Diffuse Sources of Cadmium, Nickel and Lead to Water in European Countries

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EXECUTIVE SUMMARY

A screening assessment for some diffuse sources of cadmium (Cd), nickel (Ni) and lead (Pb) to water has been undertaken to accompany the ETAP report on the sources of 8 elements (including Cd, Ni and Pb) to and discharged from European sewage treatment works. The same methodology was employed as that for a previous assessment of diffuse sources of copper to water. Available data from the scientific and grey literature, NGO, government and industry reports, were used in conjunction with available European datasets including land use, water use, average rainfall, agricultural land areas and arable and livestock breakdowns per country, soil loss as well as biosolid (sewage sludge) and inorganic fertiliser spreading on land.

Based on a combination of quantities of materials used, the concentration of the elements within the materials and estimations of loss from land it was possible to estimate loads of Cd, Ni and Pb entering European soils from biosolids, inorganic NPK fertilisers, farmyard manure (FYM – broken down into contributions from pigs, cattle, goats, poultry and sheep) and atmospheric deposition. Based on modelled loss of added elements to a variety of soils combined with reported partition coefficients, estimated losses from soil to water were calculated. Other diffuse sources quantified were septic tank discharges to water and soil erosion, where metals were considered to be part of the mineral matrices of clay minerals etc. To provide a comparison a sum of STP discharges to water were included from the accompanying ETAP report as well as that reported from the ePRTR and loads of lead entering the aquatic environment from lead sinkers and ammunition taken from a separate report focussing on lead.

A summary table is provided below for the summed EU 27 countries. Erosion and loss of agricultural soil containing metals within the mineral phase were estimated to be the greatest load for all three elements. ETAP metal loads from STPs were higher than ePRTR estimates which was not surprising owing to not all STP discharges being captured within the ePRTR. Agriculture contributed significant loads, dominated by atmospheric deposition and FYM, in particular manure from poultry, reflecting higher levels of these elements in this type of manure and the large numbers of birds reared in Europe.

Atmospheric deposition was influenced heavily by estimates for France and Spain which accounted for around 50% of the load owing to relatively high concentrations reported in rainwater and a large proportion of the country given over to agriculture. Atmospheric deposition directly into inland waters was small compared with agricultural land as is a much lower contribution to land area within most countries. However, it equated to around 50% of load to water because only ~10% of land deposition is assumed to be washed off into water compared with 100% of direct deposition onto water.

Data from another report suggested that ammunition dominated anthropogenic inputs of lead to water.

Executive Summary Table: Summary of estimated diffuse loads of Cd, Ni and Pb compared with point source data generated from other reported

	Loads (kg/d)	to soil Cd	to water Cd	to soil Ni	to water Ni	to soil Pb	to water Pb
	Soil background		521		73803		43800
ePRTR	STP		22		448		120
ETAP	STP		32		668		n/a
	Agriculture Total	271	27	2938	292	3452	301
	Septic tank		2		16		37
	Lost lead sinkers		#N/A		#N/A	#N/A	110
	Lead ammunition		#N/A		#N/A	68008	4781
	Atmospheric deposition on inland water		6.1		65		118
Agriculture break down	Sludge to land	10.5		208		395	
	NPK fertilisers	29		53		13	
	FYM fertilisers cows	11		187		136	
	FYM fertilisers pigs	3.3		84		48	
	FYM fertilisers sheep	2.8		0.96		63	
	FYM fertilisers goats	0.13		0.2		0.2	
	FYM fertilisers poultry	128		1116		815	
	Atmospheric deposition on agricultural land	86		1289		1982	

[1] Data provided from the ETAP report (2021) "Sources and fate of metals and metalloids in wastewater treatment plants. Final report to the ETAP from wca, University of Plymouth and Derac, March, 2021.

[2] Data from Arche Consulting (2020) Pb emission inventory for the environment. Final Report, 3/12/2020. Ferencz N., Eliat M. and Verdonck F.

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1 Diffuse sources of Cd, Ni, Pb to water

Sources of metal to soil are numerous (Figure 1) and very varied across European countries. Cadmium, nickel andlead occurrence in agricultural soil will be subject to the underlying geology, inputs from fertilisers and biocides. Fertilisers comprise farmyard manure, inorganic fertilisers and in some countries sewage sludge. Consequently, observed concentrations of these metals of interest in European agricultural soils will be dependent on land use which itself determines the type of fertiliser used.

This brief report represents a first pass at generating a methodology and populating estimates for loads of cadmium, nickel and lead loads for individual European countries to water from agricultural, atmospheric and septic tank sources. Owing to a constrained time and budget, it does not include an exhaustive search of the literature nor an appraisal of all of the potentially significant sources of these elements; rather an opportunity to demonstrate the methodologies that can be applied with the available datasets and their application to readily available information.

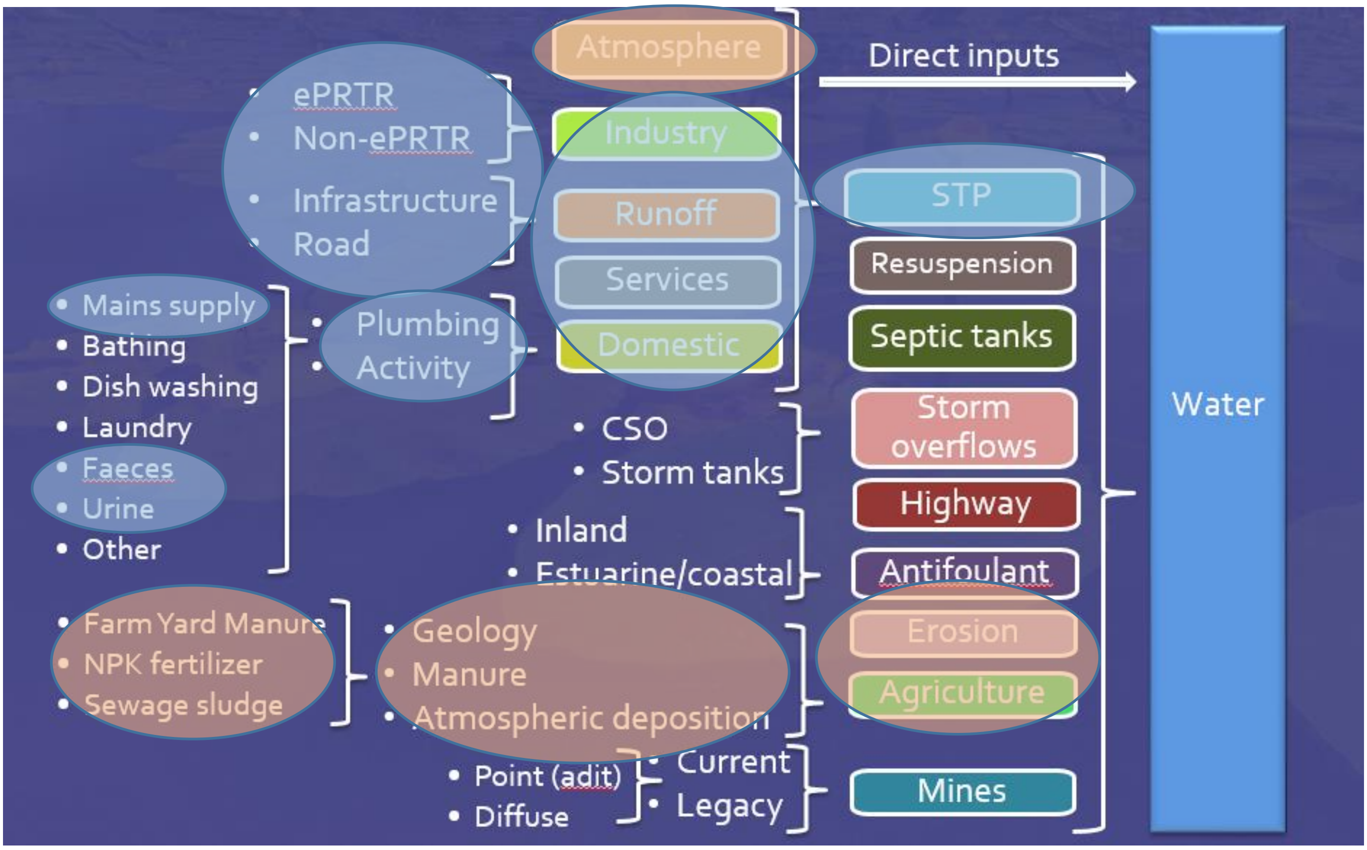

Figure 1 **A generic schematic of the major sources quantified for each metal and for individual European Countries. Pink highlights are covered in**

these calculations; blue highlights denote sources covered in the ETAP project

2 Agricultural sources

2.1 Biocide use

Based on available information there is no significant source of cadmium, nickel or lead in agricultural biocides. The elements may be present at very low concentrations as impurities in for example, copper based products, but the significance was considered too small to include in this preliminary assessment.

2.2 Cd, Ni, Pb in sewage sludge

Sewage sludge is a valuable source of nutrient that is recognised by many European countries and under the EU Sewage Sludge Directive (1986)[1]. It may be recycled to land provides it meets a number of stringent criteria, including a limit value for a number of metals including cadmium, nickel and lead, although this value may be set lower by individual countries. Sludge is typically applied to cereals, pastureland and some beet but not most fresh fruit and vegetables. However, for a variety of reasons the recycling of sludge to land varies from as high as ~80% of all sludge produced (UK) down to zero (Netherlands). Because of the tight regulation there is up to date data for sludge recycled to land and the cadmium, nickel and lead content within the sludge.

Loads to land were then calculated as reported concentrations multiplied by the dry matter recycling of sludge where appropriate. As noted some countries do not recycle sludge to land and others do not report quantities and therefore have not been included in the estimates. Where concentrations were not reported but load of sludge were, then the mean cadmium, nickel and lead concentration was applied to derive a load (Table 1).

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[1] Council Directive 86/278/EEC of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture

Table 1 EU sludge recycled to land and Cd, Ni and Pb loadings.

