichlorobenzene. The last coloumn of the table aggregates toxicity for years of reporting. Under the European Pollutant Emission Register (EPER) data was reported every three year, first in 2001 and later in 2004. After 2007 data has been reported every year.

(rural/urban AIR and fresh/sea WATER) (from the fourth column of Tables 1 and 2).

3. The North European CFs in USEtox 2.12 were used instead of the default CFs (from the fifth column of Tables 1 and 2).

4. The average impact potential has been calculated for the period between 2001 and 2017 (in the last column of Tables 1 and 2).

3.1. Human toxicity

Human toxicity impacts decreased by 26%, from 480 CTUh calculated by Nordborg et al. (2017), based on characterisation with USEtox 2.01, to 360 CTUh, based on our characterisation with USEtox 2.12 (Table 1). The calculations corroborate the earlier findings that Zinc had the highest impact potential in Sweden in 2008. Also, USEtox 2.12 attributes a smaller potential impact to zinc (41%), and a larger potential impact to mercury (25%), compared to USEtox 1.01.

To increase the precision of the results we draw distinctions between sub-compartments (freshwater/seawater and urban/rural air). The distinctions did not change the results substantially, neither in terms of absolute values of human toxicity impact potential nor in terms of pollutant rankings. One should note, however, that the parameter influencing the distinction between seawater and freshwater could be further fine-tuned. As a conservative technical rule, we classified pollutant releases into seawater, where the facilities were closer to the seacoast than 500 m. Also, CFs of major contributing pollutants to human toxicity are roughly equal for rural and urban emissions. Hence, the results with sub compartments are not significantly different from those without sub-compartments.

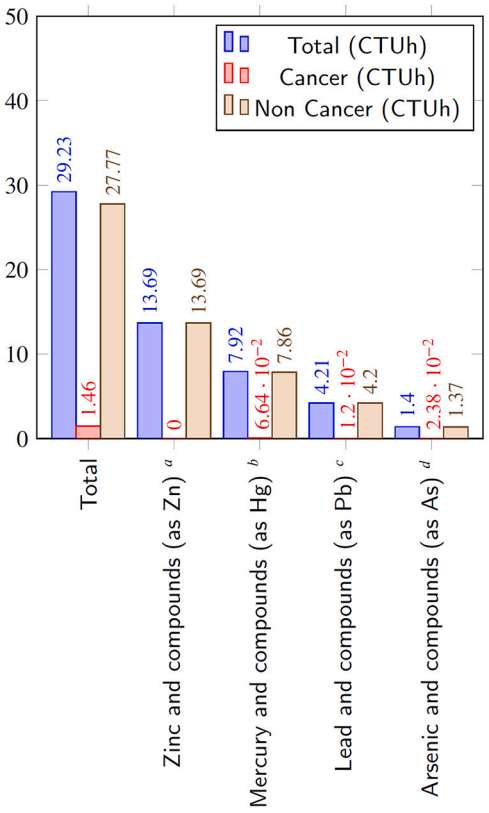
The USEtox 2.12 model allows users to choose regional emission settings, Fantke et al. (2015). As Sörme et al. (2016) and Nordborg et al. (2017) based their calculations to our best knowledge on the default USEtox model results, we tested whether and how regional settings influence results. For regionalized calculations, specific values for landscape parameters can be set by USEtox users, or region specific landscape settings can be applied by selecting one of the 25 sets of landscape parameters. For the recalculus, we used the 'Northern Europe and Northern Canada' landscape setting. Due to the compressed CFs for'Northern Europe Northern Canada' compared to 'default' values, the recalculated human toxicity collapsed to its tenth, to 29 CTUh from 360 CTUh.

Finally, we aggregated human toxicity across years from 2001 to 2017. We found that Mercury's importance in this longer period has increased by 18 percentage points to 45% compared to 27% calculated for 2008. The human risk exposure to mercury has been already elevated due to earlier pollution according to a recent river monitoring exercise in Sweden. The exercise indicated that more than 23,000 water bodies are still affected by the mercury pollution, European Environmental Agency (2018). Fish in thousands of rivers and lakes have mercury levels

Table 2
The substances with largest contribution to ecotoxicity (CTUe), emitted from Swedish point sources to air and water in 2008 and on average in the period 2001–2017, characterized with USEtox 1.01 and 2.01 and 2.12. Only the most important substances are shown. Assumptions made in characterization are given in the table footnotes, and follow Sörme et al. (2016).

