

**Emission estimates for diffuse sources  
Netherlands Emission Inventory**

**Atmospheric Deposition on  
the Netherlands and the  
Netherlands Continental Shelf**

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Authors: M. Roemer (TNO), J.H. van der Brugh (TNO), N. van Duijnhoven (Deltares)

Participating institutes:

Deltares

Netherlands National Water Board Water Unit

TNO Netherlands Organisation for Applied Scientific Research

PBL Netherlands National Environmental Assessment Agency

# Atmospheric Deposition on the Netherlands and the Netherlands Continental Shelf

## 1 Description of emission source

Atmospheric deposition of substances on water and soil can be described as load of surface water or soil via the atmosphere. In the case of the substances presented here, substance emission first takes place to the atmosphere. Emission to the atmosphere occurs from the known sources in the Netherlands and in Europe that are listed in the Emission Inventory. Once emissions from these sources have entered the atmosphere, the substances are distributed through the atmosphere and end up in the water and on the soil as a result of deposition in wet (precipitation) and dry form.

Figure 1 shows the computational area used in this study.

Deposition is calculated in this study for the Netherlands and the Netherlands Continental Shelf (NCP).

A distinction is drawn between the following emission sources:

E90141102	deposition on surface water
E40012212	deposition in the sewers
E90141103	deposition on the soil

Atmospheric deposition comes under the governmental target sector "Other sources" in the National Emission Inventory.

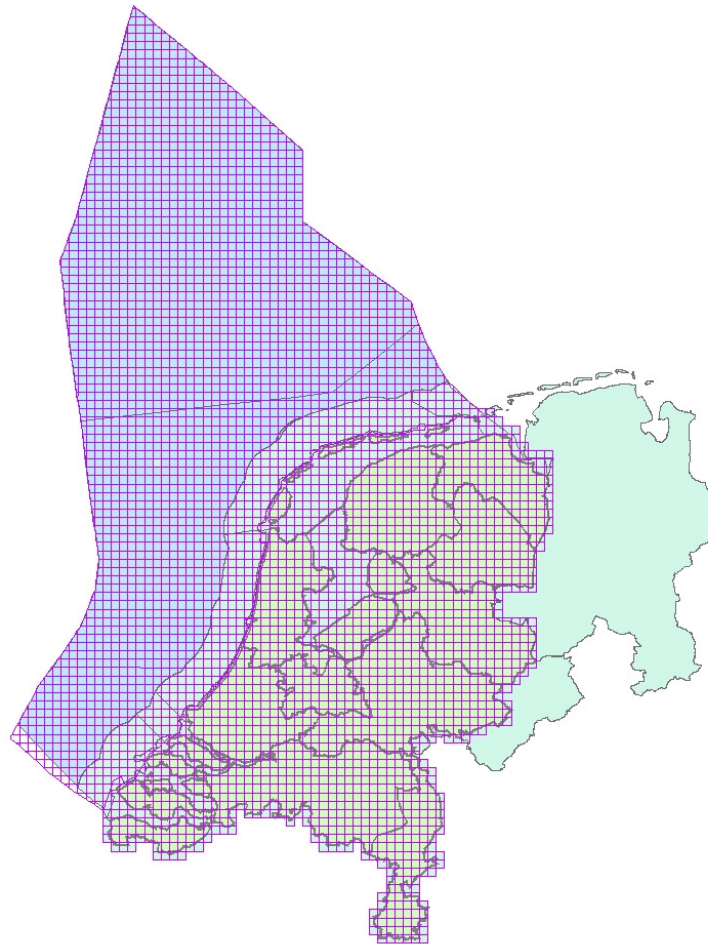


Figure 1: Summary of the computational area used in this study (only the area covered by grid markings).

It should be noted that calculations have been performed for the German part of Dollard Bay, but that this data is not included in the National Emission Inventory (NEI).

## 2 Explanation of calculation method

As described in [1], both the results of measurements and the results of model calculations are taken into account. The calculated deposition of substances at monitoring locations (see also: "comparison with measurements") is compared with the deposition measured at the location in question. The ratio between results of measurements and model calculations for all locations that is determined in this way is used to work out a measurement correction factor applied to the deposition calculated for the entire computational area (figure 1). It is implicit in this that the distribution of sources throughout the area is assumed to be well-established, but that the emission factors are not.

This approach generally works well in the area where results of measurements are available. This applies to earlier calculations performed for surface water in the Netherlands [2], and probably also to calculations of deposition on coastal waters. However, this approach leads to greater uncertainty in the case of calculations on the North Sea and the NCP. This is because there are no results of measurements for this area, and the distribution of sources may be systematically different from that on the land. Uncertainty is therefore much greater here than in the case of estimates for the land area.

### Substances

Table A1 in the appendix contains a summary of the substances examined in this study and those for which calculations have been performed. The key principle followed in selecting substances was the quality of input data, and especially data relating to emissions. The quality of emissions data has a considerable impact on the quality of the figures resulting from this study.

Emission data obtained from the National Emission Inventory or a recent emissions study by Denier van der Gon et al. [3a/b] are regarded as the most reliable source of information. Berdowski et al.'s emissions database [4] is also used for a small number of substances. Substances for which emission data can not be derived from one of the aforementioned sources are not dealt with in this study. The only exception to this is anthracene, where the results are scaled down (using a factor of 0.11) on the basis of figures for fluoranthene.

### Model calculations

The OPS (Operational model for Priority Substances) distribution and deposition model developed by the National Institute for Public Health and the Environment, RIVM, is used to determine the distribution of concentration and deposition of the various substances throughout the Netherlands. The results of the OPS model are then processed in a GIS to work out the level of load (actual net load) on unpaved soil, paved surface area, surface water and the NCP. The maps required for this purpose were supplied by PBL. The 2003 land use map and the 2002 sewer system layer in the National Emission Inventory were used to work out the distribution key for the underlying compartments. GIS processes were used to determine a division between water, paved surfaces and unpaved surfaces for each grid cell and to translate this into figures for the sewers (see also sections 8 and 10).