		Concentration (mg kg^{-1}dm)			Load (kg d^{-1})		
Country	Sludge to land t yr^{-1}dm	Cd	Ni	Pb	Cd	Ni	Pb
Albania	10300	1.9	26.3	54.9	0.1	0.7	1.5
Austria	48313	1.9	26.3	54.9	0.3	3.5	7.3
Belgium	30620	1.25	18	86	0.1	1.5	7.2
Bosnia	0	1.9	26.3	54.9	0.0	0.0	0.0
Bulgaria	22500	1.6	13	55	0.1	0.8	3.4
Croatia	1086	1.9	26.3	54.9	0.01	0.1	0.2
Cyprus	1613	6.9	21	23	0.03	0.1	0.1
Czech republic	102940	1.5	29	40	0.4	8.2	11.3
Denmark	74000	1.9	26.3	54.9	0.4	5.3	11.1
Estonia	100	2.8	19	41	0.001	0.001	0.0
Finland	4,200	0.6	30	8.9	0.0	0.3	0.1
France	299000	1.3	21	50	1.1	17.2	40.9
Germany	423497	1	25	37	1.2	29.0	42.9
Greece	21528	1.9	26.3	54.9	0.1	1.6	3.2
Hungary	28200	1.4	26	36	0.1	2.0	2.8
Iceland	n/d	1.9	26.3	54.9	n/d	n/d	n/d
Ireland	46487	1.9	26.3	54.9	0.2	3.4	7.0
Italy	315600	1.3	66	101	1.1	57.0	87.3
Latvia	3316	3.6	47	114	0.03	0.4	1.0
Liechtenstein	n/d	1.9	26.3	54.9	n/d	n/d	n/d
Lithuania	20817	1.3	25	21	0.1	1.4	1.2
Luxembourg	1138	1.9	26.3	54.9	0.01	0.1	0.2
Malta	0	1.9	26.3	54.9	0.0	0.0	0.0
Montenegro	n/d	1.9	26.3	54.9	n/d	n/d	n/d
Netherlands	0	1.9	26.3	54.9	0.0	0.0	0.0
N Macedonia	n/d	1.9	26.3	54.9	n/d	n/d	n/d
Norway	66000	1.9	26.3	54.9	0.3	4.8	9.9
Poland	108520	1.9	26.3	54.9	0.6	7.8	16.3
Portugal	13885	0.2	15	27	0.01	0.6	1.0
Romania	35000	1.9	26.3	54.9	0.2	2.5	5.3
Serbia	n/d	1.9	26.3	54.9	n/d	n/d	n/d
Slovakia	0	2.5	26	57	0.0	0.0	0.0
Slovenia	0	0.7	29	29	0.0	0.0	0.0
Spain	754740	2.1	30	68	4.3	62.0	141
Sweden	69500	0.9	15	24	0.2	2.9	4.6
Switzerland	n/d	1.9	26.3	54.9	n/d	n/d	n/d
Turkey	9260600	1.9	26.3	54.9	48.9	668	1392
UK	844400	1.9	26.3	54.9	4.5	60.9	126.9
EU27					10.5	208	395

[1] Eurostat data (2018 or latest data): https://ec.europa.eu/eurostat/web/products-datasets/product?code=env_ww_spd
[2] Mean of available data

2.3 Cadmium, nickel and lead in NPK inorganic fertiliser

Fertilisers derived from phosphate fertilisers will inevitably contain a small proportion of elemental impurities from the source mineralogy or materials used in their production. Cadmium has been highlighted as a significant impurity and has been subject to controls.

Concentrations of cadmium, nickel and lead in a range of fertilisers has been recently reported.[2] Table 2 summarises the concentrations.

Table 2 Concentrations of cadmium, nickel and lead in inorganic fertilisers

	Concentration (mg kg^{-1})		
Fertiliser type	Cd	Ni	Pb
Triple super phosphate	17.6	28.6	6.6
Other phosphate fertilisers	9.7	18.8	2.9
Nitrates fertilisers	10.7	18.8	5.1
PK only	9	15	2
Low N	6.9	18.1	5.7
High N	2.3	3.5	2.4
mean	**9.4**	**17.1**	**4.1**
median	**9.4**	**18.5**	**4.0**

Furthermore Eurostat data are available for fertilisers used within the EU.[3] Nitrate fertilisers are predominantly generated using the Haber-Bosch process utilising nitrogen and hydrogen gases to create ammonia; it was therefore assumed that trace metal impurities from this industrial process would be minimal. Based on the Eurostat country by country nutrient usage it was possible to calculate a cadmium, nickel and lead load entering European soils. Concerns regarding the cadmium content in inorganic P fertilisers has seen limits applied by some EU countries, although there appears not to an EU wide agreement at this time only proposals (Ulrich, 2019). Estimated loadings are 0.9 Ca ha^{-1}yr^{-1} based on an average application rate (Smolders and Six, 2013) based on an assumed Cd content of 36 mg Cd kg^{-1} P_2O_5, which is not dissimilar to the 21 mg Cd kg^{-1} P_2O_5 estimated using the reported data in Table 3 and probably reflects the gradual influence of using low cadmium fertilisers in the EU over the past decade.

[2] https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/545559/Material_comparators_for_materials_applied_to_land_-_manufactured_fertilisers.pdf

[3] https://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=aei_fm_usefert&lang=en

Table 3 Cadmium, nickel and lead loading to soil from inorganic fertilisers

Country	2018 or latest fertiliser use Kg–P d^{-1}	Cd Kg d^{-1}	Ni Kg d^{-1}	Pb Kg d^{-1}
Albania	38,850	0.36	0.7	0.2
Austria	35,893	0.34	0.6	0.1
Belgium	13,350	0.13	0.2	0.1
Bulgaria	91,116	0.85	1.6	0.4
Croatia	42,612	0.40	0.7	0.2
Cyprus	6,431	0.06	0.1	0.0
Czech Republic	61,489	0.58	1.1	0.3
Denmark	56,947	0.53	1.0	0.2
Estonia	11,121	0.10	0.2	0.05
Finland	30,207	0.28	0.5	0.1
France	455,447	4.27	7.8	1.9
Germany	249,273	2.33	4.3	1.0
Greece	70,872	0.66	1.2	0.3
Hungary	140,047	1.31	2.4	0.6
Iceland	5,602	0.05	0.10	0.02
Ireland	127,001	1.19	2.2	0.5
Italy	323,581	3.03	5.5	1.3
Latvia	31,433	0.29	0.5	0.1
Lithuania	61,396	0.58	1.1	0.3
Luxembourg	1,084	0.01	0.0186	0.004
Malta	153	0.001	0.003	0.001
Netherlands	16,460	0.15	0.3	0.1
Norway	23,773	0.22	0.4	0.1
Poland	404,832	3.79	6.9	1.7
Portugal	57,090	0.53	1.0	0.2
Romania	225,227	2.11	3.9	0.9
Slovenia	11,269	0.11	0.2	0.0
Slovakia	30,689	0.29	0.5	0.1
Spain	509,194	4.77	8.7	2.1
Sweden	39,151	0.37	0.7	0.2
Switzerland	11,118	0.10	0.2	0.05
Turkey	622,872	5.83	10.7	2.6
UK	224,668	2.10	3.8	0.9
EU27	**3,103,365**	**29**	**53**	**13**

2.4 Cadmium, nickel and lead in farmyard manure

Farmyard manure (FYM) is a waste product of animal husbandry, but also a valuable fertiliser. It was therefore assumed that all animal manure produced in the EU is returned to soil of the EU. Most farms spread their animal manure back onto soil. In some cases (mostly poultry) it may be processed into manure, but the assumption is it will still return to the country's soil of origin. Data were firstly collected on the concentration of cadmium, nickel and lead in FYM (Table 4). As can be seen from the table below, there is significant variation between studies likely to reflect, breed, age and feeding regimes.

Table 4 Cadmium, nickel and lead content of FYM

	Concentration (mg kg^{-1})			
FYM type	**Cd**	**Ni**	**Pb**	**Reference**
Cattle	0.2	15.4	4.26	Deltares (2017)
Cattle	0.36	5.76	4.94	Bolan et al. (2010)
Cattle	0.45	0.13	0.34	Adesoye et al. (2014)
Cattle	0.42		5.2	Wang et al. (2013)
Cattle	0.7	9.6	7.5	Chauhan et al. (2008)
Cattle	0.27	6.3	4.1	Sager (2007)
Cattle		5.8		Svane and Karring (2009)
Mean	**0.38**	**6.23**	**4.53**	
Pig	0.28	14.4	9.81	Deltares (2017)
Pig	0.02	0.15	0.28	Adesoye et al. (2014)
Pig	0.64			Xu et al. (2015)
Pig	0.59		6.8	Wang et al. (2013)
Pig	0.66	11.3	9.0	Chauhan et al. (2008)
Pig		10.38		Svane and Karring (2009)
Pig	0.395	10.7	2.25	Sager (2007)
Mean	**0.39**	**9.93**	**5.63**	
Poultry	0.13	5.98	0.6	Deltares (2017)
Poultry	1.3	8.49	7.66	Bolan et al. (2010)
Poultry	0.53	1.89	1.17	Ulkpe and Chokor (2018)
Poultry	0.08	0.40	0.36	Adesoye et al. (2014)
Poultry	0.89		5.1	Wang et al. (2013)
Poultry	0.25	8.0	6.7	Chauhan et al. (2008)
Poultry	0.46	8.5	5.4	Sager (2007)
Mean	**0.90**	**7.5**	**5.48**	
Goat	**0.15**	**0.24**	**0.24**	**Adesoye et al. (2014)**
Sheep	**0.7**	**0.24**[1]	**15.8**	**Wang et al. (2016)**

[1] assumed to be the same as goats owing to lack of data.