Authors	Sörme et al. (2016)	Nordborg et al. (2017)	This study (2021)	This study (2021)	This study (2021)	This study (2021)
USEtox - region	na	na	default	default	North Europe	North Europe
USEtox version	1.01	2.01	2.12	2.12	2.12	2.12
USEtox - Subcompartments	R.A. F.W.	R.A. F.W.	R.A. F.W.	R.A. U.A. F.W. S.W.	R.A. U.A. F.W. S.W.	R.A. U.A. F.W. S.W.
Year of observations	2008	2008	2008	2008	2008	2001-2017
PollutantName			**Impact potential (CTUe)**			
Copper and compounds (as Cu) [a]	6.40E+08	1.10E+11	5.47E+08	5.33E+08	5.81E+09	5.16E+09
Zinc and compounds (as Zn) [b]	4.00E+09	1.40E+10	1.29E+10	1.24E+10	1.33E+11	1.40E+11
Nickel and compounds (as Ni) [c]	1.20E+08	2.30E+09	1.22E+09	1.19E+09	1.27E+10	1.14E+10
Cadmium and compounds (as Cd) [d]	5.30E+06	1.20E+09	9.12E+08	8.82E+08	9.54E+09	1.01E+10
Fluoranthene	7.90E+08	9.40E+08	8.44E+05	8.44E+05	4.57E+06	2.59E+06
Chromium and compounds (as Cr) [e]	6.60E+06	4.90E+08	1.86E+08	1.69E+08	1.79E+09	2.98E+09
Halogenated organic compounds (as AOX) [f]	4.60E+08	4.60E+08	2.12E+08	1.74E+08	7.60E+08	1.05E+09
Arsenic and compounds (as As) [g]	6.00E+07	6.40E+07	5.87E+07	5.64E+07	6.03E+08	4.73E+08
Polycyclic aromatic hydrocarbons (PAHs) [h]	3.40E+07	4.90E+07	6.12E+06	6.12E+06	3.72E+07	1.38E+07
Mercury and compounds (as Hg) [i]	2.70E+06	2.60E+06	2.00E+06	1.91E+06	1.92E+07	1.87E+07
Lead and compounds (as Pb) [j]	1.60E+06	2.80E+06	2.30E+07	2.23E+07	2.27E+08	1.56E+08
Total	**6.29E+09**	**1.30E+11**	**1.61E+10**	**1.55E+10**	**1.64E+11**	**1.71E+11**
			Impact potential (in % of the total)			
Copper and compounds (as Cu) [a]	10.18%	84.81%	3.40%	3.45%	3.53%	3.01%
Zinc and compounds (as Zn) [b]	63.61%	10.79%	80.27%	80.30%	80.82%	81.73%
Nickel and compounds (as Ni) [c]	1.91%	1.77%	7.58%	7.71%	7.73%	6.65%
Cadmium and compounds (as Cd) [d]	0.08%	0.93%	5.67%	5.71%	5.81%	5.87%
Fluoranthene	12.56%	0.72%	0.01%	0.01%	0.00%	0.00%
Chromium and compounds (as Cr) [e]	0.10%	0.38%	1.15%	1.09%	1.09%	1.74%
Halogenated organic compounds (as AOX) [f]	7.31%	0.35%	1.32%	1.12%	0.46%	0.61%
Arsenic and compounds (as As) [g]	0.95%	0.05%	0.37%	0.36%	0.37%	0.28%
Polycyclic aromatic hydrocarbons (PAHs) [h]	0.54%	0.04%	0.04%	0.04%	0.02%	0.01%
Mercury and compounds (as Hg) [i]	0.04%	0.00%	0.01%	0.01%	0.01%	0.01%
Lead and compounds (as Pb) *j*	0.03%	0.00%	0.14%	0.14%	0.14%	0.09%

Notes: R.A. = Rural Air, U.A. = Urban Air, F.W.= Fresh Water, S.W.= Sea Water, [a] As Cu(II). [b] As Zn(II).[c] As Ni(II). [d] As Cd(II). [e] As Cr(VI). [f] As 1,4-dichlorobenzene. [g] As As(V). [h] As benzo[a]pyrene. [i] As Hg(II). [j] As Pb(II). The last coloumn of the table aggregates toxicity for years of reporting. Under the European Pollutant Emission Register (EPER) data was reported every three year, first in 2001 and later in 2004. After 2007 data has been reported every year.



Notes: [a] As Zn(II). [b] As Hg(II). [c] As Pb(II). [d] As As(V).

Fig. 1. Contribution of substances to cancer, non-cancer and total human toxicity (CTUh), emitted from Swedish point sources to air and water (2008), characterized with USEtox 2.12. Only the total contribution and the four substances with largest contributions are shown.

that necessitate issuing health advisory guidelines for fishermen and consumers. Hence, further releases of mercury can increase the human health risk exposure.

We followed Sörme et al. (2016), who assumed equal weighting between cancer and non-cancer cases due to a lack of more precise insights into the issue. When the results were decomposed, they showed that non-cancer human toxicity dominated the aggregated human toxicity impact potentials. Non-cancer human toxicity impact potential was 28 CTUh of the total 29 CTUh in 2008 calculated with USEtox 2.12, and cancer related CTUh was only a fraction of the total, 1 CTUh, (Fig. 1).

3.2. Ecotoxicity

Ecotoxicity impacts have been decreasing, from 1.3E+11 CTUe, based on characterisation with USEtox 2.01, to 1.6E+10 CTUe, based on characterisation with USEtox 2.12. The main reason for this change is that the ecotoxicity impact potential associated with copper dropped by 3 orders of magnitude in the latest model version. It should be noted that this change between the USEtox 2.12 and 2.01 versions reversed a similar magnitude of increase between USEtox 2.01 and 1.01. Hence, the baseline results are closer to Sörme et al. (2016) than to Nordborg et al. (2017). Copper is extremely toxic to aquatic life in high concentrations, causing mortality, reduced growth, and reproduction (United States Environmental Protection Agency, 2014). Sanstead (1995) showed that risk of copper deficiency is increased when the molar ratio of zinc to copper (Zn:Cu) is high. For comparison, copper had the highest ecotoxicity potential for releases to fresh-water, but accounted for less than 1% of quantity-based releases in Nova Scotia, Taylor et al. (2020).

The latest 2.12 USEtox model version identifies metals (Ni, Cd and Cu) as a priority group of substances from an ecotoxicological perspective similarly to its precedent 2.01 and 1.01 model versions.