### The OPS model

The OPS model calculates the concentration of a substance in atmosphere and in precipitation at a determined location (the receptor) as a result of an emission elsewhere [5]. RIVM supplied version 4.1 of the OPS model for the present study. The contribution to concentration and deposition (in both wet and dry form) on the receptor is calculated separately for all sources (maritime shipping on the NCP, the Netherlands, Europe) using what are known as trajectories. These trajectories describe the pathway travelled by atmosphere (incorporating the substance emitted) from the source to the receptor. Local distribution is calculated by means of a mathematical description of a plume, known as the Gaussian plume formula. The spatial resolution capacity of the model is largely determined by the resolution of the emission data used. A resolution capacity of 5x5 km is used in this study. Allocation via the GIS is performed with a spatial resolution of 5x5 km for the NCP and of 1x1 km for the rest of the computational area (the territory of the Netherlands, including freshwater inland waterways and the one-mile coastal zone).

Three sets of input data are needed to perform the model calculations: emission data, meteorological data and substance-specific data. The following input data are used for the two latter parameters:

### **Meteorological data**

Of course, meteorological parameters such as wind direction and wind speed play an important part in these calculations. The calculations reported here were conducted on the basis of meteorological data for 2000 so as to allow for the best possible comparison between the results of modelling and of measurements. The measurements used to scale the calculated outcomes for some substances are those from 2000. The approximation parameter  $Z_0$  varies according to the receptor, and is based on the LGN3 map. LGN3 is the default land use map used by OPS4.1.

### **Substance-specific data**

Substances can be decomposed by reactions with ozone or hydroxyl radicals while being transported through the atmosphere. Losses occur through events such as dry and wet deposition. The OPS model takes account of the influence of these loss processes. The OPS contains substance-specific parameters for individual substances (such as heavy metals and NO<sub>x</sub>). For other substances the data derived from previous reports is entered [6].

Table 1 summarises rates of deposition into water, as used in this study in order to calculate dry deposits into surface waters. It also shows the substance properties used for substances that are not normally included in the OPS.

### **Comparison with measurements**

The calculated wet deposit fluxes for metals and concentrations of NO<sub>x</sub> and NH<sub>3</sub> were compared with the corresponding figures from the RIVM national rainwater measurement network [7]. For some PAHs comparisons were made with figures from the 2000-2001 TNO measurement network [8,9]. The stations are the regional stations of the national measurement network. The figures for 2000 were used for both measurement networks. Both measurement networks have national coverage (see also footnote 3 of table A.1). The model results are scaled to reflect the situation in 2000 by means of trend factors. The philosophy behind this method is that little emission information is available for a number of substances, and that a correction on the basis of the ratio of measurements to modelling results would give a load calculation that properly reflects the measured fluxes. No correction factor was applied for NO<sub>x</sub> and NH<sub>3</sub>, as emissions of these substances are quite well established. The measurement correction factor is defined as the ratio between the measurement result and the modelling result. (If measurements produce higher figures than the modelling results, the measurement correction factor is greater than 1, and the fluxes to the sub-soil are increased by applying the factor).

It should be noted that the measurement correction factor is large for a number of metals (table 1). This may be due to inaccurate measurements. See also chapter 12.

Table 1: Deposition rates above water ( $V_{water}$ ) in  $cm.s^{-1}$  (centimetres per second) for the various substances in this study, the division into gas phase and aerosol phase, and the parameters entered for non-standard substances. The measurement correction factors are shown in the final column.

substances	Vd <sup>1</sup> water $cm.s^{-1}$	ps <sup>2</sup>	substance properties	mol	Mst. corr. factor
Cadmium	0.10	1	OPS	112	3.418
Mercury	0.15	1	OPS	201	1
Lead	0.10	1	OPS	207	3.823
Arsenic	0.10	1	OPS	75	3.527
Chromium	0.10	2	OPS	52	0.58
Copper	0.10	1	OPS	64	2.568
Nickel	0.10	1	OPS as Cu	59	1.018
Selenium	0.10	1	OPS	79	1
Zinc	0.10	1	OPS	65	5.075
Hexachlorobenzene	0.10	0	Rc=21.3;Da=0.01;W=0;Dc=5.93 <sup>E-02</sup>	284	1
Polychlorobiphenyl	0.30	0	Rc=14.3;Da=0.0;W=1000;Dc=5.53 <sup>E-02</sup>	327	5
Hexachlorohexane	0.19	0	Rc=2.33;Da=0.09;W=453000;Dc=5.86 <sup>E-02</sup>	291	1
Benzo(a)pyrene	0.45	0	OPS	252	0.254
Benzo(b)fluoranthene	0.45	0	Rc=66666;Da=1,25;W=100000;Dc=6,300 <sup>E-02</sup>	252	0.958
Benzo(k)fluoranthene	0.45	0	Rc=2,32;Da=0.41;W=340000;Dc=6,300 <sup>E-02</sup>	252	0.985
Indeno(1,2,3-cd)pyrene	0.45	1	Dc=6,019 <sup>E-2</sup>	276	0.337
Benzene	0.01	0	OPS	78	1
Benzo(g,h,i)perylene	0.45	0	Rc=1,47;Da=1,25;W=73000;Dc=6,019 <sup>E-2</sup>	276	0.294
Endosulfan	0.40	0	Rc=166;Da=1,8;W=1.55;Dc=4.957 <sup>E-02</sup>	407	1
Fluoranthene	0.43	0	Rc=9,1;Da=0,43;W=5200;Dc=6,3 <sup>E-2</sup>	252	0.294
polybromide diphenyl ethers	0.01	1	Dc=3.22 <sup>E-02</sup>	964	1
Hexachlorobutadiene	0.10	0	W=0;Dc=6.19 <sup>E-02</sup>	261	10
Pentachlorophenol	0.45	0	Rc=2.05;Da=0.05;W=130000;Dc=6.126 <sup>E-02</sup>	267	0.1
Tetrachloroethene	0.01	0	Rc=250;Da=0.0439;W=0.8;Dc=7,809 <sup>E-2</sup>	164	1
Trichloroethene	0.01	0	Rc=111,1;Da=0,521;W=1,94;Dc=8,77 <sup>E-02</sup>	130	1
NOx	0.2	0	OPS	46	1
NH3	1.0	0	OPS	17	1
Naphthalene	0.14	0	Rc=16666;Da=4,1;W=125;Dc=8,838 <sup>E-2</sup>	128	1

1 deposition rates taken from [6] and the references therein.