Based on concentrations of the metals in manure and knowledge of average manure production per day (Lorimer et al., 2004) it was possible to calculate the mean amount of cadmium, nickel and lead excreted per head of each category of animal. Eurostat databases (2018) provided data of head of farm animal per country[4]. Poultry data were generated from the amount slaughtered per year[5], divided by the average chicken weight (2.5kg)[6]. With this data collated it was a simple matter of multiplying the amount of metal excreted per day by the number of animals to derive a load, all of which is assumed to be returned to agricultural land (Table 5).

[4] https://ec.europa.eu/eurostat/documents/3217494/9455154/KS-FK-18-001-EN-N.pdf/a9ddd7db-c40c-48c9-8ed5-a8a90f4faa3f

[5] https://ec.europa.eu/eurostat/databrowser/view/tag00043/default/table?lang=en

[6] https://beyondthetreat.com/chicken-weight/

Table 5 Animal numbers and mass of Cd, Ni and Pb in manure (n/d = no data)

	Million animals					Cd (kg day^{-1})					Ni (kg day^{-1})					Pb (kg day^{-1})				
	Cows	Pigs	Sheep	Goats	Poultry	Cows	Pigs	Sheep	Goats	Poultry	Cows	Pigs	Sheep	Goats	Poultry	Cows	Pigs	Sheep	Goats	Poultry
Belgium	2.4	6.1	:	:	179	0.3	0.14	n/d	n/d	4.4	5.7	3.54	n/d	n/d	38.7	4.1	2.01	n/d	n/d	28.3
Bulgaria	0.6	0.6	1.3	0.3	46	0.1	0.01	0.06	0.00	1.1	1.4	0.35	0.020	0.005	9.9	1.0	0.20	1.32	0.01	7.2
Czech Rep	1.4	1.5	:	:	67	0.2	0.03	n/d	n/d	1.7	3.3	0.87	n/d	n/d	14.5	2.4	0.49	n/d	n/d	10.6
Denmark	1.6	12.8	:	:	64	0.2	0.29	n/d	n/d	1.6	3.8	7.43	n/d	n/d	13.7	2.8	4.21	n/d	n/d	10.0
Germany	12.3	27.6	1.6	0.1	634	1.8	0.63	0.07	0.00	15.7	29.1	16.02	0.025	0.002	136.8	21.1	9.08	1.62	0.00	100.0
Estonia	0.3	0.3	:	:	14	0.0	0.01	n/d	n/d	0.3	0.7	0.17	n/d	n/d	3.0	0.5	0.10	n/d	n/d	2.2
Ireland	6.7	1.6	3.9	:	67	1.0	0.04	0.18	n/d	1.7	15.9	0.93	0.060	n/d	14.4	11.5	0.53	3.95	n/d	10.5
Greece	0.6	0.7	8.6	3.8	92	0.1	0.02	0.39	0.04	2.3	1.4	0.41	0.132	0.063	19.9	1.0	0.23	8.71	0.06	14.5
Spain	6.5	30	16	3.1	694	0.9	0.68	0.72	0.03	17.2	15.4	17.41	0.246	0.052	150.0	11.2	9.87	16.21	0.05	109.6
France	18.6	13.1	6.9	1.2	679	2.7	0.30	0.31	0.01	16.8	44.0	7.60	0.106	0.020	146.7	32.0	4.31	6.99	0.02	107.2
Croatia	0.5	1.1	0.6	0.1	27	0.1	0.03	0.03	0.00	0.7	1.2	0.64	0.009	0.002	5.8	0.9	0.36	0.61	0.00	4.2
Italy	6.3	8.6	7.2	1	520	0.9	0.20	0.32	0.01	12.9	14.9	4.99	0.111	0.017	112.3	10.8	2.83	7.30	0.02	82.0
Cyprus	0.1	0.4	:	:	11	0.0	0.01	n/d	n/d	0.3	0.2	0.23	n/d	n/d	2.3	0.2	0.13	n/d	n/d	1.7
Latvia	0.4	0.3	0.1	0	14	0.1	0.01	0.00	0.00	0.3	0.9	0.17	0.002	0.000	3.0	0.7	0.10	0.10	0.00	2.2
Lithuania	0.7	0.6	0.2	0	40	0.1	0.01	0.01	0.00	1.0	1.7	0.35	0.003	0.000	8.7	1.2	0.20	0.20	0.00	6.3
Luxembourg	0.2	0.1	:	:	4	0.0	0.00	n/d	n/d	0.1	0.5	0.06	n/d	n/d	0.9	0.3	0.03	n/d	n/d	0.7
Hungary	0.9	2.9	1.1	0.1	213	0.1	0.07	0.05	0.00	5.3	2.1	1.68	0.017	0.002	46.0	1.5	0.95	1.11	0.00	33.6
Malta	0	0	0	0	2	0.0	0.00	0.00	0.00	0.0	0.0	0.00	0.000	0.000	0.3	0.0	0.00	0.00	0.00	0.3
Netherlands	4	12.3	1	0.5	166	0.6	0.28	0.04	0.01	4.1	9.5	7.14	0.015	0.008	35.8	6.9	4.05	1.01	0.01	26.1
Austria	1.9	2.8	0.4	0.1	85	0.3	0.06	0.02	0.00	2.1	4.5	1.63	0.006	0.002	18.4	3.3	0.92	0.41	0.00	13.4
Poland	6	11.9	:	:	1037	0.9	0.27	n/d	n/d	25.6	14.2	6.91	n/d	n/d	224.0	10.3	3.91	n/d	n/d	163.7
Portugal	1.7	2.2	2.2	0.3	141	0.2	0.05	0.10	0.00	3.5	4.0	1.28	0.034	0.005	30.4	2.9	0.72	2.23	0.01	22.2
Romania	2	4.4	10	1.5	193	0.3	0.10	0.45	0.02	4.8	4.7	2.55	0.154	0.025	41.7	3.4	1.45	10.13	0.03	30.4
Slovenia	0.5	0.3	:	:	28	0.1	0.01	n/d	n/d	0.7	1.2	0.17	n/d	n/d	6.0	0.9	0.10	n/d	n/d	4.4
Slovakia	0.4	0.6	0.4	0	29	0.1	0.01	0.02	0.00	0.7	0.9	0.35	0.006	0.000	6.2	0.7	0.20	0.41	0.0000	4.5
Finland	0.9	1.1	:	:	56	0.1	0.03	n/d	n/d	1.4	2.1	0.64	n/d	n/d	12.0	1.5	0.36	n/d	n/d	8.8
Sweden	1.4	1.4	0.6	:	65	0.2	0.03	0.03	n/d	1.6	3.3	0.81	0.009	n/d	14.1	2.4	0.46	0.61	n/d	10.3
UK	9.8	4.7	23.3	0.1	796	1.4	0.11	1.05	0.00	19.7	23.2	2.73	0.359	0.002	172.0	16.8	1.55	23.61	0.00	125.6
Switzerland	1.6	1.4	0.2	:	40	0.2	0.03	0.01	n/d	1.0	3.8	0.81	0.003	n/d	8.6	2.8	0.46	0.20	n/d	6.3
Montenegro	0.1	0	0.7	0	1	0.0	0.00	0.03	0.00	0.0	0.2	0.00	0.011	0.000	0.1	0.2	0.00	0.71	0.00	0.1
Albania	0.5	0.2	1.7	:	2	0.0	0.00	0.09	0.00	n/d	0.7	0.12	0.029	0.002	#N/A	0.5	0.07	1.93	0.00	#N/A
Serbia	0.9	2.9	33.7	0.2	37	0.1	0.00	0.08	n/d	0.0	1.2	0.12	0.026	n/d	0.4	0.9	0.07	1.72	n/d	0.3
Turkey	16.1	:	1	10.6	879	0.1	0.07	1.51	0.00	0.9	2.1	1.68	0.519	0.003	7.9	1.5	0.95	34.15	0.00	5.8
Bosnia	0.4	0.5	:	0.1	28	2.3	n/d	0.04	0.11	21.7	38.1	n/d	0.015	0.177	189.9	27.7	n/d	1.01	0.18	138.7
Kosovo	:	:	:	:	18	0.1	0.01	n/d	0.00	0.7	0.9	0.29	n/d	0.002	6.0	0.7	0.16	n/d	0.00	4.4
EU27						**11**	**3.3**	**2.8**	**0.13**	**128**	**187**	**84**	**0.96**	**0.20**	**1116**	**136**	**48**	**63**	**0.20**	**815**

2.5 Background Cd, Ni and Pb in soil

Based on underlying geology there will be a background signal of metals from natural soils. The Foregs database[7] provides concentrations across numerous countries (Table 6). For reference Smolders and Six (2013) report a cadmium background concentration in European soils of 0.28 mg kg^{-1}. Consideration was given to using the GEMAS metals dataset for soil concentrations. However the GEMAS data was generated for agricultural soils which were already subject to anthropogenic contamination. The objective of the calculations below was to estimate background, 'natural' sources of the elements of interest to generate loads to water from natural erosion processes. The Foregs dataset was designed to target unamended soil and therefore better reflects the natural geology of the different regions.

Table 6 Background Cd, Ni and Pb concentrations in European soils

	Mean soil concentration (mg Kg^{-1})		
Country	**Ni**	**Pb**	**Cd**
Albania	652.5	13.5	0.36
Austria	25.2	27.1	0.37
Belgium	29.8	32.8	0.87
Switzerland	55.3	36.2	0.54
Czech Republic	17.5	28.1	0.26
Germany	16.8	25.9	0.34
Denmark	3.4	4.3	0.04
Estonia	9.1	11.6	0.14
Spain	25.6	26.9	0.26
Finland	9.3	5.5	0.07
France	23.7	36.3	0.41
Greece	171.0	39.2	0.83
Croatia	35.5	19.7	0.33
Hungary	18.2	13.8	0.17
Ireland	22.0	19.5	0.51
Italy	83.4	35.6	0.37
Lithuania	7.5	8.7	0.11
Latvia	8.1	8.2	0.09
Netherlands	9.3	26.9	0.29
Norway	12.4	8.1	0.09
Poland	7.4	10.7	0.17
Portugal	13.2	18.2	0.08
Sweden	6.5	10.0	0.09
Slovakia	22.9	34.5	0.31
Slovenia	39.8	29.2	0.59
United Kingdom	18.5	38.4	0.28
Mean	**51.7**	**21.9**	**0.31**

[7] http://weppi.gtk.fi/publ/foregsatlas/ForegsData.php

Furthermore, an extensive database is available on soil loss across the EU[8], for various land types and scenarios. Metal losses to water can therefore be calculated by multiplying the soil loss by the metal concentration (Table 7).