One can obtain roughly equivalent calculation results, no matter whether sub-compartments of air (rural and urban) and water (sea- and freshwater), into which pollutants are released, are differentiated or not. The reason for this is that ecotoxicity CFs of pollutants with the largest potential are roughly similar for the urban and rural air emissions. Furthermore, only a small fraction of emissions into water are classified as seawater emissions due to our conservative parameter for the classification (emissions into water are classified as seawater emissions when the facility distance to seacost is less than 500 m).

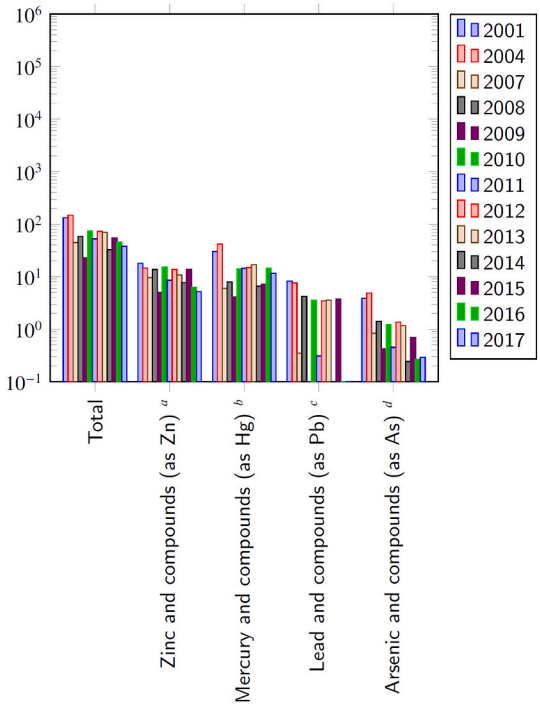
3.3. Trend of toxicity in Sweden

Following the suggestion from Nordborg et al. (2017), we broadened the analysis and monitored the toxic impacts from (Swedish) economic activities over time, in the period between 2001 and 2017. The first reporting year under the European Pollutant Release and Transfer Register (*E*-PRTR) was 2007. The E-PRTR succeded the European Pollutant Emission Register (EPER) under which data was reported every three year, first in 2001 and later in 2004. The EPER data is part of the E-PRTR dataset published by the European Environmental Agency, and hence used in our analysis.

Figs. 2 and 3 show downward sloping total human toxicity and ecotoxicity impacts over time. The downward trend of human toxicity is more obvious for zinc and arsenic than for mercury or lead.

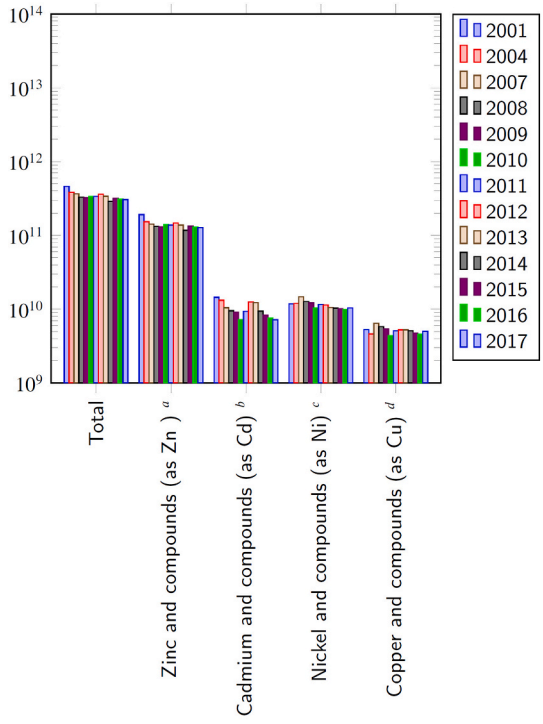
Ecotoxicity impacts have been also decreasing in total, in particular in early 2000s and less dynamically recently. Especially Cd's 5% annual average decrease can be categorized as significant. Koh et al. (2016) reported a decreasing ecotoxicity trend for the US between 1999 and 2013 using the Toxic Release Inventory (TRI) of the US.

Time trends could be seen as a proxy of data quality because



Notes: [a] As Zn(II). [b] As Hg(II). [c] As Pb(II). [d] As As(V).

Fig. 2. Trend of contribution of substances to human toxicity (CTUh), emitted from Swedish point sources to air and water (2001–2017), characterized with USEtox 2.12. Only the total contribution and the four substances with largest contributions are shown.



Notes: [a] As Zn(II). [b] As Cd(II). [c] As Ni(II). [d] As Cu(II).

Fig. 3. Trend of contribution of substances to ecotoxicity (CTUe), emitted from Swedish point sources to air and water (2001–2017), characterized with USEtox 2.12. Only the total contribution and the four substances with largest contributions are shown.

substantial volatility across years could indicate data errors if there was no significant change in production or technology, Sörme et al. (2016). The tangents of the curves were more-or-less stable suggesting stability of data quality except for fluoranthene discussed in the next section.

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3.4. Toxicity map of Sweden

Figs. 4 and 5 present the Swedish human toxicity and ecotoxicity maps. Potential toxicity impacts the south-western coast and north-eastern coast of Sweden. Areas around Gothenburg, Stockholm, Borlange can be identified as densely populated areas with higher human toxicity risk from industrial emission.

It should be remarked, however, that Persson et al. (2019) show that the real impact of Swedish consumption in terms of use and emissions of hazardous substances mostly has an impact outside the Swedish borders, mainly in other EU countries and only 10–24% of the pressure occurs within the Swedish national borders.