2 particle size division; 0: gas; 1: ultra-fine; 2: fine

Rc surface resistance for dry deposition ( $s.cm^{-1}$ ).

Da chemical decomposition in the atmosphere (% per hour)

W ratio between the concentration in precipitation and in atmosphere (non-dimensional). This ratio is significant for part of the wet deposition (rain-out).

Dc diffusion coefficient ( $cm^2.s^{-1}$ ) is equal to  $(mol\ weight)^{-0.5}$ . This coefficient is significant for the description of part of the wet deposition (wash-out).

### 3 Activity rates

The activity rate for deposition on surface water and on soil is in principle emission into atmosphere. This means that the ratio of a particular emission in a particular year to the deposition in that year can be used to calculate deposition in another year on the basis of emission in that other year. The scaling factor for a substance for a particular year is therefore equal to the emission for the year in question divided by the emission in the reference year for which the deposition is being calculated. In this fact sheet, deposition is calculated on the basis of emissions that are divided into three categories:

- emissions from maritime shipping on the NCP (only nickel and NO<sub>x</sub>)
- emissions from other Dutch sources
- emissions from other emission sources in Europe

Table 2: Emissions into atmosphere in (kg/year, unless otherwise specified), and origin and reference year of the data

	Dutch sources		European sources (excluding NL)		Maritime shipping (NCP)	
	emission	origin	emission	origin	emission	origin
Cadmium	2363	ER-2003 <sup>1</sup>	326 tonnes	DvdG-2000 <sup>2</sup>		
Mercury	571	ER-2003	315 tonnes	DvdG-2000		
Lead	41 tonnes	ER-2003	12995 tonnes	DvdG-2000		
Arsenic	976	ER-2003	481 tonnes	DvdG-2000		
Chromium	2990	ER-2003	1960 tonnes	DvdG-2000		
Copper	86 tonnes	ER-2003	4990 tonnes	DvdG-2000 <sup>3</sup>		
Nickel	18.6 tonnes	ER-2003	3730 tonnes	DvdG-2000	28.6 tonnes	Ref.6
Selenium	706	ER-2003	473 tonnes	DvdG-2000		
Zinc	97 tonnes	ER-2003	16881 tonnes	DvdG-2000		
Hexachlorobenzene	598	DvdG-2000	18 tonnes	DvdG-2000		
Polychlorobiphenyl	160	DvdG-2000	133 tonnes	DvdG-2000		
Hexachlorohexane	0	DvdG-2000	254 tonnes	DvdG-2000		
Benzo(a)pyrene	1610	ER-2000	773 tonnes	DvdG-2000		
Benzo(b)fluoranthene	360	ER-2000	842 tonnes	DvdG-2000		
Benzo(k)fluoranthene	759	ER-2000	327 tonnes	DvdG-2000		
Indeno(1,2,3-cd)pyrene	700	ER-2000	613 tonnes	DvdG-2000		
Benzene	2911 tonnes	ER-2000	157 ktonnes	LE-2000 <sup>4</sup>		
Benzo(g,h,i)perylene	1147	ER-2000	10 tonnes	Berdw-1990 <sup>5</sup>		
Endosulfan	0	DvdG-2000	703 tonnes	DvdG-2000		
Fluoranthene	35 tonnes	ER-2005	8231 tonnes	Berdw-1990		
polybromide diphenyl ethers	180	DvdG-2000	8.5 tonnes	DvdG-2000		
Hexachlorobutadiene	0	DvdG-2000	64	DvdG-2000		
Pentachlorophenol	21 tonnes	ER-2005	657 tonnes	DvdG-2000		
Tetrachloroethene	825 tonnes	ER-2005	196 ktonnes	Berdw-1990		
Trichloroethene	31 tonnes	ER-2005	222 ktonnes	Berdw-1990		
NO <sub>x</sub> <sup>6</sup>	371 ktonnes	ER-2005	16430 ktonnes	LE-2000	116 ktonnes	Ref.6
NH <sub>3</sub> <sup>6</sup>	152 ktonnes	ER-2000	6295 ktonnes	LE-2000		
Naphthalene	243 tonnes	ER-2005	15.7 ktonnes	>>benzene <sup>7</sup>		

1: ER: National Emission Inventory, followed by the year for which the information is requested; the 2006 database is used for metals, and the 2007 database for other substances.

2: DvdG: Denier van de Gon et al., (2005). The figures relate to the year 2000 [3a/b]

3: the variant containing a high emission estimate for brake lining is used for foreign copper emissions

4: LE-2000: from the LOTOS-EUROS system [10].

5: Berdw: Berdowski et al., 1997 [4]

6: Emissions are given as NO<sub>x</sub> and NH<sub>3</sub> (this is how the OPS model enters the data); the modelling results (deposition/loads) are given in the unit N.

7: foreign naphthalene emissions are scaled with benzene emissions (10% of benzene)

### Trend factors

The deposition fluxes calculated by the OPS model are scaled with trend factors in order to ensure that the fluxes are those of the reference year (2000). These trend factors are also used to work out the

loads (pollution) for years other than 2000. The trend factors for 1990 to 2020 are given in table 3 (Netherlands), table 4 (Europe) and table 5 (shipping).

Most of the trend factors are based on trends in emissions (see section below).