[8] https://ec.europa.eu/eurostat/statistics-explained/index.php/Agri-environmental_indicator_-_soil_erosion

Table 7 Cd, Ni and Pb average loss from European soils. Yellow cells indicate use of average data.

			Load (kg day^{-1})		
Country	**Agricultural areas and natural grassland total soil loss (t ha^{-1})**	**Agricultural area (km²)**	**Cd**	**Ni**	**Pb**
Albania	3.2	11740	3.77	536	140
Austria	7	26538	18.66	1,283	1,376
Belgium	1.6	13561	5.16	177	195
Bosnia	3.2	17796	4.81	813	344
Bulgaria	3.3	50303	13.92	2,349	995
Croatia	3.5	14857	4.70	505	280
Cyprus	3.5	1319	0.39	65	28
Czech Republic	2.6	35232	6.42	439	705
Denmark	0.5	26325	0.14	12	15
Estonia	0.5	10042	0.19	12	16
Finland	0.4	22719	0.17	23	14
France	2.3	290202	74.40	4,334	6,637
Germany	1.75	166451	26.80	1,339	2,069
Greece	4.9	52881	58.57	12,131	2,784
Hungary	2.1	53438	5.35	560	422
Iceland	3.2	15551	4.21	710	301
Ireland	0.9	45160	5.69	245	216
Italy	11	128433	142.58	32,263	13,773
Kosovo	3.2	4195	1.13	192	81
Latvia	0.7	19379	0.34	30	30
Lithuania	0.8	29472	0.71	48	56
Luxembourg	3.4	1316	0.38	63	27
Malta	4.7	116	0.05	8	3
Netherlands	0.3	18224	0.43	14	40
N. Macedonia	3.2	12641	3.42	577	244
Norway	3.2	9825	0.77	108	70
Poland	1.5	145396	10.28	444	637
Portugal	3.1	35914	2.54	401	555
Romania	4.2	134137	47.24	7,972	3,376
Serbia	3.2	34869	9.43	1,592	674
Slovakia	3.8	19195	6.24	458	690
Slovenia	14.8	4779	11.50	771	565
Spain	4.6	242019	77.82	7,802	8,212
Sweden	1	30004	0.72	53	82
Switzerland	3.2	15147	7.23	740	484
Turkey	3.2	382390	103.45	17,461	7,393
UK	1.6	173570	21.45	1,403	2,921
EU27			**521**	**73,803**	**43,800**

2.6 Atmospheric deposition of Cd, Ni, Pb onto agricultural soil and inland waters

Concentrations of metals such as cadmium in precipitation decreased significantly between the 1980's and 2000, but have stabilised to a degree since then (Smolders and Six, 2013). Rainfall data are provided Table 8 based on outputs from the European EMEP monitoring data from 2017.[9] Atmospheric deposition of Cd, Ni and Pb to soil were based on multiplying this concentration for each country by the annual rainfall by the total agricultural area and inland water area available via the Eurostat databases (Tables 8 and 9).

Deposition rates for cadmium ranged from a minimum, maximum and mean of 0.07, 0.69 and 0.238 µg Cd l^{-1}. Converting to kg day^{-1} by multiplying by the average country's rainfall and agricultural area then computing the mean deposition as an annual load per hectare provided a mean across Europe of 0.17 35 g Cd ha^{-1} yr^{-1} which was around half of the 0.35 g Cd ha^{-1} yr^{-1} reported by Smolders and Six (2013). This probably reflects improving air quality over the last decade as well as the different assumptions used in the calculations.

[9] https://projects.nilu.no/ccc/reports/cccr3-2019_HM_and_POP_measurements_2017_FINAL.pdf

Table 8 Cd, Ni and Pb loads from atmospheric deposition onto agricultural land.

			Concentration in rainfall (µg L^{-1})[1]			Atmospheric load to agricultural land (Kg d^{-1})		
	Annual rainfall (mm)	**Agricultural area (Km^2)**	**Cd**	**Ni**	**Pb**	**Cd**	**Ni**	**Pb**
Albania	1000	11740	0.02	0.33	0.59	0.77	11	19
Austria	607	26538	0.02	0.33	0.59	1.05	15	26
Belgium	760	13561	0.025	0.2	0.67	0.71	6	19
Bosnia	1008	17796	0.02	0.33	0.59	1.17	16	29
Bulgaria	595	50303	0.02	0.33	0.59	1.95	27	48
Croatia	1021	14857	0.02	0.33	0.59	0.99	14	24
Cyprus	320	1319	0.02	0.33	0.59	0.03	0	1
Czech republic	527	35232	0.0185	0.28	0.58	0.94	14	29
Denmark	736	26325	0.023	0.25	0.65	1.21	13	34
Estonia	660	10042	0.043	0.65	0.55	0.78	12	10
Finland	651	22719	0.013	0.22	0.30	0.53	9	12
France	650	290202	0.04	0.64	0.89	19.54	328	462
Germany	933	166451	0.013	0.21	0.39	5.53	89	167
Greece	371	52881	0.02	0.33	0.59	1.28	18	32
Hungary	563	53438	0.02	0.33	0.59	1.96	27	48
Iceland	798	15551	0.016	0.44	0.42	0.54	15	14
Ireland	731	45160	0.019	0.14	0.42	1.72	13	38
Italy	944	128433	0.02	0.33	0.59	7.91	110	195
Kosovo	700	4195	0.02	0.33	0.59	0.19	3	5
Latvia	636	19379	0.02	0.33	0.59	0.80	11	20
Lithuania	683	29472	0.02	0.33	0.59	1.31	18	32
Luxembourg	875	1316	0.02	0.33	0.59	0.08	1	2
Malta	553	116	0.02	0.33	0.59	0.00	0	0
Netherlands	760	18224	0.0295	0.21	0.76	1.12	8	29
North Macedonia	830	12641	0.02	0.33	0.59	0.68	10	17
Norway	774	9825	0.0113333	0.2	0.38	0.24	4	8
Poland	686	145396	0.027	0.28	0.41	7.37	75	111
Portugal	753	35914	0.02	0.33	0.59	1.76	25	43
Romania	595	134137	0.02	0.33	0.59	5.21	73	128
Serbia	700	34869	0.02	0.33	0.59	1.59	22	39
Slovakia	523	19195	0.02325	0.44	1.36	0.64	12	37
Slovenia	523	4779	0.007	0.12	0.34	0.05	1	2
Spain	456	242019	0.0690	1.22	1.40	20.85	367	422
Sweden	539	30004	0.02025	0.083	0.26	0.90	4	11
Switzerland	1800	15147	0.02	0.33	0.59	1.78	25	44
Turkey	400	382390	0.02	0.33	0.59	9.98	139	246
United Kingdom	754	173570	0.0094	0.096	0.22	3.37	34	80
EU27						**86**	**1289**	**1982**

[1] Yellow cells are median reported values for EU countries

Table 9 Cd, Ni and Pb loads from atmospheric deposition onto inland waters.

			Concentration in rainfall (µg L^{-1})[1]			Atmospheric load to agricultural land (Kg d^{-1})		
	Annual rainfall (mm)	Agricultural area (Km^2)	Cd	Ni	Pb	Cd	Ni	Pb
Albania	1000	1350	0.02	0.33	0.59	0.088	1.23	2.17
Austria	607	914	0.02	0.33	0.59	0.036	0.50	0.89
Belgium	760	242	0.025	0.2	0.67	0.013	0.10	0.34
Bosnia	1008	130	0.02	0.33	0.59	0.009	0.12	0.21
Bulgaria	595	1047	0.02	0.33	0.59	0.041	0.57	1.00
Croatia	1021	566	0.02	0.33	0.59	0.038	0.53	0.93
Cyprus	320	21	0.02	0.33	0.59	0.000	0.01	0.01
Czech republic	527	668	0.0185	0.28	0.58	0.018	0.27	0.55
Denmark	736	756	0.023	0.25	0.65	0.035	0.38	0.99
Estonia	660	2191	0.043	0.65	0.55	0.170	2.57	2.18
Finland	651	53749	0.013	0.22	0.30	1.261	20.76	28.42
France	650	3920	0.04	0.64	0.89	0.264	4.43	6.24
Germany	933	4990	0.013	0.21	0.39	0.166	2.66	4.99
Greece	371	1324	0.02	0.33	0.59	0.032	0.45	0.79
Hungary	563	2618	0.02	0.33	0.59	0.096	1.34	2.37
Iceland	798	8898	0.016	0.44	0.42	0.311	8.55	8.16
Ireland	731	12253	0.019	0.14	0.42	0.466	3.43	10.30
Italy	944	2373	0.02	0.33	0.59	0.146	2.04	3.60
Kosovo	700	49	0.02	0.33	0.59	0.002	0.03	0.05
Latvia	636	2764	0.02	0.33	0.59	0.115	1.60	2.83
Lithuania	683	1753	0.02	0.33	0.59	0.078	1.09	1.93
Luxembourg	875	10	0.02	0.33	0.59	0.001	0.01	0.01
Malta	553	0	0.02	0.33	0.59	0.000	0.00	0.00
Netherlands	760	3579	0.0295	0.21	0.76	0.220	1.53	5.66
North Macedonia	830	330	0.02	0.33	0.59	0.018	0.25	0.44
Norway	774	35083	0.0113333	0.2	0.38	0.843	14.87	28.50
Poland	686	5664	0.027	0.28	0.41	0.287	2.93	4.31
Portugal	753	739	0.02	0.33	0.59	0.036	0.51	0.90
Romania	595	7145	0.02	0.33	0.59	0.277	3.87	6.84
Serbia	700	593	0.02	0.33	0.59	0.027	0.38	0.67
Slovakia	523	341	0.02325	0.44	1.36	0.011	0.21	0.66
Slovenia	523	103	0.007	0.12	0.34	0.001	0.02	0.05
Spain	456	3494	0.0690	1.22	1.40	0.301	5.30	6.08
Sweden	539	66249	0.02025	0.083	0.26	1.980	8.07	24.93
Switzerland	1800	817	0.02	0.33	0.59	0.096	1.34	2.37
Turkey	400	9820	0.02	0.33	0.59	0.256	3.57	6.32
United Kingdom	754	7522	0.0094	0.096	0.22	0.146	1.49	3.45
EU27						**6.1**	**65**	**118**

[1] Yellow cells are median reported values for EU countries

2.7 Total loads of Cd, Ni, Pb to agricultural soils

Summing the different loads to soil provides an overall flux of cadmium, nickel and lead (Tables 10-12). Biocides are a significant source of copper to agricultural soil but there are no known inputs of cadmium, nickel and lead from this source. FYM and atmospheric deposition are significant and in a few cases where a high percentage of biosolids are recycled to land, this too, can be a significant trace metal source. Cadmium impurities in inorganic phosphate fertilisers are a known source to agricultural soils.