3.5. Toxicity ranking of facilities

Sörme et al. (2016) aggregated toxicity information at the level of industrial sectors and at the national level. Here, we provide summary statistics at the facility level. Understanding and measuring environmental impacts of individual companies and facilities is currently needed for environmental accounting, for informed consumer decisions and to scale up financing to clean production and sustainable finance. Jia et al. (1996) suggested that toxicity transformation of PRTRs data could be used as a more effective market-oriented tool. This statement likely holds in case of the Swedish *E*-PRTR. Furthermore, it could be used as part of the Environmental Social and Governance (ESG) assessment of firms by ESG rating companies, which is increasingly an integral element of sustainable finance. To foster further progress in the field, our dataset of the top 300 companies with the largest contribution to toxicity are published as a Mendeley dataset (doi: 10.17632/vbj4x2ngky.1)

Companies in the manufacturing sector of metals are estimated to have the largest human toxicity potential in Sweden in the period between 2001 and 2017 (Table 3). Rönnskärsverken's (Boliden Mineral AB) human toxicity was calculated to be the highest 123 CTUh and impacts both water and air subcompartments.

LKAB - Kirunagruvan' human toxicity impact has been estimated 18 CTUh. LKAB's mining activities give rise to an environmental impact on the surrounding landscape and community, in the form of emissions into the air and discharges into water, noise, vibrations and land impact, LKAB (2021). LKAB's atmospheric emissions are the byproducts of the ore processing plants including mainly carbon dioxide, nitrogen oxides, dust and acid gases, such as sulphur oxide, hydrogen fluoride and hydrogen chloride.

Outokump, another heavyweight manufacturer in the steel industry, is ranked third in terms of human toxicity impact potential (18 CTUh). In its stainless steel production, the largest environmental impacts include dust emissions into the air, water discharges from production, use of direct and indirect energy, and waste created in the production process.

At the industry level the paper manufacturing sector had the largest eco-toxicity footprint in the 2001–2017 sample period (Table 4). This result confirms the findings of Sörme et al. (2016) with USEtox 1.01, even after controlling for the sub-compartment level differences of characterisation factors. The largest zinc emitters have been identified in the paper and paper product industry. Gruvöns bruk is ranked on the top in terms of ecotoxicity impact potential, followed by Stora Enso Kvarnsveden AB and Korsnäsverken.

4. Discussions

The limitations of the national chemical footprint analysis based on the *E*-PRTR should be clearly communicated. Persson et al. (2019) demonstrated the shortage of information by comparing the number of chemical products included in the data from the Swedish System of Environmental and Economic Accounting (close to 100,000) and the number of chemicals (substance groups) included in the *E*-PRTR (less than 100). Also, the E-PRTR and its substance list is currently under revision. It is expected that the list of substances required to be reported to the E-PRTR will be extended, which will necessarily influence any chemical assessments at the national level. Furthermore, the *E*-PRTR database contains pollutant release information above thresholds set for each substance, however industries with releases below the threshold are not included. Data quality could be mentioned as well, as emissions are self-reported by facilities and often estimated and not exactly measured. One should not forget about the possible conflicts of interest embedded in chemical related reporting, which may influence data quality. Leclerc et al. (2019) argued that the E-PRTR database includes neither quantitative nor qualitative uncertainty estimates at substance level, and it appears incomplete in terms of source coverage, there may be gaps and inconsistencies in reporting across countries.

Our baseline result with the USEtox 2.12 version seems to be significantly different for fluoranthene compared to the result of Nordborg et al. (2017) and Sörme et al. (2016) with USEtox 1.01 and USEtox 2.01, respectively. Notably, CFs have not changed between the two model versions. When we compared our pollutant quantity data (total of 7.4 kg of fluoranthene in 2008 in Sweden) downloaded from the EEA E-PRTR database to the figure of 8600 kg of fluoranthene in the study by Sörme et al. (2016) the difference in toxicity results could be better reconciled. After cross-checking our E-PRTR dataset with the dataset published on the Swedish Environmental Protection Agency website, we found that either the reported fluoranthene data by Kubikenborg Aluminum AB for 2008 is not correct in the Swedish PRTR, or it is missing from the European equivalent, E-PRTR database. Sörme et al. (2016) also mentioned that in the Swedish database fluoranthene