#### PAH trend factors

Reference [3b] is used for the period from 2000 to 2020 for PAHs. For the period from 1990 to 2000 the same trend factors that feature in the Saltwater Atmospheric Deposition project [6] are used.

#### Heavy metal trend factors

For heavy metals in the period from 1990 to 2006, the trend factors are based on concentrations measured in rainwater derived from the RIVM national rainwater measurement network [7].

The trend factors for metals, apart from those for selenium, are taken from Denier van de Gon [3a].

Emission data from EMEP-Webdab [11] is used for selenium.

Table 3: Trend factors compared to the year 2000 for the Netherlands

	1990	1995	2000	2005	2006	2010	2015	2020
Cadmium	1.342	1.405	1	0.514	0.514	0.447	0.442	0.442
Mercury	2.000	1.989	1	1.000	1.000	0.950	0.940	0.950
Lead	1.263	0.954	1	0.485	0.485	0.272	0.277	0.286
Arsenic	3.350	2.261	1	0.761	0.761	0.616	0.609	0.601
Chromium	1.387	1.002	1	0.822	0.822	0.814	0.871	0.929
Copper	1.028	0.729	1	0.567	0.567	0.527	0.538	0.550
Nickel	1.292	1.477	1	0.859	0.859	0.773	0.756	0.713
Selenium	2.000	1.500	1	0.815	0.778	0.630	0.640	0.650
Zinc	1.450	1.136	1	0.618	0.618	0.568	0.580	0.599
Hexachlorobenzene	0.440	0.720	1	0.720	0.664	0.440	0.450	0.460
Polychlorobiphenyl	0.920	0.960	1	0.600	0.520	0.200	0.130	0.060
Hexachlorohexane	5.200	3.100	1	1.000	1.000	1.000	1.000	1.000
Benzo(a)pyrene	3.400	2.100	1	0.980	0.976	0.960	0.940	0.900
Benzo(b)fluoranthene	3.400	2.100	1	0.985	0.982	0.970	0.940	0.890
Benzo(k)fluoranthene	3.400	2.100	1	0.975	0.970	0.950	0.970	0.980
Indeno(1,2,3-cd)pyrene	3.400	2.100	1	0.970	0.964	0.940	0.890	0.830
Benzene	1.839	1.363	1	0.659	0.659	0.659	0.606	0.552
Benzo(g,h,i)perylene	3.400	2.100	1	0.980	0.976	0.960	0.940	0.900
Endosulfan	1.000	1.000	1	1.000	1.000	1.000	1.000	1.000
Fluoranthene*	3.400	2.100	1	0.800	0.800	0.800	0.800	0.800
polybromide diphenyl ethers	1.000	1.000	1	1.000	1.000	1.000	1.000	1.000
Hexachlorobutadiene	1.000	1.000	1	1.000	1.000	1.000	1.000	1.000
Pentachlorophenol	1.000	1.000	1	1.000	1.000	1.000	1.000	1.000
Tetrachloroethene	2.204	2.625	1	0.946	0.946	0.946	0.946	0.946
Trichloroethene	8.572	9.025	1	0.303	0.303	0.303	0.303	0.303
NOx	1.411	1.193	1	0.884	0.684	0.684	0.562	0.439
NH3	1.638	1.270	1	0.888	0.879	0.842	0.821	0.799
Naphthalene	3.400	2.100	1	0.659	0.659	0.659	0.606	0.552

\*: Trend factors are used for anthracene. Results are scaled on the basis of those for fluoranthene.

### Trend factors for nitrogen, benzene, naphthalene, trichloroethene, tetrachloroethene

For the substances NO<sub>x</sub>, NH<sub>3</sub>, NMVOS (non-methane volatile organic substances), the trend factors for the Netherlands and Europe for the period 1990 to 2010 are taken from EMEP [12]<sup>1</sup>. The Gotheborg Protocol represents the year 2010. The trends for benzene and naphthalene are scaled on the basis of those for NMVOS.

For 2020 an analysis conducted by IIASA<sup>2</sup> of the 'NEC5 variant' (NEC: National Emission Ceilings Directive) in preparing the emission ceiling for 2020 [13] is used. Figures for 2015 are obtained by linear interpolation between 2010 and 2020. As the Netherlands NMVOS emission rate in 2005 had already reached the target for 2010 (according to [12]), it was assumed that the trend factor for benzene and naphthalene for 2010 would be the same as for 2005.

The trend factors for the period 1990 to 2005 for tri- and tetrachloroethene are taken from the National Emission Inventory [14].

Table 4: Trend factors compared to the year 2000 for Europe

	1990	1995	2000	2005	2006	2010	2015	2020
Cadmium	1.342	1.405	1	0.514	0.514	0.447	0.442	0.442
Mercury	2.000	1.989	1	1.000	1.000	0.950	0.940	0.950
Lead	1.263	0.954	1	0.485	0.485	0.272	0.277	0.286
Arsenic	3.350	2.261	1	0.761	0.761	0.616	0.609	0.601
Chromium	1.387	1.002	1	0.822	0.822	0.814	0.871	0.929
Copper	1.028	0.729	1	0.567	0.567	0.527	0.538	0.550
Nickel	1.292	1.477	1	0.859	0.859	0.773	0.756	0.713
Selenium	2.000	1.500	1	0.815	0.778	0.630	0.640	0.650
Zinc	1.450	1.136	1	0.618	0.618	0.568	0.580	0.599
Hexachlorobenzene	0.440	0.720	1	0.720	0.664	0.440	0.450	0.460
Polychlorobiphenyl	0.920	0.960	1	0.600	0.520	0.200	0.130	0.060
Hexachlorohexane	5.200	3.100	1	1.000	1.000	1.000	1.000	1.000
Benzo(a)pyrene	3.400	2.100	1	0.980	0.976	0.960	0.940	0.900
Benzo(b)fluoranthene	3.400	2.100	1	0.985	0.982	0.970	0.940	0.890
Benzo(k)fluoranthene	3.400	2.100	1	0.975	0.970	0.950	0.970	0.980
Indeno(1,2,3-cd)pyrene	3.400	2.100	1	0.970	0.964	0.940	0.890	0.830
Benzene	1.478	1.177	1	0.929	0.659	0.879	0.678	0.477
Benzo(g,h,i)perylene	3.400	2.100	1	0.980	0.976	0.960	0.940	0.900
Endosulfan	1.000	1.000	1	1.000	1.000	1.000	1.000	1.000
Fluoranthene	3.400	2.100	1	0.800	0.800	0.800	0.800	0.800
polybromide diphenyl ethers	1.000	1.000	1	1.000	1.000	1.000	1.000	1.000
Hexachlorobutadiene	1.000	1.000	1	1.000	1.000	1.000	1.000	1.000
Pentachlorophenol	1.000	1.000	1	1.000	1.000	1.000	1.000	1.000
Tetrachloroethene	2.204	2.625	1	0.946	0.946	0.946	0.946	0.946
Trichloroethene	8.572	9.025	1	0.303	0.303	0.303	0.303	0.303
NO <sub>x</sub>	1.311	1.098	1	0.991	0.684	0.894	0.646	0.398
NH <sub>3</sub>	1.258	1.029	1	0.996	0.879	0.975	0.833	0.691
Naphthalene	3.400	2.100	1	0.929	0.659	0.879	0.678	0.477

