Diffuse sources of heavy metals in the Rhine basin

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Abstract An estimate of diffuse sources of heavy metals (Hg, Cd, Cu, Zn, Pb, Cr, Ni) in the Rhine catchment stressed the urban storm water discharges in the German part and drainage flow in the Dutch part as the most important pathways. Additional sources are erosion and, to a far lesser extent, atmospheric deposition on open water areas. All other pathways were of minor importance. Meanwhile, after reduction of the point sources by between 72–95%, the diffuse sources dominate the total emissions. For several metals the anthropogenic diffuse sources amounted to 40–80%, the point sources to 15–40% and the geogeneous sources to 5–40%. The estimated inputs sufficiently agreed with the loads of the river Rhine. For the estimation, mean values were used for the water masses and the substance concentrations of the different hydrological pathways. It is recommended to undertake further studies on diffuse sources of heavy metals in urban areas and on the possibilities to improve urban storm water management. The calculation methods and the recommendations of the International Commission for the Protection of the Rhine (ICPR) are explained in detail.

Keywords Diffuse sources; drainage; emissions; heavy metals; Rhine; storm waters

Occasion and procedure

Within the framework of the International Commission for the Protection of the Rhine (ICPR), interest has recently focused on the diffuse sources of heavy metals. It became obvious that the operational targets of the ICPR would not be reached even though the point sources were reduced by between 72% and 95% and the concentrations in the river dropped too. Problems exist especially for copper, zinc, mercury, cadmium and lead. For copper and zinc it was established that they are so widely used in construction and installation that the required reduction is not within reach in the foreseeable future.

The ICPR subgroup ÒMethods, foundation and models for the inventory of the diffuse sourcesÓ(established in autumn 1995) selected the sources and pathways of diffuse metal emissions. With the support of an expert team of the German Rhine Protection Commission (DKRR) as well as other Swiss, Dutch and French experts, an intensive literature study was carried out about diffuse metal pathways and their rates and concentrations. Previous investigations of the heavy metal sources in the ICPR only considered the point sources. Inventories made by Behrendt (1993, 1994) and Vink *et al.* (1999) firstly also included the diffuse sources.

Figure 1 shows the considered sources (left) and pathways (right). The metal transports on these different pathways were estimated with the help of statistical data, concentrations etc. averaged over the whole basin. The estimations were carried out in two conceptionally different ways:

¥ *Farmyard seepage, spraydrift, fertiliser and manure runoff.* The masses of used mineral fertiliser and organic manure and their mean metal content were determined. A probable share of losses to waters was assumed with expert judgement.

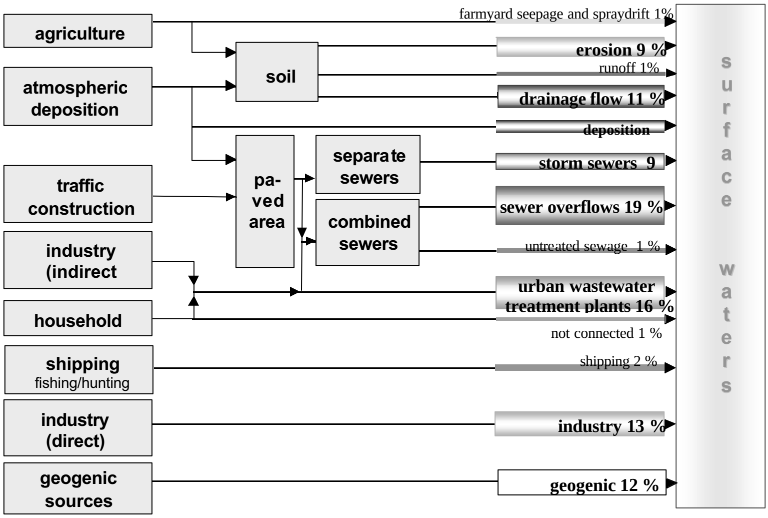

Figure 1 Sources and pathways of heavy metals in the Rhine (Hg, Cd, Cu, Zn, Pb, Cr, Ni – relative shares)

¥ *All other pathways*. The water masses were estimated (in the case of erosion the fine soil masses) and mean metal concentrations chosen from literature.