Table 10 Total cadmium loads agricultural land (nd = no data).

	Agricultural land	Total FYM	Biosolids	P fertiliser	Atmospheric deposition	Biocides	Total
Country	Km^2	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$
Albania	11740	0.20	0.05	0.36	0.77	0.0	1.4
Austria	26538	2.46	0.25	0.34	1.05	0.0	4.1
Belgium	13561	4.91	0.10	0.13	0.71	0.0	5.8
Bosnia	17796	0.76	0.00	n/d	1.17	0.0	1.9
Bulgaria	50303	1.29	0.10	0.85	1.95	0.0	4.2
Croatia	14857	0.79	0.01	0.40	0.99	0.0	2.2
Cyprus	1319	0.29	0.03	0.06	0.03	0.0	0.4
Czech Republic	35232	1.90	0.42	0.58	0.94	0.0	3.8
Denmark	26325	2.09	0.39	0.53	1.21	0.0	4.2
Estonia	10042	0.39	0.00	0.10	0.78	0.0	1.3
Finland	22719	1.53	0.01	0.28	0.53	0.0	2.4
France	290202	20.10	1.06	4.27	19.54	0.0	45.0
Germany	166451	18.14	1.16	2.33	5.53	0.0	27.2
Greece	52881	2.80	0.11	0.66	1.28	0.0	4.9
Hungary	53438	5.52	0.11	1.31	1.96	0.0	8.9
Iceland	15551	0.09	0.00	0.05	0.54	0.0	0.7
Ireland	45160	2.83	0.25	1.19	1.72	0.0	6.0
Italy	128433	14.29	1.12	3.03	7.91	0.0	26.4
Kosovo	4195	0.44	n/d	n/d	0.19	0.0	0.6
Latvia	19379	0.41	0.03	0.29	0.80	0.0	1.5
Lithuania	29472	1.12	0.07	0.58	1.31	0.0	3.1
Luxembourg	1316	0.13	0.01	0.01	0.08	0.0	0.2
Malta	116	0.04	0.00	0.00	0.00	0.0	0.0
Netherlands	18224	5.00	0.00	0.15	1.12	0.0	6.3
N Macedonia	12641	0.13	0.00	n/d	0.68	0.0	0.8
Norway	9825	0.00	0.35	0.22	0.24	0.0	0.8
Poland	145396	26.78	0.57	3.79	7.37	0.0	38.5
Portugal	35914	3.88	0.01	0.53	1.76	0.0	6.2
Romania	134137	5.62	0.18	2.11	5.21	0.0	13.1
Serbia	34869	2.61	0.00	n/d	1.59	0.0	4.2
Slovakia	19195	0.80	0.00	0.29	0.64	0.0	1.7
Slovenia	4779	0.77	0.00	0.11	0.05	0.0	0.9
Spain	242019	19.54	4.34	4.77	20.85	0.0	49.5
Sweden	30004	1.88	0.17	0.37	0.90	0.0	3.3
Switzerland	15147	1.26	0.00	0.10	1.78	0.0	3.1
Turkey	382390	24.22	48.87	5.83	9.98	0.0	88.9
UK	173570	22.25	4.46	2.10	3.37	0.0	32.2
EU27		**168**	**15.0**	**31.2**	**89.6**	**0.0**	**303**

Table 11 Total nickel loads agricultural land (nd = no data).

	Agricultural land	Total FYM	Biosolids	P fertiliser	Atmospheric deposition	Biocides	Total
Country	Km^2	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$
Albania	11740	1.7	0.7	0.7	10.7	0.0	13.8
Austria	26538	24.5	3.5	0.6	14.7	0.0	43.2
Belgium	13561	47.9	1.5	0.2	5.6	0.0	55.3
Bosnia	17796	7.2	0.0	n/d	16.3	0.0	23.6
Bulgaria	50303	11.7	0.8	1.6	27.2	0.0	41.2
Croatia	14857	7.6	0.1	0.7	13.8	0.0	22.2
Cyprus	1319	2.8	0.1	0.1	0.4	0.0	3.4
Czech Republic	35232	18.7	8.2	1.1	14.2	0.0	42.2
Denmark	26325	24.9	5.3	1.0	13.3	0.0	44.5
Estonia	10042	3.9	0.0	0.2	11.8	0.0	15.9
Finland	22719	14.8	0.3	0.5	8.8	0.0	24.4
France	290202	198.5	17.2	7.8	327.9	0.0	551.4
Germany	166451	182.0	29.0	4.3	88.6	0.0	303.8
Greece	52881	21.9	1.6	1.2	17.8	0.0	42.5
Hungary	53438	49.9	2.0	2.4	27.4	0.0	81.7
Iceland	15551	0.8	0.0	0.1	14.9	0.0	15.9
Ireland	45160	31.3	3.4	2.2	12.7	0.0	49.5
Italy	128433	132.3	57.0	5.5	110.3	0.0	305.2
Kosovo	4195	3.8	n/d	n/d	2.7	0.0	6.5
Latvia	19379	4.1	0.4	0.5	11.2	0.0	16.3
Lithuania	29472	10.7	1.4	1.1	18.3	0.0	31.5
Luxembourg	1316	1.4	0.1	0.0	1.0	0.0	2.6
Malta	116	0.3	0.0	0.0	0.1	0.0	0.4
Netherlands	18224	52.4	0.0	0.3	7.8	0.0	60.5
N Macedonia	12641	0.9	0.0	n/d	9.5	0.0	10.4
Norway	9825	0.0	4.8	0.4	4.2	0.0	9.3
Poland	145396	245.2	7.8	6.9	75.1	0.0	335.0
Portugal	35914	35.7	0.6	1.0	24.6	0.0	61.9
Romania	134137	49.1	2.5	3.9	72.6	0.0	128.1
Serbia	34869	12.2	0.0	n/d	22.2	0.0	34.4
Slovakia	19195	7.5	0.0	0.5	12.0	0.0	20.0
Slovenia	4779	7.4	0.0	0.2	0.8	0.0	8.4
Spain	242019	183.1	62.0	8.7	367.1	0.0	620.9
Sweden	30004	18.3	2.9	0.7	3.7	0.0	25.5
Switzerland	15147	13.3	0.0	0.2	24.8	0.0	38.2
Turkey	382390	228.2	667.7	10.7	139.1	0.0	1045.7
UK	173570	198.3	60.9	3.8	34.4	0.0	297.4
EU27		**1586**	**269**	**57**	**1323**	**0**	**3235**

Table 12 Total lead loads agricultural land (nd = no data).

	Agricultural land	Total FYM	Biosolids	P fertiliser	Atmospheric deposition	Biocides	Total
Country	Km^2	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$	$Kg\ d^{-1}$
Albania	11740	2.9	1.5	0.2	19	0	23.5
Austria	26538	18.0	7.3	0.1	26	0	51.3
Belgium	13561	34.4	7.2	0.1	19	0	60.6
Bosnia	17796	5.2	0.0	n/d	29	0	34.1
Bulgaria	50303	9.8	3.4	0.4	48	0	61.6
Croatia	14857	6.1	0.2	0.2	24	0	30.8
Cyprus	1319	2.0	0.1	0.0	1	0	2.8
Czech Republic	35232	13.5	11.3	0.3	29	0	54.3
Denmark	26325	17.0	11.1	0.2	34	0	62.8
Estonia	10042	2.8	0.0	0.0	10	0	12.8
Finland	22719	10.7	0.1	0.1	12	0	22.9
France	290202	150.4	40.9	1.9	462	0	655
Germany	166451	131.8	42.9	1.0	167	0	342
Greece	52881	24.5	3.2	0.3	32	0	59.6
Hungary	53438	37.3	2.8	0.6	48	0	89.0
Iceland	15551	0.6	0.0	0.0	14	0	14.9
Ireland	45160	26.5	7.0	0.5	38	0	72.0
Italy	128433	103.0	87.3	1.3	195	0	387
Kosovo	4195	2.8	n/d	n/d	5	0	7.5
Latvia	19379	3.1	1.0	0.1	20	0	24.1
Lithuania	29472	8.0	1.2	0.3	32	0	41.8
Luxembourg	1316	1.0	0.2	0.0	2	0	3.1
Malta	116	0.3	0.0	0.0	0	0	0.4
Netherlands	18224	38.1	0.0	0.1	29	0	67.0
N Macedonia	12641	2.5	0.0	n/d	17	0	19.4
Norway	9825	0.0	9.9	0.1	8	0	18.0
Poland	145396	177.9	16.3	1.7	111	0	307
Portugal	35914	28.1	1.0	0.2	43	0	72.8
Romania	134137	45.5	5.3	0.9	128	0	180
Serbia	34869	42.4	0.0	n/d	39	0	81.7
Slovakia	19195	5.8	0.0	0.1	37	0	43.2
Slovenia	4779	5.4	0.0	0.0	2	0	7.7
Spain	242019	146.9	140.5	2.1	422	0	711
Sweden	30004	13.8	4.6	0.2	11	0	29.8
Switzerland	15147	9.7	0.0	0.0	44	0	53.6
Turkey	382390	167.6	1391.9	2.6	246	0	1808
UK	173570	167.6	126.9	0.9	80	0	375
EU27		1229	522	14	2062	0	3826

2.8 Loss of Cd, Ni, Pb from agricultural soils

2.8.1 Metal leached in the dissolved phase

There is a scarcity of data for the quantity of metals leached from soil. Two studies have measured copper loss from agricultural soils, one of which also includes zinc. Sadovnikova et al (1996) undertook a bass balance of copper fluxes into and out of soil including the main sources, uptake into crops or livestock and leaching (Table 13). This was completed for grass and arable crops for sand, sand-calcareous, clay, clay-calcareous, loess and peat soils. A mean of 8.8% loss for Cu was calculated (median=10.8%, n=13, 95% confidence interval=2.95%). A second study (Monteiro et al., 2010) undertook a practical study which measured copper and zinc fluxes across different soil types (silty, loam, clay, sand) in the UK and reported a 9% and 10.1% loss respectively (Table 13).