<sup>1</sup> EMEP: Co-operative programme for monitoring and evaluation of the long-range transmission of atmospheric pollutants in Europe. Gothenburg Protocol: Protocol describing the national emission ceilings for European countries for the substances NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub> and NMVOS

<sup>2</sup> IIASA: International Institute for Applied Systems Analysis. IIASA produces a large number of predictions for the European Commission in the area of energy, emissions and atmospheric pollution.



### Maritime shipping trend factors

It is unclear what future emissions from maritime shipping will be. It is anticipated that the size of this sector will continue to increase. Measures are being prepared to reduce discharge from this sector. In view of the uncertainty over the reduction in emission factors, it has been decided to keep the emissions at the same level for the period after 2005.

Table 5: Trend factors compared to the year 2000 for shipping

	1990	1995	2000	2005	2006	2010	2015	2020
Nickel	1.292	1.477	1	0.859	0.859	0.773	0.756	0.713
NOx	0.753	0.879	1	1.163	1.163	1.163	1.163	1.163

## 4 Emission factors

Emission factors describe emissions per unit for a given parameter. For example: emissions from a car per kilometre travelled. The term 'emission factor' is not appropriate for describing atmospheric deposition. See the section on Activity rates.

## 5 Effects of policy measures

Restricting emissions at an international scale is the most effective way of controlling atmospheric deposition. In the case of NO<sub>x</sub>, NH<sub>3</sub>, NMVOC, heavy metals and persistent organic substances, there is a protocol under the Convention on the Long Range Transboundary Air Pollution (CLRTAP), drawn up under the auspices of the UN-ECE<sup>3</sup>. As part of this protocol a large number of countries have entered into agreements on reporting requirements and emission-reduction obligations. This has been one factor in the decline of emissions of these substances for a number of years. It is expected that emissions will continue to fall in future. The OPS calculations are, wherever possible, based on actual emissions into atmosphere. This means that the effects of measures are taken into account via the trend factors in the final calculations of load caused by atmospheric deposition.

## 6 Time series of emission factors

The method used to estimate atmospheric deposition does not involve emission factors, see section 4.

## 7 Deposition in the Netherlands and the Netherlands Continental Shelf

The OPS model calculates deposition for all compartments separately. Tables 6 to 9 show total deposition (for all catchments combined) for the three different source groups (shipping on the NCP, the Netherlands and Europe). The Netherlands sources exclude the NCP sources, and the European sources exclude the Netherlands and NCP sources. The NCP is offshore water until just before the coast. The one-mile zone does not come under the NCP in this study. It is counted as surface water, as are the IJsselmeer, the Waddenzee and the Zeeuwse waterways. The reference year for the emissions in the calculations is 2000.

Section 8 contains an explanation of how the distribution among the various compartments was devised.

<sup>3</sup> UN-ECE stands for: United Nations Economic Commission for Europe. Emissions are collected in this context under the EMEP programme. (EMEP: Co-operative programme for monitoring and evaluation of the long-range transmission of atmospheric pollutants in Europe).

A summary of the total deposition by atmospheric deposition caused by the three source groups combined for the period from 1990 to 2020 is shown in:

- Table 6 (deposition on NCP)
- Table 7 (deposition on unpaved surfaces)
- Table 8 (deposition on paved surfaces draining into the sewers)
- Table 9 (deposition on saltwater and freshwater surface water including one-mile coastal zone).

*Table 6: Deposition (kg/year, unless otherwise specified) of the NCP due to emissions in the Netherlands and Europe, and by maritime shipping emissions on the NCP*