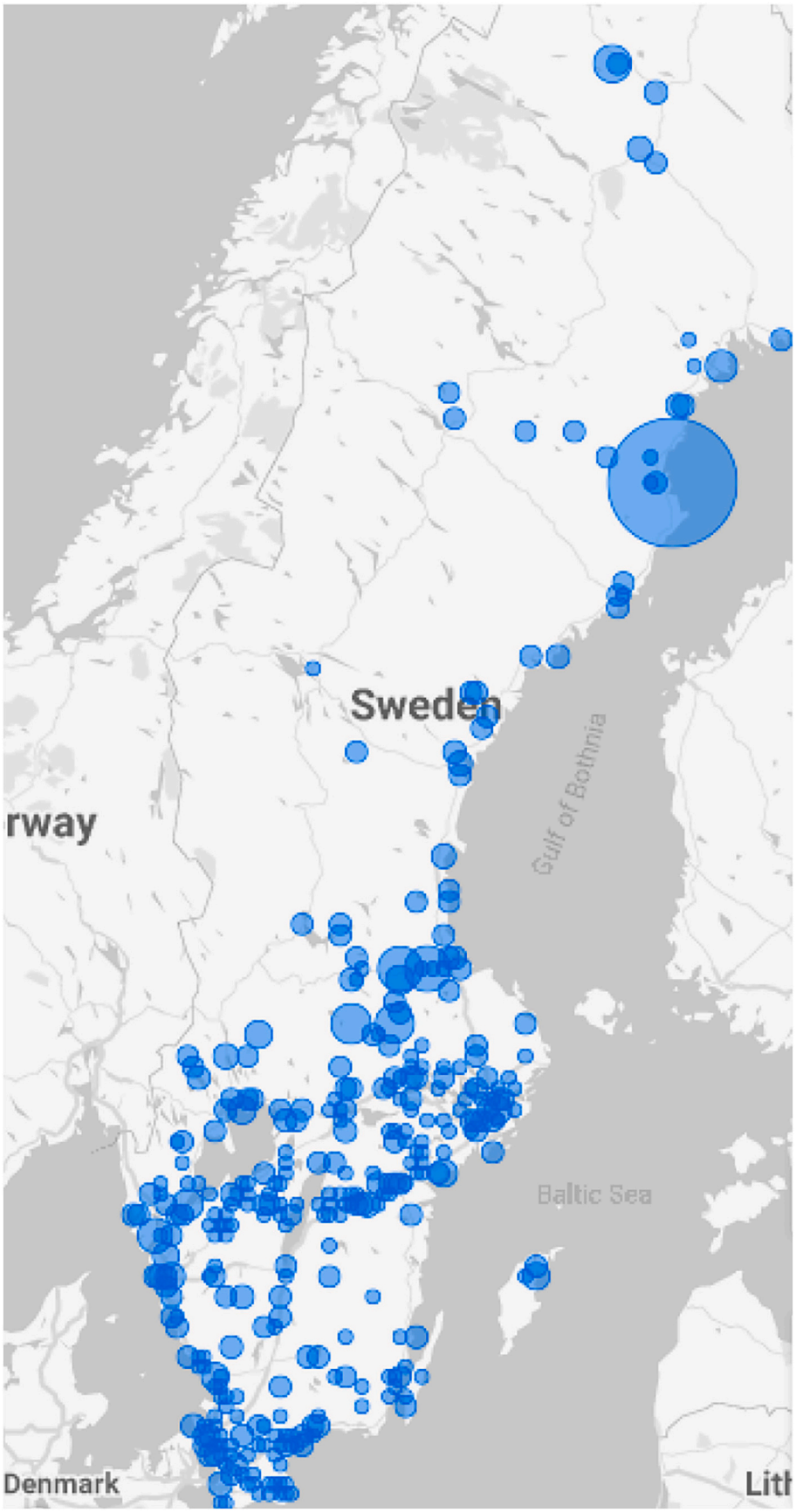

Fig. 4. The human toxicity (CTUh) of substances emitted from Swedish point sources to air and water in the period 2001–2017 with UsEtox 2.12. Calculations are based on the USEtox Northern European CFs. Coordinates are rounded to the 1st decimal (10 km precision) and toxicity is aggregated at this level.