For comparison, the geogeneous sources were calculated by multiplying the unconsidered water discharge components with the background concentrations. For the discussion, the results of the point sources inventory are also included.

Detailed description of estimations

All the results are summarised in Table 16. Data from the literature were listed in IKSR (1998).

Farmyard seepage and spraydrift, fertiliser and manure runoff

Table 1 Used mass of organic and mineral fertilisers (in 1000 t/a)

Type of fertiliser	Switzerland	France	The Netherlands	Germany	
Cattle manure	812	1410	4285	5730	Dry weight, figures by Eurich-Menden *et al.* (1997) for Germany, converted by livestock statistics of the German Rhine basin
Pig manure	113	18	596	1080	
Poultry manure	23	63	412	105	
P-fertiliser	44	205	123	490	Dry weight, figures for Germany from Statistical Yearbook (1996); 21.7% of German agricultural areas are situated in the Rhine basin; mean nutrient content of fertilisers: 28% N; 20% P_2O_5; 35% K_2O; 50% CaO
N-fertiliser	44	368	967	1385	
K-fertiliser	32	100	62	414	
Ca-fertiliser	45	4	558	795	

Probable share of used masses, which is inserted into freshwaters:

¥ Manure via farmyard seepage and spraydrift (0.2%). For cattle dung in France, this is 5%.

¥ Mineral fertiliser via farmyard seepage and spraydrift (0.01%). In The Netherlands due to many open ditches this is 1.2% (Braun *et al.*, 1991; Hamm, 1991; Werner and Wodsak, 1994: BUWAL, 1996; Coppoolse *et al.*, 1993).

¥ Manure and fertiliser via runoff: Switzerland (1%), France (0.3%), Germany (0.3%) and The Netherlands (0.1%).

Table 2 Mean metal content of fertilisers (mg/kg dry weight)

	Hg	Cd	Cu	Zn	Pb	Cr	Ni	
Cattle manure	0.2	0.3	40	170	12	7	9	
Pig manure	0.4	0.6	370	880	13	11	17	BUWAL (1993);
Poultry manure	0.1	0.3	70	380	9	11	14	BUWAL (1991);
P-fertiliser	0.03	14	27	180	9	710	21	ICK-Landbow (1997);
N-fertiliser	0.01	0.2	8	50	6	4	5	Wilcke and Doehler (1995);
K-fertiliser	0.02	0.2	5	25	6	4	5	FAC (1992)
Ca-fertiliser	0.01	0.2	6	98	13	2	3	

Erosion

Table 3 Mean metal content of fine soil (mg/kg dry weight)

Hg	Cd	Cu	Zn	Pb	Cr	Ni	
0.1	0.3	20	60	27	29	33	BUWAL (1993), LABO (1995)

Mean soil loss of arable land: 2.7 t/ha a (Braun *et al.*, 1991; Hamm, 1991; Werner and Wodsak, 1994)

Table 4

	Switzerland	France	Germany	The Netherlands
Area of arable land (km^2) (local statistics)	1506	657	25700	467
Sedimentation at the foot of the slopes (Braun *et al.*, 1991; Werner and Wodsak, 1994)	60%	90%	90%	99%

Drainage flow

Table 5 Mean metal content of drainage water (µg/l)

Hg	Cd	Cu	Zn	Pb	Cr	Ni	
0.05	2	15	200	15	3	10	Univ. Kiel (1990); Fiedler and Roesler (1993); Koch (1993); Wilcke and Doehler (1995)

Table 6 Discharge of drained area (10^6 m^3/a)

Switzerland	France	The Netherlands	Germany	
244	143	855	313	Drained area: 8% of 39,190 km^2 arable and grassland (local statistics); mean specific drain flow: 100 mm. Univ. Kiel (1990); Traub-Eberhard *et al.* (1994)

Atmospheric deposition

Table 7 Mean deposition rate (mg/m^2*a)

Hg	Cd	Cu	Zn	Pb	Cr	Ni	
0.02	0.2	3.0	25	4.0	0.5	1.5	KIGA (1991); Baart *et al.* (1995); Somhorst and Stolk (1996); Wilcke and Doehler (1995); UBA (1994/95); Brechtel (1989)