	1990	1995	2000	2005	2006	2010	2015	2020
Chromium	5,393	3,896	3,887	3,195	3,195	3,163	3,387	3,611
Nickel	19,078	21,804	14,763	12,681	12,681	11,413	11,160	10,525
Copper	65,770	46,604	63,954	36,243	36,243	33,706	34,431	35,156
Zinc	372,484	291,749	256,886	158,639	158,639	145,948	149,121	153,880
Arsenic	20,270	136,82	6,052	4,605	4,605	3,730	3,684	3,638
Cadmium	3,361	3,519	2,505	1,287	1,287	1,120	1,107	1107
Lead	101,776	76,862	80,573	39,095	39,095	21,893	22,284	23,066
Mercury	1,802	1,792	901	901	901	856	847	856
Selenium	2,715	2,040	1,379	1,124	1,073	869	882	896
Benzo(a)pyrene	2,135	1,319	628	615	613	603	590	565
Benzo(b)fluoranthene	8,875	5,481	2,610	2,571	2,563	2,532	2,454	2,323
Benzo(k)fluoranthene	3,311	2,045	974	950	945	925	945	954
Indeno(1,2,3-cd)pyrene	2,013	1,243	592	574	571	556	527	491
Benzo(ghi)pyrene	1,017	628	299	293	292	287	281	269
Hexachlorobenzene	0.726	1.189	1.651	1.189	1.096	0.726	0.743	0.759
Hexachlorohexane	825	492	159	159	159	159	159	159
Polychlorobiphenyl	46.575	48.600	50.625	30.375	26.325	10.125	6.581	3.038
Endosulfan	376	376	376	376	376	376	376	376
Hexachlorobutadiene	2.469	2.469	2.469	2.469	2.469	2.469	2.469	2.469
polybromide diphenyl ethers	27	27	27	27	27	27	27	27
Pentachlorophenol	609	609	609	609	609	609	609	609
Total N (ktonnes N/yr)	60	51	47	46	45	43	35	27
NOx (ktonnes N/year)	33	28	26	26	25	23	18	13
NH3 (ktonnes N/year)	28	22	21	20	20	19	17	15
Benzene	44,890	35,235	29,136	25,701	25,460	24,497	19,390	14,278
Naphthalene	76,031	46,960	22,362	19,626	19,445	18,721	14,855	10,986
Fluoranthene	7,957	4,915	2,340	1,872	1,872	1,872	1,872	1,872
Trichloroethene	8,870	9,338	1,035	313	313	313	313	313
Tetrachloroethene	14,010	16,682	6,356	6,012	6,012	6,012	6,012	6,012
Anthracene	875	541	257	206	206	206	206	206

Table 7: Deposition (kg/year, unless otherwise specified) of unpaved surfaces in the Netherlands due to emissions in the Netherlands and Europe, and by maritime shipping emissions on the NCP

	1990	1995	2000	2005	2006	2010	2015	2020
Chromium	5,303	3,831	3,822	3,142	3,142	3,110	3,330	3,550
Nickel	17,660	20,183	13,665	11,739	11,739	10,565	10,330	9,743
Copper	89,151	63,171	86,689	49,127	49,127	45,688	46,670	47,653
Zinc	509,670	399,199	351,496	217,066	217,066	199,701	204,042	210,554
Arsenic	16,928	11,427	5,054	3,846	3,846	3,115	3,077	3,038
Cadmium	4,811	5,038	3,586	1,843	1,843	1,603	1,585	1,585
Lead	149,914	113,216	118,682	57,586	57,586	32,248	32,824	33,976
Mercury	2,064	2,053	1,032	1,032	1,032	980	970	980
Selenium	2,229	1,682	1,174	957	913	740	751	763
Benzo(a)pyrene	1,771	1,094	521	510	508	500	490	469
Benzo(b)fluoranthene	3,037	1,876	893	880	877	867	840	795
Benzo(k)fluoranthene	3,191	1,971	939	915	910	892	910	920
Indeno(1,2,3-cd)pyrene	1,561	964	459	445	442	431	409	381
Benzo(ghi)pyrene	3,360	2,075	988	968	964	949	929	889
Hexachlorobenzene	5	9	12	9	8	5	6	6
Hexachlorohexane	1,012	603	195	195	195	195	195	195
Polychlorobiphenyl	175	183	191	114	99	38	25	11
Endosulfan	622	622	622	622	622	622	622	622
Hexachlorobutadiene	1.84	1.84	1.84	1.84	1.84	1.84	1.84	1.84
polybromide diphenyl ethers	49.50	49.50	49.50	49.50	49.50	49.50	49.50	49.50
Pentachlorophenol	669	669	669	669	669	669	669	669
Total N (ktonnes N/yr)	141	113	94	87	86	80	72	64
NOx (ktonnes N/year)	39	33	29	28	27	24	18	13
NH3 (ktonnes N/year)	102	80	65	59	59	57	54	51
Benzene	91,276	69,092	53,104	39,769	39,592	38,885	33,454	27,987
Naphthalene	8,163	5,042	2,401	1,768	1,761	1,734	1,505	1,274
Fluoranthene	51,395	31,744	15,116	12,093	12,093	12,093	12,093	12,093
Trichloroethene	410,523	432,187	47,890	14,494	14,494	14,494	14,494	14,494
Tetrachloroethene	569,925	678,610	258,539	244,576	244,576	244,576	244,576	244,576
Anthracene	5,653	3,492	1,663	1,330	1,330	1,330	1,330	1,330

Table 8: Deposition (kg/year, unless otherwise specified) of sewers in the Netherlands due to emissions in the Netherlands and Europe, and by maritime shipping emissions on the NCP

	1990	1995	2000	2005	2006	2010	2015	2020
Chromium	850	614	612	503	503	498	534	569
Nickel	3,347	3,825	2,590	2,225	2,225	2,002	1,958	1,847
Copper	16,085	11,397	15,641	8,863	8,863	8,243	8,420	8,598
Zinc	83,201	65,167	57,380	35,435	35,435	32,600	33,309	34,372
Arsenic	2,626	1,773	784	597	597	483	477	471
Cadmium	775	812	578	297	297	258	255	255
Lead	24,120	18,216	19,095	9,265	9,265	5,188	5,281	5,466
Mercury	319	318	160	160	160	152	150	152
Selenium	355	270	198	161	154	125	126	128
Benzo(a)pyrene	283	175	83	82	81	80	78	75
Benzo(b)fluoranthene	464	287	136	134	134	132	128	121
Benzo(k)fluoranthene	721	445	212	207	206	201	206	208
Indeno(1,2,3-cd)pyrene	247	152	73	70	70	68	65	60
Benzo(ghi)pyrene	649	401	191	187	186	183	179	172
Hexachlorobenzene	0.98	1.61	2.23	1.61	1.48	0.98	1.01	1.03
Hexachlorohexane	185	110	35	35	35	35	35	35
Polychlorobiphenyl	32	34	35	21	18	7	5	2
Endosulfan	105	105	105	105	105	105	105	105
Hexachlorobutadiene	0.28	0.28	0.28	0.28	0.28	0.28	0.28	0.28
polybromide diphenyl ethers	8	8	8	8	8	8	8	8
Pentachlorophenol	189	189	189	189	189	189	189	189
Total N (ktonnes N/yr)	7.25	6.14	5.38	5.06	4.90	4.26	3.32	2.37
NOx (ktonnes N/year)	7.25	6.14	5.38	5.06	4.90	4.26	3.32	2.37
NH3 (ktonnes N/year)*	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Benzene	19,507	14,676	11,133	8,060	8,033	7,926	6,940	5,945
Naphthalene	1,757	1,085	517	369	368	364	321	277
Fluoranthene	13,029	8,048	3,832	3,066	3,066	3,066	3,066	3,066
Trichloroethene	68,675	72,299	8,011	2,425	2,425	2,425	2,425	2,425
Tetrachloroethene	118,324	140,888	53,676	50,777	50,777	50,777	50,777	50,777
Anthracene	1,433	885	422	337	337	337	337	337