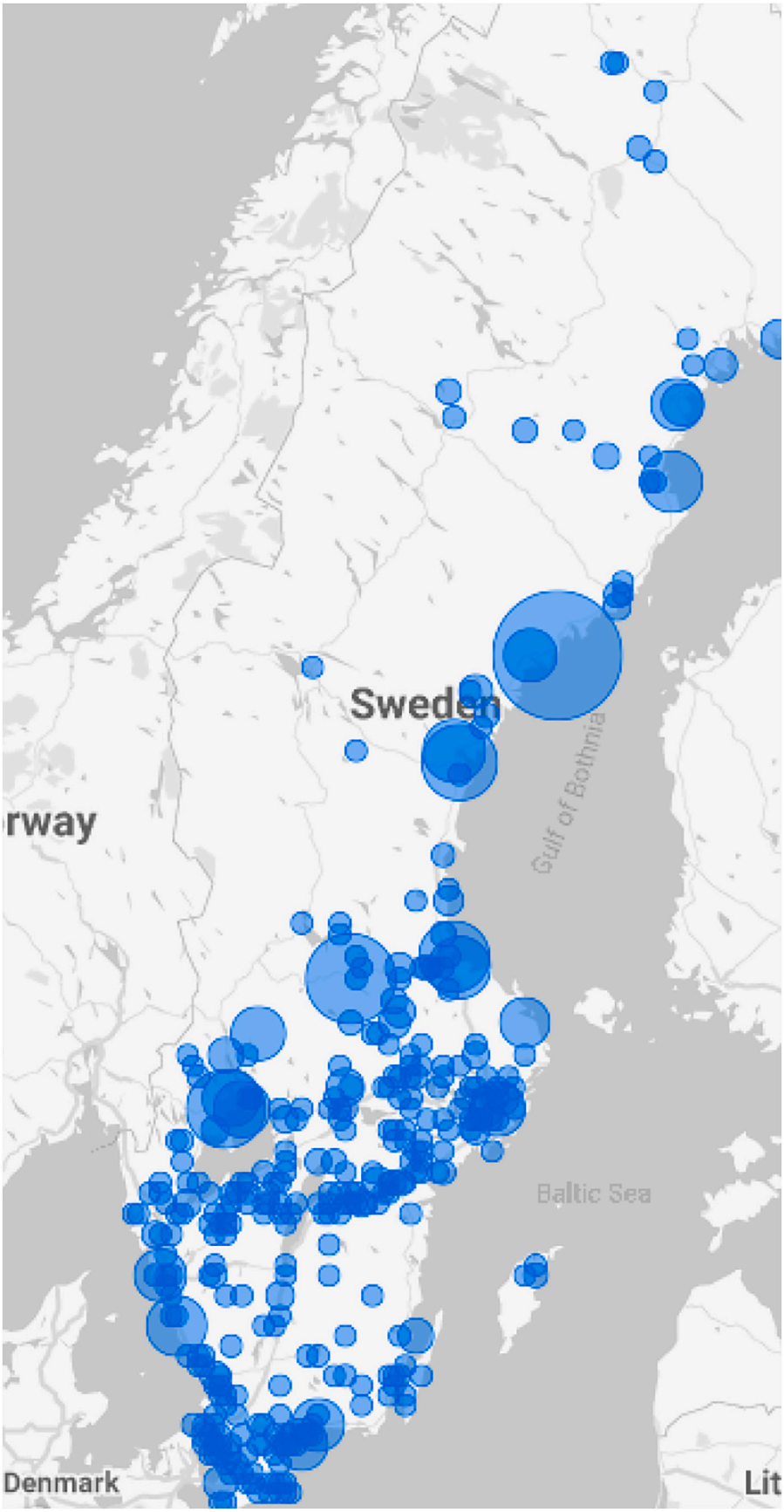

Fig. 5. The ecotoxicity (CTUe) of substances emitted from Swedish point sources to air and water in the period 2001–2017 with UsEtox 2.12. Calculations are based on the USEtox Northern European CFs. Coordinates are rounded to the 1st decimal (10 km precision) and toxicity is aggregated at this level.

Table 3
The facilities with the largest contribution to human toxicity (CTUh), emitted from Swedish point sources to air and water in 2001–2017, characterized with USEtox 2.12. Only the most relevant 20 facilities are shown. Assumptions made in characterisation are given in the footnotes of Table 2, and follow Sörme et al. (2016). Calculations were based on the subcompartment level.

Company name	Main NACE economic activity	Human toxicity (CTUh)	Eco toxicity (CTUe)	Human toxicity (rank)	Eco toxicity (rank)
RÖNNSKÄRSVERKEN	Copper production	1.23E+02	7.49E+10	1	6
LKAB - Kirunagruvan	Mining of iron ores	1.82E+01	2.69E+09	2	94
Outokumpu Stainless AB, Avesta	Manufacture of basic iron and steel and of ferro-alloys	1.80E+01	1.76E+10	3	34
OVAKO Hofors AB	Manufacture of basic iron and steel and of ferro-alloys	1.53E+01	1.05E+10	4	45
OVAKO BAR AB	Manufacture of basic iron and steel and of ferro-alloys	1.40E+01	3.18E+09	5	88
Sandvik AB	Activities of head offices	1.18E+01	3.14E+09	6	89
OVAKO Steel AB	Manufacture of basic iron and steel and of ferro-alloys	1.08E+01	1.53E+09	7	125
Vargön Alloys AB	Manufacture of basic iron and steel and of ferro-alloys (ECSC)*	1.07E+01	1.06E+10	8	44
AB Sandvik Materials Technology	Manufacture of steel tubes	1.03E+01	2.28E+09	9	100
Ovako Sweden AB, Hofors	Manufacture of basic iron and steel and of ferro-alloys	9.00E+00	4.81E+09	10	81
SSAB Tunnplåt AB	Manufacture of basic iron and steel and of ferro-alloys	8.13E+00	1.01E+10	11	47
INEOS Sverige AB	Manufacture of plastics in primary forms	7.65E+00	5.88E+06	12	236
FUNDIA SPECIAL BAR AB	Manufacture of basic iron and steel and of ferro-alloys (ECSC)*	7.48E+00	2.19E+09	13	103
Höganäs Halmstadsverken	Manufacture of basic iron and steel and of ferro-alloys	6.86E+00	6.07E+09	14	72
Hagfors Järnverk	Manufacture of basic iron and steel and of ferro-alloys	6.76E+00	2.52E+10	15	25
Sävenäs	Steam and air conditioning supply	6.73E+00	1.81E+09	16	110
SSAB Oxelösund AB	Manufacture of basic iron and steel and of ferro-alloys	6.67E+00	5.42E+09	17	76
HÖGDALENVERKET	Steam and air conditioning supply	6.54E+00	5.29E+06	18	237
AB Sandvik Materials Technolog	Activities of head offices	5.84E+00	1.90E+09	19	109
Eka Chemicals AB	Manufacture of other inorganic basic chemicals	5.65E+00	5.23E+06	20	239

Table 4
The facilities with the largest contribution to eco-toxicity (CTUe), emitted from Swedish point sources to air and water in 2001–2017, characterized with USEtox 2.12. Only the most relevant 20 facilities are shown. Assumptions made in characterisation are given in the footnotes of Table 2, and follow Sörme et al. (2016). Calculations were based on the subcompartment level.