Table 13 Copper and zinc loss from agricultural soils

	Cu (g $ha^{-1}yr^{-1}$)			Zn (g $ha^{-1}yr^{-1}$)			
Soil type	**Input**	**Leaching**	**Ratio**	**Input**	**Leaching**	**Ratio**	**Reference**
Silty clay	207	5	0.024	1564	61	0.039	1
Clay	218	12	0.055	1605	88	0.055	1
Sand	209	8	0.038	1557	327	0.210	1
Loam	206	4	0.019	1549	77	0.050	1
Clay loam	206	4	0.019	1541	78	0.051	1
Sand	206	5	0.024	1541	110	0.071	1
Sand calcareous	345	59	0.171	1039	377	0.363	1
Clay	256	29	0.113	868	86	0.099	1
Clay calcareous	239	24	0.100	811	43	0.053	1
Grass							2
Sand	240	36	0.15				2
Sand calcareous	193	17	0.088				2
Clay	252	13	0.052				2
Clay calcareous	272	12	0.044				2
Loess	306	24	0.078				2
Peat	213	26	0.122				2
Arable							2
Sand	345	59	0.171				2
Sand calcareous	256	29	0.113				2
Clay	239	24	0.100				2
Clay calcareous	295	17	0.058				2
Loess	335	36	0.107				2
Peat	251	61	0.243				2
Mean			**0.09**			**0.101**	

1 Monteiro et al., 2010. Groenenberg et al 2006.

Data that are available for cadmium, nickel and lead includes soil:water partitioning coefficients (Table 14). With these data it is possible to plot Kp versus loss ratio (Figure 2).

Table 14 Soil partition coefficients

	Kp (l kg^{-1})	Ratio of loss from soil	Reference
Copper	1200	0.090	Sheppard et al 2006
Cadmium	1700	0.09041	Sheppard et al 2006
Nickel	1700	0.09041	Sheppard et al 2006
Lead	18000	0.07861	Sheppard et al 2006
Zinc	150	0.101	Sheppard et al 2006

Fitting a log plot between the two points – accepting the issues associated with plotting between two points, a curve can be used to ratio the soil loss ratio for cadmium, nickel and lead based on their partition coefficients (Table 14). These loss values could then be applied to the total loads applied to generate a loss of metals to water via leaching. The predicted 9% loss of cadmium was significantly lower than the values reported elsewhere using partitioning models suggesting between 40 and 100% of cadmium may be lost via leaching (Smolders and Six, 2013). For the purposes of this exercise, for consistency the values in Table 14 were used.

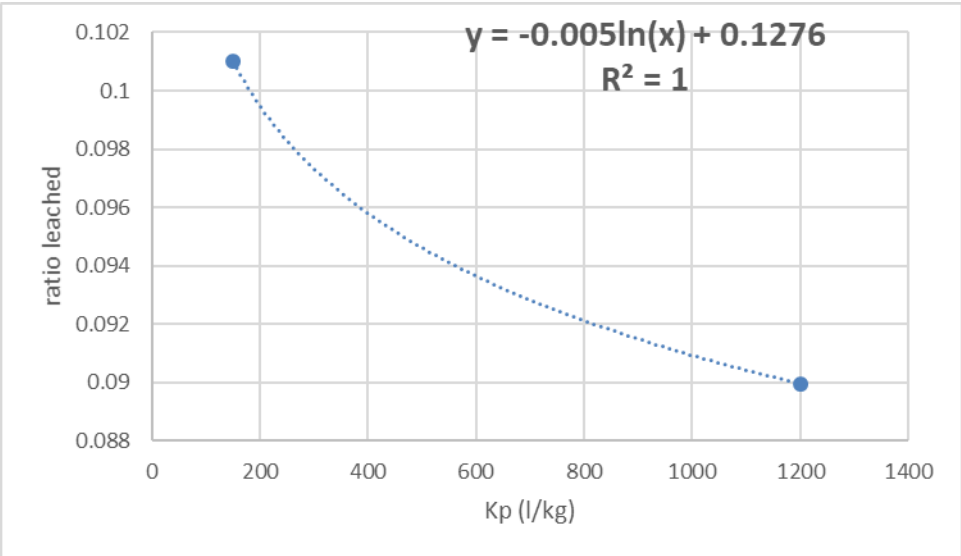

Figure 2 Plot of Kp versus ratio of Cu and Zn leached from soil

2.8.2 Metal transported into water in the particulate/mineral phase

Data are also available for soil loss by rain erosion across Europe[10], broken down by country for different land use and categorised for total, moderate and severe conditions. Total soil loss data were selected (tonnes ha^{-1} $year^{-1}$) and multiplied by background soil cadmium, nickel and lead concentrations and by total agricultural areas within each country to provide a load of metal lost via soil being washed off of land (Table 15).

There are two comparisons for the loss of metals from agricultural land (i) a reported loss of soil multiplied by a background mineral concentration of metals (broadly speaking assumed to be particulate) and (ii) a calculated summed load applied per year from fertilisers and atmospheric deposition, multiplied by a proportion that is leached rather than taken up into crops or adsorbed to the soil matrix (assumed to be mostly dissolved in nature). As can be seen in the figure below, loss of metal associated with the soil is far higher than that leached from inputs, although the leached, assumed more soluble metal may be more bioavailable. The leached volume only comprises 10% for cadmium and 1% for nickel and lead (Figure 3). Cadmium returns a high percentage possibly owing to higher proportion (relatively) in the inorganic P fertilisers which are relatively soluble. Furthermore, background concentrations of cadmium in soils are also much lower than Ni and Pb, compared with concentrations (and therefore loads) in anthropogenic sources such as FYM, biosolids and the aforementioned inorganic fertilisers.

[10] https://ec.europa.eu/eurostat/statistics-explained/index.php/Agri-environmental_indicator_-_soil_erosion

Table 15 Metal loads entering the aquatic environment via soil loss (yellow cells represent means used for calculations)

	Soil loss (kg d^{-1})			Total loss based on source inputs (kg d^{-1})		
	Cd	**Ni**	**Pb**	**Cd**	**Ni**	**Pb**
Albania	3.77	536	140	**0.13**	**1.25**	**1.85**
Austria	18.66	1,283	1,376	**0.37**	**3.91**	**4.03**
Belgium	5.16	177	195	**0.53**	**5.00**	**4.76**
Bosnia	4.81	813	344	**0.17**	**2.13**	**2.68**
Bulgaria	13.92	2,349	995	**0.38**	**3.73**	**4.84**
Croatia	4.70	505	280	**0.20**	**2.01**	**2.42**
Cyprus	0.39	65	28	**0.04**	**0.31**	**0.22**
Czech Republic	6.42	439	705	**0.35**	**3.81**	**4.27**
Denmark	0.14	12	15	**0.38**	**4.02**	**4.94**
Estonia	0.19	12	16	**0.12**	**1.43**	**1.01**
Finland	0.17	23	14	**0.21**	**2.21**	**1.80**
France	74.40	4,334	6,637	**4.07**	**49.9**	**51.5**
Germany	26.80	1,339	2,069	**2.46**	**27.5**	**26.90**
Greece	58.57	12,131	2,784	**0.44**	**3.84**	**4.69**
Hungary	5.35	560	422	**0.80**	**7.38**	**6.99**
Iceland	4.21	710	301	**0.06**	**1.44**	**1.17**
Ireland	5.69	245	216	**0.54**	**4.47**	**5.66**
Italy	142.58	32,263	13,773	**2.38**	**27.6**	**30.4**
Kosovo	1.13	192	81	**0.06**	**0.59**	**0.59**
Latvia	0.34	30	30	**0.14**	**1.47**	**1.89**
Lithuania	0.71	48	56	**0.28**	**2.85**	**3.28**
Luxembourg	0.38	63	27	**0.02**	**0.23**	**0.24**
Malta	0.05	8	3	**0.00**	**0.04**	**0.03**
Netherlands	0.43	14	40	**0.57**	**5.47**	**5.26**
N Macedonia	3.42	577	244	**0.07**	**0.94**	**1.52**
Norway	0.77	108	70	**0.07**	**0.84**	**1.41**
Poland	10.28	444	637	**3.48**	**30.29**	**24.1**
Portugal	2.54	401	555	**0.56**	**5.59**	**5.72**
Romania	47.24	7,972	3,376	**1.19**	**11.6**	**14.2**
Serbia	9.43	1,592	674	**0.38**	**3.11**	**6.42**
Slovakia	6.24	458	690	**0.16**	**1.80**	**3.39**
Slovenia	11.50	771	565	**0.08**	**0.76**	**0.61**
Spain	77.82	7,802	8,212	**4.47**	**56.14**	**55.9**
Sweden	0.72	53	82	**0.30**	**2.30**	**2.34**
Switzerland	7.23	740	484	**0.28**	**3.46**	**4.21**
Turkey	103.45	17,461	7,393	**8.04**	**94.54**	**142**
UK	21.45	1,403	2,921	**2.91**	**26.89**	**29.5**
EU27	**521**	**73,791**	**43,784**	**27.4**	**292**	**301**