\*: NH<sub>3</sub> deposition on paved surfaces is set at 0 because of the alkaline nature of the surface

Table 9: Deposition (kg/year, unless otherwise specified) of surface water (saltwater and freshwater) in the Netherlands due to emissions in the Netherlands and Europe, and by maritime shipping emissions on the NCP

	1990	1995	2000	2005	2006	2010	2015	2020
Chromium	1,203	869	867	713	713	706	756	805
Nickel	4,315	4,932	3,339	2,868	2,868	2,581	2,524	2,381
Copper	16,801	11,905	16,338	9,258	9,258	8,610	8,796	8,981
Zinc	97,484	76,355	67,231	41,518	41,518	38,197	39,027	40,273
Arsenic	3,716	2,508	1,109	844	844	684	675	667
Cadmium	1,039	1,088	775	398	398	346	342	342
Lead	29,120	21,992	23,053	11,186	11,186	6,264	6,376	6,600
Mercury	420	418	210	210	210	200	198	200
Selenium	484	366	259	211	202	163	166	168
Benzo(a)pyrene	591	365	174	170	170	167	163	157
Benzo(b)fluoranthene	2,176	1,344	640	631	629	621	602	570
Benzo(k)fluoranthene	776	479	228	222	221	217	221	224
Indeno(1,2,3-cd)pyrene	532	328	156	152	151	147	139	130
Benzo(ghi)pyrene	309	191	91	89	89	87	85	82
Hexachlorobenzene	0.127	0.208	0.289	0.208	0.192	0.127	0.130	0.133
Hexachlorohexane	116	69	22	22	22	22	22	22
Polychlorobiphenyl	9.03	9.43	9.82	5.89	5.11	1.96	1.28	0.59
Endosulfan	109	109	109	109	109	109	109	109
Hexachlorobutadiene	0.367	0.367	0.367	0.367	0.367	0.367	0.367	0.367
polybromide diphenyl ethers	7	7	7	7	7	7	7	7
Pentachlorophenol	133	133	133	133	133	133	133	133
Total N (ktonnes N/yr)	24.06	19.63	16.90	15.80	15.50	14.30	12.37	10.42
NOx (ktonnes N/year)	10.68	9.08	7.99	7.54	7.31	6.37	5.01	3.65
NH3 (ktonnes N/year)	13.38	10.55	8.91	8.26	8.19	7.93	7.35	6.77
Benzene	17,074	12,989	10,091	7,759	7,718	7,554	6,411	5,262
Naphthalene	30,613	18,908	9,004	6,817	6,784	6,653	5,692	4,725
Fluoranthene	2,860	1,767	841	673	673	673	673	673
Trichloroethene	3,077	3,240	359	109	109	109	109	109
Tetrachloroethene	5,640	6,716	2,559	2,420	2,420	2,420	2,420	2,420
Anthracene	1,433	885	422	337	337	337	337	337

## 8 Release into environmental compartments

Figure 2 shows the final destinations of atmospheric deposition (emissions into water/soil, etc.) to be:

- A unpaved surfaces
- B paved surfaces with no sewer connection
- C paved surfaces with sewer connection
- D water (NCP + salt- and freshwater surface water)

It is assumed that of the load that falls on paved surfaces without a sewer connection, 20% ends up in surface water and 80% on unpaved surfaces (soil). This breakdown means that the final destinations of atmospheric deposition can be defined as:

- 1) unpaved surfaces (see table 7)
- 2) sewers (see table 8)
- 3) water
  - a NCP (see table 6)
  - b fresh- and saltwater surface water (see table 9)

In view of the alkaline nature of paved surfaces, the NH<sub>3</sub> load in sewers is judged to be zero.

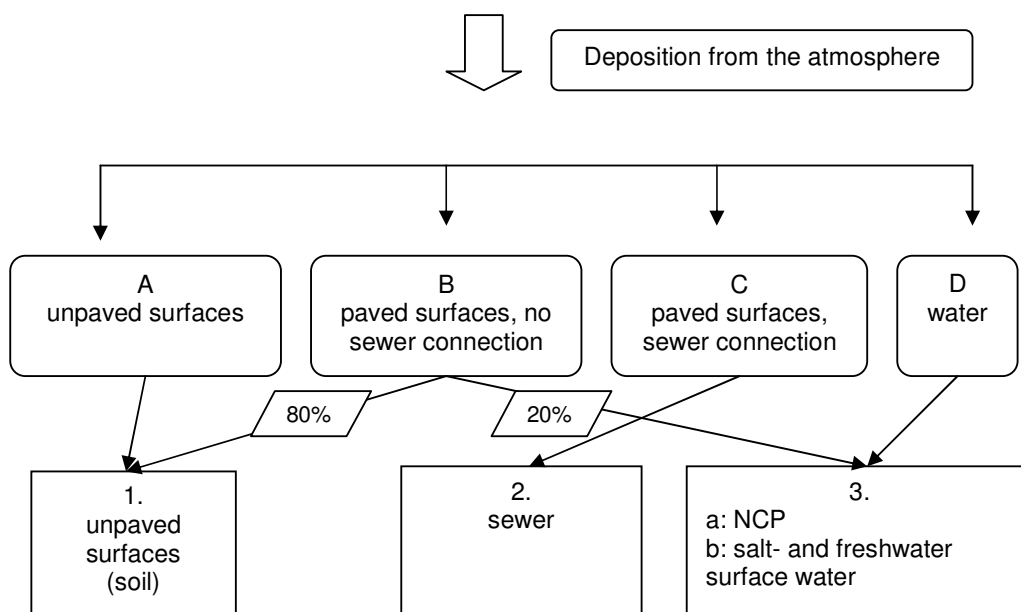


Figure 2: Breakdown of atmospheric deposition between environmental compartments.