Table 8 Freshwater surface area (km^2)

Switzerland	France	The Netherlands	Germany
8.7 (down the great Swiss lakes)	37.7	3020	1250 (Behrendt: GIS-based estimation)

Storm sewers

Table 9 Mean metal concentration in storm sewer outflows (μmg/l)

Hg	Cd	Cu	Zn	Pb	Cr	Ni	
0.4	3	100	400	80	25	45	Muschack (1989); Boller and Häfliger (1996); Heinzmann (1994); Xanthopoulos (1993); Pfeiffer and Hahn (1994); Siecker (1993); Rossi (1995); Krauth

Table 10 Rainwater discharge (10^6 m^3/a)

Switzerland	France	Germany	The Netherlands
39.7	19	263	52

Estimation example (Germany: Fuchs *et al.* (1994), based on local statistics and construction standards): 11% urban area of 99,740 km^2 German Rhine basin area 40% paved area 800 mm/a precipitation 50% specific runoff = 1.755 10^6 m^3/a whole rainwater discharge 15% separate sewer systems = 263 10^6 m^3/a

Sewer and treatment plant overflows

Table 11 Mean metal concentration in sewer and treatment plant overflows (μg/l)

Hg	Cd	Cu	Zn	Pb	Cr	Ni	
0.5	2	60	280	60	10	30	Geiger (1990); Harremöes (1986); Mueller (1995); Schäfer (1995); Fuchs *et al.* (1994); Brombach *et al.* (1992); Schultz *et al.* (1993); Menacher *et al.* (1993); BUWAL (1988)

Table 12 Overflow discharge (10^6 m^3/a)

Switzerland	France	Germany	The Netherlands
45	63	817	128

Estimation example (Germany; Fuchs *et al.* (1994)): Wastewater mass: 130 l per inhabitant and day 34 million inh. 85% combined system + business water: 0.8% business area of 99,740 km^2 German Rhine basin area 0.5 l/s * ha wastewater rate + rainwater: 1.755 10^6 m^3/a whole rainwater discharge 85% combined systems = whole combined systems discharge: 1862 10^6 m^3/a. Overflow statistics: overflows occur on 65 days per year with 45% of a daily discharge = overflow discharges: 817 10^6 m^3/a

Untreated sewers

Table 13 Mean metal concentration in combined sewers: like sewer and treatment plant overflows (see Table 11)

	Switzerland	France	Germany	The Netherlands
Inhabitant share	0	11%	1%	0
Whole combined systems discharge (10^6 m^3/a)	905	31	1862	610

Unconnected inhabitants (scattered dwellings)

Table 14 Mean metal concentration in wastewater: like sewer and treatment plant overflows (see Table 11)

	Switzerland	France	Germany	The Netherlands
Inhabitant share:	3.9%	7%	7%	1.5%

Whole combined systems discharge: see Table 13.
Treatment rate during soil passage etc. 0.8 (own assumption)

Direct uses for shipping

Copper containing antifoulings for sporting boats: Input per boat: 25 g/a (estimated from sales and boat numbers in The Netherlands) boat number: 750,000 (statistics and own estimation).

Zinc electrodes: Use for corrosion protection: 10% of the business ships life time of one electrode (100 g): two years (Bentum, 1994) business ship number in the whole Rhine catchment: 10169 (CCNR, 1997).

Lead containing propeller shaft grease: Used by 80% of the business ships (Bentum, 1994). Input per ship: 1.5 kg/a (CIW/CUTVO, 1997) see above.

Usage of lead and zinc for fishing and hunting was not included. These inputs occur in metal form hardly available for organisms.