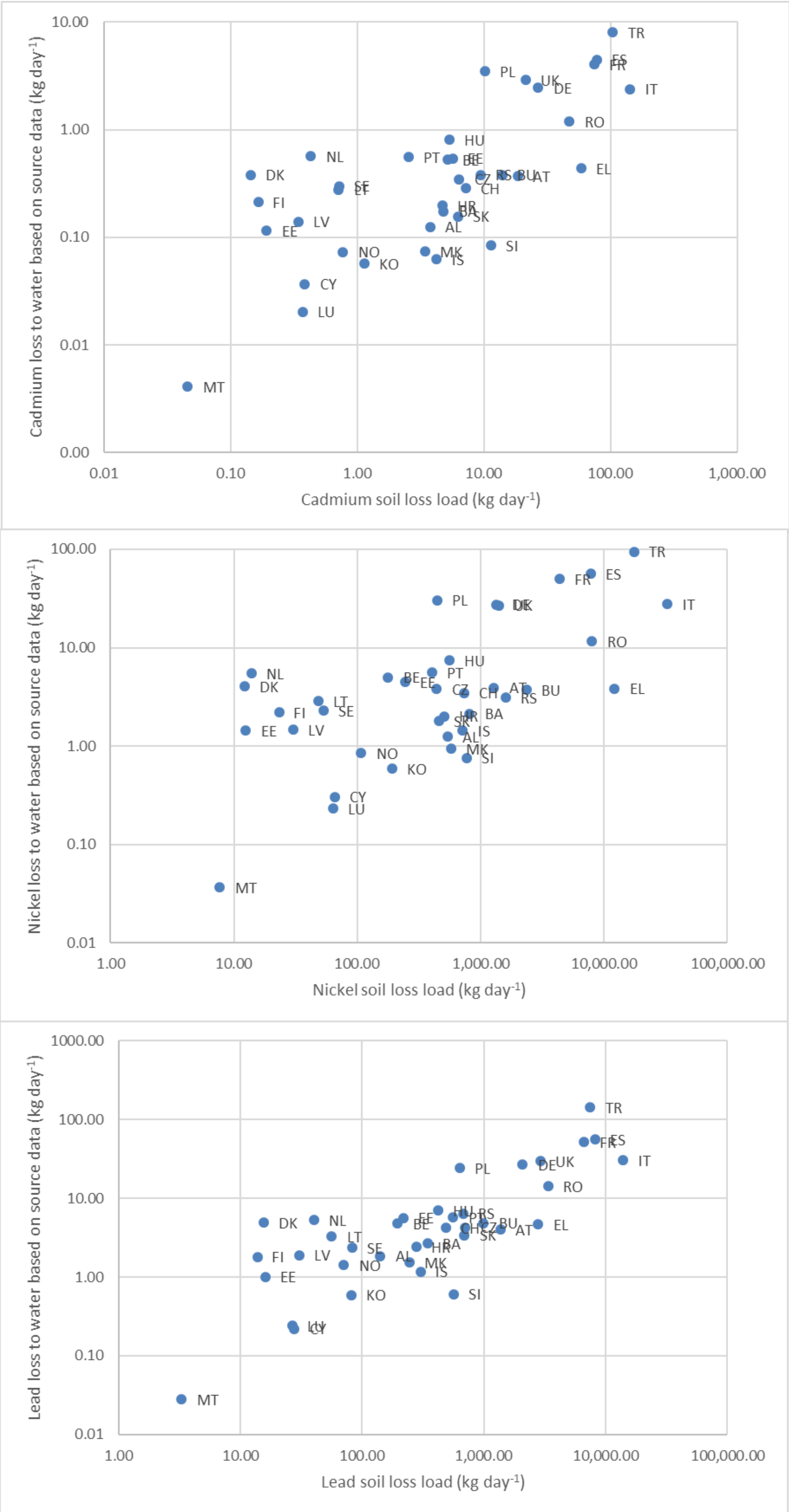

Figure 3 **Comparison between soil loss Cd, Ni and Pb loads with loads to water from the total loads to agricultural soil.**

3 Industrial emissions to water

Data for the emission of cadmium to water for Europe was provided by the European Pollutant Release and Transfer Register (ePRTR)[11]. In 2017, 36 kg/d of cadmium was estimated to be released to water, 22 kg/d by STPs; 663 kg/d of Ni discharged (448 kg/d by STPs) and 211 kg/d of lead (120 kg/d by STPs).

4 Septic tank discharges

Domestic septic tanks are private sewage treatment. Across Europe based on Eurostat data the percentage of people on septic tanks range from 50% for Albania, to ~60% for many Eastern European countries through to high 90's % in many parts of Northern Europe. As an example recent data suggest that 3% of properties in England and Wales have their own treatment facility (although not necessarily in the form of a septic tank), with an estimated 300,000 septic tank systems operating in rural areas.

Septic tanks receive raw sewage, which then separates into solids and liquids. The liquids (effluent) flow out of the system, usually to a soak-away or land drainage system. Some of the solids float to the top of the tank where they form scum, including detergents, cooking fats, and other non-decomposable materials. The heavier solids remain at the bottom of the tank, where anaerobic bacteria produce methane and hydrogen sulphide and decompose them into a sludge layer. The scum and the sludge need to be periodically removed and are commonly taken to a STP for treatment and reuse.

Septic tanks are characterised as:

- Having a high spatial dispersion;
- Are often unknown in number and location;
- Lacking information about operating conditions, maintenance and hence performance;
- Creating difficulty in engagement with "operators" because of their number and relative lack of technical and financial resources;
- Having poor characterisation of the nature of their discharge i.e. where does it go, how much of it is there?; and
- In many cases, providing difficulty or impossibility of monitoring their effluent quality, and hence a difficulty in demonstrating the need to improve.

There is uncertainty about the impact of septic tanks on water quality for four main reasons:

a) A lack of information about the location, number and condition of septic tanks;

[11] https://prtr.eea.europa.eu/#/home

b) The difficulty of, and general lack of, monitoring the effects of septic tanks discharges to surface water and groundwater, there is also a lack of information and associated difficulties of transport pathways and connectivity to received waters not listed;

c) The increasing need, driven by new legislation such as the WFD, to consider a wider range of potential pollutants than has been dealt with in the past. Historical concern has centred principally on BOD and suspended solids, with some account being taken of nutrients at larger treatment works. Under the WFD a wider range of substances, including nutrients, metals and a large number of trace organic substances has to be considered in, to use the parlance of the WFD, assessing the "pressures" that might prejudice the achievement of "good status" for a water body; and,

d) A lack of a clear understanding about the relative importance of septic tanks in relation to other pressures. In rural areas these other pressures stem from sources such as agricultural activity (e.g. use of fertilisers, plant protection products, veterinary pharmaceuticals) and forestry.

e) These issues need to be overcome in order to estimate substance load from septic tanks to surface waters.

4.1 Numbers and locations of septic tanks

There are little data available for the numbers or location of septic tanks across Europe. Individual countries often make estimates based on known mains sewerage data or via mapping exercises. However, based on the Eurostat data for percentage of people on mains sewerage, those on septic tanks can be estimated based on the difference in populations.

4.2 Factors applied to predict inputs to watercourses

Input loads of metals have been derived on a *per capita* basis using data taken from the ETAP influent sewage treatment works concentrations. Eurostat data are available for domestic water use for each European country and this was multiplied by calculated or measured concentrations to derive a per capita load. Table 16 provides a summary of the input loads from domestic sewage based on predicted loads per country relating to summed loads and those estimated from measured integrated domestic wastewater from the literature, applied across all countries. The loads are then amended by a number of key parameters prior to possible discharge into a near-by water body:

- Efficiency of removal of treatment
- Drainage field serviceability
- Slope to water body
- Soil type
- Distance to water body

Table 16 Metal loads entering septic tanks on a per capita basis

	Cadmium (mg capita^{-1} day^{-1})		Nickel (mg capita^{-1} day^{-1})		Lead (mg capita^{-1} day^{-1})[1]	
	Based on calculated load	Based on measured loads	Based on calculated load	Based on measured loads	Based on calculated load	Based on measured loads
Albania	0.172	0.162	1.02	1.37	No data	3.26
Austria	0.092	0.072	0.63	0.61	No data	1.44
Belgium	0.078	0.055	0.53	0.47	No data	1.11
Bosnia	0.073	0.050	0.49	0.42	No data	1.00
Bulgaria	0.081	0.060	0.56	0.51	No data	1.20
Croatia	0.091	0.071	0.61	0.60	No data	1.42
Cyprus	0.177	0.171	1.00	1.45	No data	3.44
Czech Republic	0.074	0.051	0.56	0.43	No data	1.02
Denmark	0.097	0.079	0.63	0.67	No data	1.26
Estonia	0.076	0.054	0.51	0.46	No data	1.09
Finland	0.127	0.069	0.76	0.59	No data	1.39
France	0.108	0.086	0.67	0.73	No data	1.73
Germany	0.083	0.073	0.58	0.62	No data	1.47
Greece	0.227	0.225	1.30	1.91	No data	4.53
Hungary	0.080	0.057	0.55	0.49	No data	1.15
Iceland	0.135	0.120	0.83	1.02	No data	2.42
Ireland	0.086	0.065	0.58	0.55	No data	1.31
Italy	0.122	0.136	0.78	1.15	No data	2.72
Kosovo	0.069	0.045	0.49	0.39	No data	0.91
Latvia	0.097	0.078	0.64	0.66	No data	1.56
Lithuania	0.065	0.041	0.47	0.35	No data	0.82
Luxembourg	0.137	0.123	0.81	1.05	No data	2.48
Malta	0.091	0.070	0.60	0.60	No data	1.41
Netherlands	0.088	0.078	0.57	0.66	No data	1.57
N. Macedonia	0.200	0.195	1.10	1.65	No data	1.35
Norway	0.152	0.106	0.92	0.90	No data	2.14
Poland	0.077	0.053	0.54	0.45	No data	1.07
Portugal	0.114	0.097	0.71	0.82	No data	1.94
Romania	0.067	0.043	0.48	0.37	No data	0.87
Serbia	0.094	0.074	0.61	0.63	No data	1.49
Slovakia	0.092	0.063	0.57	0.53	No data	0.98
Slovenia	0.068	0.043	0.46	0.37	No data	1.27
Spain	0.106	0.088	0.72	0.75	No data	1.77
Sweden	0.124	0.083	0.83	0.70	No data	1.66
Switzerland	0.089	0.102	0.59	0.87	No data	1.96
Turkey	0.086	0.064	0.58	0.55	No data	1.30
UK	0.084	0.076	0.60	0.64	No data	1.52
EU27 mean	**0.101**	**0.081**	**0.65**	**0.69**	**No data**	**1.61**

[1] Lead was not included in the ETAP list so a default of domestic wastewater (median = 12 µg L^{-1}) for the UK was used (Gardner et al., 2013).