Company name	Main NACE economic activity	Human toxicity (CTUh)	Eco toxicity (CTUe)	Human toxicity (rank)	Eco toxicity (rank)
Gruvöns bruk	Manufacture of paper and paperboard	1.12E+00	1.08E+11	56	1
STORA ENSO KVARNSVEDEN AB	Manufacture of paper and paperboard	1.75E+00	1.06E+11	37	2
Korsnäsverken	Manufacture of paper and paperboard	1.95E+00	9.24E+10	36	3
Metsä Board Sverige AB, Husums fabr	Manufacture of pulp	8.40*E*-01	8.91E+10	64	4
M-real Sverige AB, Husums fabrik	Manufacture of paper and paperboard	1.42E+00	8.30E+10	46	5
RÖNNSKÄRSVERKEN	Copper production	1.23E+02	7.49E+10	1	6
Södra Cell Värö	Manufacture of pulp	1.05E+00	7.31E+10	58	7
Nya Svenska Rayon AB	Manufacture of man-made fibres	0.2011915766	67321265080	106	8
Skoghalls Bruk	Manufacture of paper and paperboard	2.88E+00	6.61E+10	30	9
Skutskärs Bruk	Manufacture of pulp	9.45*E*-01	6.44E+10	62	10
HENRIKSDALS RENINGSVERK	Sewerage	0.3773718127	63131888459	83	11
Domsjö Fabriker AB	Manufacture of pulp	0.4148755524	59209901527	80	12
Södra Cell Mörrum	Manufacture of pulp	9.92E-01	5.91E+10	60	13
HALLSTA PAPPERSBRUK	Production of electricity	0.3941339881	53890197252	81	14
STORA ENSO NYMÖLLA AB	Manufacture of pulp	0.333693123	49097236051	88	15
Ortvikens pappersbruk	Manufacture of paper and paperboard	1.57E+00	4.86E+10	40	16
Smurfit Kappa Kraftliner Piteå	Manufacture of paper and paperboard	2.60E+00	4.77E+10	31	17
SCA Ortviken	Manufacture of paper and paperboard	1.55E+00	4.57E+10	41	18
SCA Munksund	Manufacture of paper and paperboard	1.41E+00	3.90E+10	47	19
SCA Östrands massafabrik	Manufacture of pulp	6.03-01	3.85E+10	69	20

emissions decreased very much, from 11,500 kg in 2007 to just 9 kg in 2009 and this structural change can be seen as a technical indicator of data errors.

Our study addresses point source emissions, which are emitted from unique places and hence are easier to identify. The *E*-PRTR data covers pollutants which enter the environment from point sources, for example from smokestacks or from discharge pipes of EU facilities. Nonpoint source pollution is much more difficult to observe or track and is covered neither by the E-PRTR database nor by our study. Nonpoint source pollutants are emitted in a wider area usually in lower concentration, but can later concentrate after for example transmitted away by meteorological movements, by rain, rivers or wind, Nonpoint source pollutants can be emitted by motor vehicles, unreported smaller facilities and by almost any kind of smaller scale human activities. Hence, nonpoint source pollution can increase total pollution and toxicity impacts. As a final remark, our calculations only focus on point source pollutant releases and hence, underestimate the real national toxicity potential. Diffuse emissions of some pollutants have been shown to be larger than point sources nationally in Sweden (Sörme et al., 2016).

Our methodology followed Sörme et al. (2016) and calculated the chemical footprint of Swedish facilities by the additive aggregation formula. Such a choice is common in the international practice, however it has important consequences (Erhart, 2021). An undesirable feature of additive aggregations is the implied full compensation, such that high

emission in some pollutants can be compensated for by sufficiently low values in other pollutants. An alternative approach would be to use a non-compensatory formula (for example based on a geometric aggregation function) at the facility, industry or national level. Furthermore, the additive toxicity calculation formula in our analysis does not take into account the large number of possible interactions. Especially, the investigation of toxicity consequences from zinc's interaction with other pollutants could be a potential research direction. Sanstead (1995) for example gave evidence that risk of copper deficiency is increased when the molar ratio of zinc to copper (Zn:Cu) is high.

A further limitation of the chemical footprint analysis is that the list of pollutants in the *E*-PRTR and the USEtox model can not be fully matched. Another obstacle is the possible inaccuracy and inconsistency in pollutant release estimates Sullivan and Gouldson (2007), Taylor et al. (2020).

Sörme et al. (2016), Nordborg et al. (2017) noted that the recommended CFs for organic substances have an estimated uncertainty range of up to 2 and 3 orders of magnitude for ecotoxicity and human toxicity, respectively, primarily related to input data (Rosenbaum et al., 2008). Hence the previous authors have not considered these uncertainties in the calculations, and neither did we. Since metals were recognized as a priority group of pollutants for both human toxicity and ecotoxicity, we follow Sörme et al. (2016) and remark that all metal CFs are classified as "indicative" in both model versions. The related uncertainties have not been quantified, but are larger than the uncertainties associated with organic substances, considering that USEtox is primarily developed for organic substances (Rosenbaum et al., 2008).

Our study confirms the likely importance of (eco)toxicity impacts of metals, although challenges associated with assessing this impact have been prevailing. We found that Mercury's importance in the longer period between 2001 and 2017 has increased by 18 percentage points to 45% compared to 27% calculated for 2008.

We aimed at increasing the precision of earlier calculations by Sörme et al. (2016), Nordborg et al. (2017) and draw distinctions between subcompartments (freshwater/seawater and urban/rural air). The distinctions did not change the results substantially, neither in terms of absolute human toxicity impact potential nor in terms of pollutant rankings. One should note, however, that the parameter influencing the distinction between seawater and freshwater could be better optimized. We classified pollutant releases into seawater, where the facilities were closer to the seacoast than 500 m. This choice can be interpreted as a simple and conservaive technical rule which needs further optimization in the future.