Geogeneous sources

Table 15 Geogeneous metal concentration (µg/l)

Hg	Cd	Cu	Zn	Pb	Cr	Ni	
0.01	0.018	1.0	3.5	0.83	2.5	1.1	Compilation by Schudoma (1993)

Yearly mean discharge minus already considered discharge components (industrial and communal wastewater, drainage and storm waters): 22,000 10^6 m^3/a.
For The Netherlands no geogeneous discharge was assumed

Results for the Rhine basin

Table 16 Summary of emissions for the ICPR-Rhine basin (downstream of the Alpine Lakes, see also Figures 1 and 2)

(Usually only two significant figures)	Hg	Cd	Cu	Zn	Pb	Cr	Ni
	(kg/a)		(t/a)				
Anthropogeneous diffuse sources							
Farmyard seepage and spraydrift	20	56	5.9	21	1.3	1.9	1.0
Erosion	100	310	21.0	63	28.0	30.3	34.5
Fertiliser and manure runoff	9	52	3.7	12	0.6	2.2	0.5
Drainage flow	78	3100	23.0	310	23.5	4.7	15.5
Atmospheric deposition	95	950	14.0	120	19.0	2.4	7.1
Storm sewers	150	1100	37.5	150	30.0	9.3	17.0
Sewer and treatment plant overflows	530	2100	63.0	290	63.0	10.5	31.5
Untreated sewers	25	100	3.0	14	3.0	0.5	1.5
Not connected inhabitants	24	90	2.8	13	2.8	0.5	1.4
Shipping	–	–	19.0	63	13.0	–	–
Geogeneous sources	220	390	22.0	76	18.0	55.0	24.0
Point sources							
Urban sewage treatment plants	380	1060	66.0	510	27.0	12.5	29.0
Industry	440	730	48.0	140	16.0	22.0	35.5
Total	**2100**	**10000**	**330**	**1800**	**250**	**150**	**200**

Figure 1 shows the mean relative values for the pathways of the viewed metals, i.e. the percent shares of one pathway in the whole emission of the metal were averaged. The single results for all metals are presented in Figure 2. Figure 3 shows the regional distribution over the Rhine basin countries.

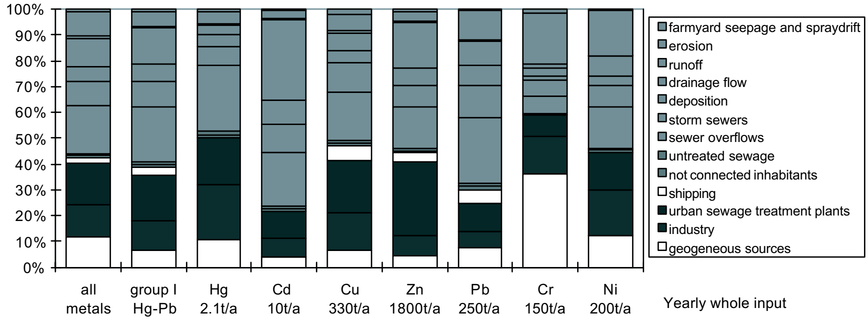

Figure 2 Pathways of heavy metals in the Rhine basin: single results (from bottom to top: White: geogeneous; Black: industry; Straight: urban; Oblique: agriculture)

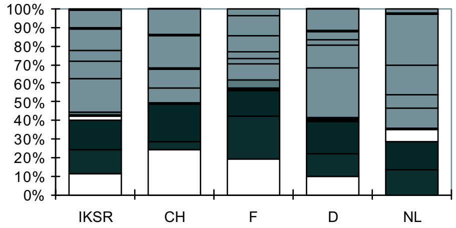

Figure 3 Pathways of heavy metals in the Rhine basin: regional distribution (legend see Figure 2)

Comparison of emissions with river loads

For assessment of the plausibility of the source estimations, the estimated emissions were compared with the river loads at two points:

¥ Weil am Rhein (near Basel): river loads compared with the outflow of the Alpine lakes and the inputs in the Swiss Rhine basin downstream of these lakes added by the emissions in the German basin of Lake Constance.

¥ Bimmen/Lobith (at the GermanÐDutch border): river loads compared with the sum of Swiss lakes output loads and emissions for Switzerland, France and German (see Table 17).

In view of the big uncertainties of both the emissions as well as the loads assessments, the agreement of their figure can be considered as good. That confirmed, that the used methods were valuable for the source apportionment for heavy metals in the framework of the ICPR.