The export coefficient database allows each of these parameters to be adjusted, however, it has been possible to generate default values based on a number of factors (Table 17). The parameters are multiplied together to generate an overall transmission factor.

Table 17 Variables and factors used for 'modelling' the transmission of metals through septic tanks (Comber et al., 2013b)

Variable	**Method**	**Comment**	**Scaling[1]**
Removal efficiency	Currently provided for by 'good', 'moderate' and 'poor' providing factors in table below	Varies according to substance	Fractional transmission: **G = 0.8** M = 0.9 P = 1
Distance	The EA database provides a distance from OSWwTS (?) to nearest water body. An exponential 'decay' factor has been applied, taking account of a specified distance where influence is reduced by 50%.	The specified 'half distance' may be varied and is currently arbitrary.	Fractional transmission: G = 0.5 **M = 0.7** P = 1
Slope to watercourse	Fixed via PSYCHIC output 1km grid data for slope based on digital terrain data	Probably as goods as required	Fractional transmission: G = 0.8 **M = 0.9** P = 1
Soil type/permeability	Fixed via PSYCHIC output 1km grid data for soil type	Probably as goods as required	Fractional transmission: G = 0.8 **M = 0.9** P = 1
Drainage field serviceability	Expert judgement		Fractional transmission: G = 0.8 **M = 0.9** P = 1
Overall transmission factor			**0.41**

[1] **bold** = value used for this assessment

The generic derivation of the factor used were taken from Comber et al., (2013). The assumptions used to generate output loads are detailed below:

- The amount of substance reaching the water body is reduced on the basis of the distance of the tank to the nearest water body using an exponential decay formula:

  $$2.718282^{\left(\frac{-\mathrm{LN}(2)}{100}\times \mathrm{distance}\right)}$$

  Where '100' is the number of metres for the emission concentration to be reduced by half and 'distance' is the distance in meters from the tank to the water course.

- The amount of substance reaching the water body is reduced on the basis of the soil type for each 1km^2 grid using:

- Soil permeability data (Comber et al., 2013)
  - If soil =>50% clay then assume no loss (multiply by factor of 1)
  - If soil =>50% sand then assume loss due to infiltration (multiply by factor of 0.8)
  - For other assume nominal loss due to infiltration (multiply by factor of 0.9)

- Slope to water body (Comber et al., 2013)
  - If slope >10 then assume no loss (multiply by factor of 1)
  - If slope > 5 then multiply by factor of 0.9
  - If slope > 2 then multiply by factor of 0.8
  - If slope > 1 then multiply by factor of 0.7
  - If slope < 1 then multiply by factor of 0.6

- If no grid data then worst case is assumed and a default factor of one value is applied (i.e. no reduction)

Combining these factors allowed estimates of losses of elements from septic tanks per country (Table 18).

Table 18 Metal loads entering the aquatic environment via septic tanks

	Cadmium (kg day^{-1})		Nickel (kg day^{-1})		Lead (kg day^{-1})[1]	
	Based on calculated load	Based on measured loads	Based on calculated load	Based on measured loads	Based on calculated load	Based on measured loads
Albania	0.10	0.09	0.60	0.80	No data	1.91
Austria	0.02	0.01	0.11	0.11	No data	0.25
Belgium	0.04	0.03	0.30	0.26	No data	0.62
Bosnia	0.07	0.05	0.45	0.38	No data	0.91
Bulgaria	0.06	0.05	0.44	0.40	No data	0.94
Croatia	0.07	0.05	0.46	0.45	No data	1.07
Cyprus	0.02	0.02	0.11	0.15	No data	0.36
Czech Republic	0.05	0.03	0.35	0.27	No data	0.64
Denmark	0.02	0.01	0.12	0.12	No data	0.23
Estonia	0.01	0.01	0.05	0.05	No data	0.11
Finland	0.04	0.02	0.26	0.20	No data	0.47
France	0.56	0.45	3.48	3.79	No data	8.99
Germany	0.08	0.07	0.56	0.60	No data	1.43
Greece	0.07	0.07	0.38	0.55	No data	1.31
Hungary	0.06	0.04	0.42	0.37	No data	0.88
Iceland	0.00	0.00	0.01	0.01	No data	0.03
Ireland	0.06	0.05	0.41	0.39	No data	0.92
Italy	0.18	0.20	1.16	1.70	No data	4.03
Kosovo	0.02	0.01	0.11	0.09	No data	0.20
Latvia	0.02	0.01	0.11	0.12	No data	0.28
Lithuania	0.03	0.02	0.19	0.14	No data	0.32
Luxembourg	0.00	0.00	0.00	0.00	No data	0.00
Malta	0.00	0.00	0.00	0.00	No data	0.00
Netherlands	0.00	0.00	0.02	0.02	No data	0.06
N. Macedonia	0.05	0.05	0.28	0.43	No data	0.35
Norway	0.05	0.03	0.28	0.28	No data	0.66
Poland	0.32	0.22	2.24	1.87	No data	4.45
Portugal	0.07	0.06	0.44	0.51	No data	1.21
Romania	0.29	0.19	2.09	1.61	No data	3.78
Serbia	0.10	0.08	0.66	0.68	No data	1.62
Slovakia	0.07	0.04	0.41	0.38	No data	0.70
Slovenia	0.02	0.01	0.11	0.09	No data	0.30
Spain	0.06	0.05	0.38	0.40	No data	0.94
Sweden	0.07	0.04	0.45	0.38	No data	0.89
Switzerland	0.00	0.00	0.02	0.03	No data	0.07
Turkey	0.29	0.22	1.96	1.86	No data	4.40
UK	0.06	0.06	0.44	0.47	No data	1.11
EU27 total	**2.35**	**1.83**	**15.6**	**15.5**	**No data**	**36.6**

The data provided in the table above shows that septic tank inputs are lower than agriculture, with reasonable agreement between the summed loads from domestic sources and a mean applied measured load.

CONCLUSIONS

This short screening exercise has been undertaken to estimate diffuse loads of Cd, Ni and Pb to soil and water based on a methodology developed for copper and utilising readily available datasets. It is by no means an exhaustive analysis of data available but designed to provide an indication of the magnitude of inputs from some key known sources which have been put into perspective with data generated in other projects (Table 18).

The breakdown of loads of Cd, Ni and Pb to STPs across Europe is available within the ETAP report and lead sources to water from Arche Consulting (2020).

The data generated from this investigation suggested that soil erosion containing natural background metals within the mineral phase is the single largest load of Cd, Ni and Pb to water, owing to the vast quantities of soil lost from European agricultural land each year. However significant loads of metals ware also associated with farmyard manure, in particular from poultry, atmospheric deposition. Agricultural emissions of these elements is of a similar magnitude to STP discharges.

Atmospheric deposition directly into inland waters was small compared with agricultural land as is a much lower contribution to land area within most countries. However, it equated to around 50% of load to water because only ~10% of land deposition is assumed to be washed off into water compared with 100% of direct deposition onto water.

Septic tanks appear to be only a minor contribution owing to the high proportions of EU populations on mains sewerage and (generally) disconnection between septic tanks and watercourses.

Lead was shown to be an anomaly owing to its specialised use in angling sinkers and in particular, ammunition which was shown to be the second most significant source to water after soil erosion.

Table 18 Summary of estimated diffuse loads of Cd, Ni and Pb compared with point source data generated from other reported

	Loads (kg/d)	to soil Cd	to water Cd	to soil Ni	to water Ni	to soil Pb	to water Pb
	Soil background		521		73803		43800
ePRTR	STP		22		448		120
ETAP	STP		32		668		n/a
	Agriculture Total	271	27	2938	292	3452	301
	Septic tank		2		16		37
	Lost lead sinkers		#N/A		#N/A	#N/A	110
	Lead ammunition		#N/A		#N/A	68008	4781
	Atmospheric deposition on inland water		6.1		65		118
Agriculture break down	Sludge to land	10.5		208		395	
	NPK fertilisers	29		53		13	
	FYM fertilisers cows	11		187		136	
	FYM fertilisers pigs	3.3		84		48	
	FYM fertilisers sheep	2.8		0.96		63	
	FYM fertilisers goats	0.13		0.2		0.2	
	FYM fertilisers poultry	128		1116		815	
	Atmospheric deposition on agricultural land	86		1289		1982	

[1] Data provided from the ETAP report (2021) "Sources and fate of metals and metalloids in wastewater treatment plants. Final report to the ETAP from wca, University of Plymouth and Derac, March, 2021.

[2] Data from Arche Consulting (2020) Pb emission inventory for the environment. Final Report, 3/12/2020. Ferencz N., Eliat M. and Verdonck F.

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