As the E-PRTR dataset of the European Environmental Agency (EEA) does not contain information on Biochemical Oxygen Demand (BOD), we could not analyse the role of BOD in aquatic systems. BOD and organic pollution frequently stem from waste water treatment plant discharges, industrial effluents and agricultural run-off. Organic pollution may cause severe de-oxygenation of freshwater, and lead to the disappearance of fish and aquatic invertebrates. However, it should be noted that one of the key point sources of organic waste and BOD are in the paper industries, the ecotoxity of which is presented in our analysis. According to the EEA, the share of monitored river sites with BOD not satisfying recommendations for salmonid waters exceeds 50% in three out of 23 assessed countries: Kosovo under UNSCR 1244/99, Spain and Sweden, (European Environmental Agency, 2021).

Fig. 6 presents the scatter plot of the human toxicity (CTUh) versus the ecotoxicity (CTUe) of substances emitted from Swedish point sources to air and water in the period 2008 calculated with UsEtox 2.12. The results suggest that major emitters either have human toxicity potential or ecotoxicity potential. There are only a few facilities which have toxicity potential in both ways. The Pearson correlation coefficient between the CTUh and CTUe is 0.1, not significantly different from zero. As a consequence, there are economic activities, which have toxicity impact potential either for the humans or for the environment. All this could pose a difficulty for designing a clear industrial transformation by supporting some industries and penalizing others. Careful management of this trade-off between human and ecotoxicity remains hence an important research question and a hot public policy issue.

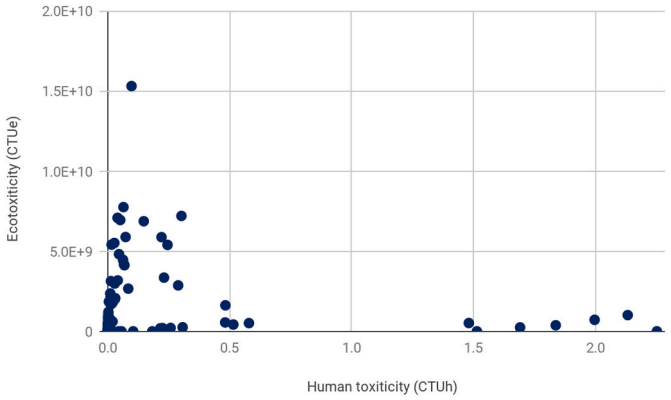

Fig. 6. Scatter plot of the human toxicity (CTUh) versus the ecotoxicity (CTUe) of substances emitted from Swedish point sources to air and water in the period 2008 calculated with UsEtox 2.12. Only the major emitters are shown. Calculations are based on the Northern European CFs.

5. Conclusions

We conclude that the 2.12 USEtox model version produces relatively consistent results with earlier versions, except for copper, whose ecotoxicity characterisation factor has been changed substantially. Our results are significantly important due to the fact that USEtox subcompartment level toxicity characterisation factors are used for the first time and matched with point source industrial pollutant releases on the basis of EUROSTAT GISCO population density and distance-to-coast grid data. We showed that results are largely invariant to the choice of air sub-compartments, as the urban air and rural air characterisation factors for most toxic substances are roughly equal. Characterisation factors for freshwater and seawater are less similar, but our conservative choice of classifying emissions into water as seawater emissions, when the source point distance to the seacoast is less than 500 m, does not influence the results at the aggregated level. Better identification methods to capture pollutant releases into seawater and freshwater can help advancing towards a more precise chemical footprint analysis.

We recalculated the results of Sörme et al. (2016) and Nordborg et al. (2017) with the 'Northern Europe and Northern Canada' regional USEtox landscape setting instead of the default setting and showed that the calculated human toxicity for 2008 collapsed to its tenth, to 29 CTUh from 360 CTUh.

We aggregated human toxicity across years from 2001 to 2017. We found that Mercury's importance in this longer period has increased by 18 percentage points to 45% compared to 27% calculated for 2008.

Following Nordborg et al. (2017)'s suggestion we broadened the analysis and monitored the toxic impacts from economic activities in Sweden over time, in the period between 2001 and 2017. Our analysis shows a downward sloping total human toxicity and ecotoxicity impacts over time. The decreasing trend of human toxicity is more obvious for zinc and arsenic than for mercury or lead. Ecotoxicity impacts have been also decreasing in total, in particular in early 2000s and less dynamically recently. Especially Cd 5% annual average decrease can be categorized

as significant. Uncertainties are large, especially in the case of metals due to input data and hence, we should clearly communicate this uncertainty. We confirm the results of Nordborg et al. (2017) that USEtox pinpoints a substance (Zn) which has been described as "relatively harmless" to humans, and this may need further research to resolve.

Also, we calculated toxicity at the facility level. Understanding and measuring environmental impacts of individual companies and facilities is currently needed for environmental accounting, for informed consumer decisions and to scale up financing to clean production and sustainable finance. Companies in the manufacturing sector of metals are estimated to have the largest human toxicity potential in Sweden in the period between 2001 and 2017. At the industry level the paper manufacturing sector had the largest eco-toxicity footprint. Our results suggest that major emitters either have human toxicity potential or ecotoxicity potential. There are only a few large facilities which have toxicity potential in both ways. All this could pose a difficulty for designing clear transformation by supporting some industries and penalizing others. Careful management of this trade-off between human and ecotoxicity remains to be, hence, an important research question and a hot public policy issue.

Author statement

All persons who meet authorship criteria are listed as authors, and all authors certify that they have participated sufficiently in the work to take public responsibility for the content, including participation in the concept, methodology, software, data curation, writing, original draft preparation, visualisation.

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.

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