Table 17 Comparison of emissions with river loads

	Hg	**Cd**	**Cu**	**Zn**	**Pb**	**Cr**	**Ni**
	[kg/a]		**[t/a]**				
Weil am Rhein (near Basel)							
Estimated emissions	740	1850	55.0	290	26.5	39.5	80.5
Mean river load	430	1000	51.0	260	34.0	21.0	37.0
Deviation of yearly loads (1991–1996)	230–800	480–2400	42–61	150–300	16.5–50	10–29	26–51.5
Bimmen/Lobith (Germany–Dutch border)							
Estimated emissions	2100	7350	275	1400	205	150	220
Mean river load	2550	5500	490	2000	330	325	300
Deviation of yearly loads (1991–1996)	1700–3500	3500–9700	330–645	1450–3000	180–500	155–530	185–345

Discussion

The results show that in the Rhine basin the diffuse metal sources dominate in comparison with the point sources from communal sewage plants and industry. Anthropogeneous diffuse sources account for about 80% of Cd sources. 75% of Pb, 60% of Cu and Zn, 55% of Ni, 50% of Hg and 40% of Cr sources, respectively. Nearly half of the diffuse sources or 30% of all sources can be attributed to storm waters from streets and roofs of the cities, which reach the river either via separate storm sewers or via overflows of combined systems. Due to the wide use of combined systems the overflows dominate, although the concentrations of most of the considered metals are higher in storm water than in the normal sewage, which is usually the more contaminated water. In the German part the combined system is installed in 85% of all communities. Due to a strong reduction in leaded fuel use and installation of settling equipment for amalgam residues from dentists the urban diffuse sources might already be reduced for lead and mercury.

Usually there are insufficient facilities to keep the metals in the storm water from directly flowing into the rivers. Combined systems overflow usually about 50 times a year, and storm water enters the river together with sewage – both insufficiently cleaned. Only the first, usually extremely polluted part, is held back.

Other important diffuse emission pathways are drainage and erosion with about a third of all diffuse sources. Drainage is especially important in the Dutch Rhine Basin, where plenty of ditches alone account for 40% of all diffuse sources. In the case of Cd, the drain flows dominate in comparison with all other sources. Erosion is of greater importance for nickel and chromium. The deposition from the atmosphere on the open water areas takes a share of about 5%. It is of greater importance for Cd and Hg. Sources of depositions come from the waste incinerating plants, for example, where the metals get to from thermometers or batteries, as well as from industrial or other combustions. Again deposition is of higher importance in The Netherlands due to the bigger portion of freshwater areas there (see Table 8). All other considered pathways (farmyard seepage, spraydrift and runoff during manure spreading, untreated sewage, households which are not connected with public sewage and uses at or in the waters) contribute less than 5% to the diffuse sources.

Geogeneous sources have a share of about 5–10% of all or about 15% of diffuse sources. After the reduction of the point sources of up to 95%, this low geogeneous share shows the great dimension of the remaining problem: the actual emissions are on average 10 fold higher than the natural ones. In the worst cases, Zn and Cd, the emissions are 25 fold higher. Only for chromium is the geogeneous share already significant. But the concentrations of that metal are below the targets already.

Recommendations

Because of the uncertain knowledge about diffuse sources of heavy metals more consolidated examinations are necessary. The used metal amounts and the possible share of inputs into the waters should be investigated for the ranges of use of the heavy metals mercury, cadmium, copper, zinc and lead. The summarised estimates within the Rhine Commission should be deepened by an extended literature study and selective analytical studies on the sources and pathways. The inputs from traffic outside cities should be included in dependence on the traffic densities and the pathways between the streets and the waters, respectively. Geographical information systems may be useful for the summary of knowledge and data, as it is shown nowadays in the investigations on nutrient and pesticide sources in Germany.

The situation in urban storm water treatment calls most urgently for action to minimise diffuse sources. More adequate settling and infiltration basins are needed. They could be created as humid biotopes also improving the residential areas. Moreover the use of heavy metals in construction and traffic should be tested for their necessity and alternatives be found.

In agriculture, diffuse heavy metal inputs are usually caused by drainage flow as well as runoff or erosion events. The drainage flow, especially for Cd, plays an important role. The Cd content of the soils has been enriched with mineral phosphate fertilisers, atmospheric deposition, and other sources. Here only long-term remedy is possible with the help of Cd-poor fertilisers and shut down of tile drainage. All measures against erosion, e.g. mulch seeding or tillage across the slope, are helpful against metal entries too.

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