The four individual components cover the following area in the Netherlands:

A	unpaved surfaces	2.939 10 <sup>6</sup> ha
B	paved surfaces with no sewer connection	0.101 10 <sup>6</sup> ha
C	paved surfaces with sewer connection	0.326 10 <sup>6</sup> ha
Da	NCP	5.727 10 <sup>6</sup> ha
Db	surface water	0.787 10 <sup>6</sup> ha

## 9 Description of emission pathways to water

Emissions into water arise from direct emissions into surface water and indirectly as a result of emissions from the sewer system, combined sewer overflows, and effluents from waste water treatment plants. The fact sheet " Effluents from waste water treatment plants and sewer systems " [15] describes this in further detail. Emissions take place via rainwater flow (rwf).

## 10 Spatial allocation

Load per substance is worked out using the OPS model on a 5x5 kilometre grid. GIS software is then used to allocate depositions to the individual compartments. Another 5x5 kilometre grid is used for the NCP, while a 1x1 kilometre grid is used for land. Load per substance is recorded directly in the National Emission Inventory for each area or each grid cell. The GIS software uses the 2003 land use map and the 2002 sewer units map. The land use file divides land use in the Netherlands into principal groups which are further subdivided into categories. A threshold ranging from 0.1 to 1 hectare is used for these categories [16].

The spatial allocation of emissions from 2000 (as calculated by OPS) is used as a basis for distribution for all other years. The emission figures for other years are adjusted on the basis of trend factors, but spatial allocation remains unchanged.

## 11 Comments and changes in regard to previous version

### Fluxes

Table B1 shows a comparison of deposition flux on the four different compartments as calculated in this study, and the flux derived from measurements in the TNO measurement network [9] and the RIVM national rainwater measurement network [7]. Comparison with measurements was the principle used to determine deposition fluxes in previous studies. This provides a good match for some substances, but for others the differences are still quite significant. This comparison led us to introduce measurement correction factors, as shown in table 1, for a number of substances (PCB, PCP and HBU). The deposition figures above water have been altered for HCB and PCB (from 0.01 to 0.10 and from 0.07 to 0.30 respectively).

Furthermore, it is clear from table B1 that the fluxes of various substances vary considerably depending on the subsoil. This is due to the physical and chemical properties of the substances in question.

### NCP load

Comparison with earlier studies is difficult because the areas for which the calculations were performed (including catchments) vary. The only exception to this is the study "Factsheet Atmosferische depositie op zoute wateren" [6]. Table C1 contains a summary of the emissions, measurement correction factors and load on the NCP in both studies.

Taking the emissions and the measurement correction factors into account, it can be concluded that for some substances there is a reasonable correlation in respect of their contribution to NCP load. For other substances there are significant differences, largely due to the differences in emissions.

### Substances

The following substances were dealt with in previous studies but are not covered in this study:

- benzo(a)anthracene,
- carbendazim,
- chlorothanolil,
- diuron,
- fenanthrene,
- isoproturon,
- organotin compounds,
- simazine
- tin compounds

These substances have been left out because the emission data for them was judged to be of insufficient quality.

## 12 Accuracy and indicated subjects for improvement

The method used in Emission Inventory publications has been followed as far as possible in classifying the quality of information. It is based on the CORINAIR (CORe emission INventories AIR) methodology<sup>4</sup>, which applies the following quality classifications:

- A: a value based on a large number of measurements from representative sources;
- B: a value based on a number of measurements from some of the sources that are representative of the sector;
- C: a value based on a limited number of measurements, together with estimates based on technical knowledge of the process;
- D: a value based on a small number of measurements, together with estimates based on assumptions;
- E: a value based on a technical calculation on the basis of a number of assumptions.

Element of emission calculation	Reliability classification
Activity rates	C
Emission factors	N/a
Load	C to E
Distribution among compartments	B
Emission pathways to water	C
Spatial allocation	B

The activity rate here relates to emissions of the various substances. The quality of emission data for heavy metals and persistent organic compounds is generally quite poor. However, the emission data used in this study can certainly be described as the best available. It is probably most appropriate to apply category C. A 'load' category has been added as there is no sense in having a category for emission factors. The quality varies from C to E because of the significant differences in the quality of emission figures, combined with the paucity of knowledge as to the deposition of persistent organic compounds. The extent of uncertainty varies depending on the substance and area in question. The correspondence between measurements and the results of calculations varies markedly depending on the substance, and therefore different adjustment factors (used to bring deposition calculations more closely into line with deposition measurements) are applied to individual substances. A significant difference between deposition calculations and deposition measurements has been found for copper in particular. The calculated deposition is five times lower than the figure based on measurements. Uncertainty in respect of deposition calculations for copper is therefore high especially in areas (such as the NCP) for which no measurements are available. No measurements at all are available for some substances (such as selenium and various PAHs). This increases the uncertainty for these substances. Category D is therefore the most appropriate one for this element. The major factor in the difference between deposition calculations and deposition measurements is probably related to the equally significant difference between deposition fluxes obtained from two measuring methods. In 2 (comparison with measurements) it was stated that the results of measurements of metals from the national rainwater network were out of line with the results of measurements of metals measured in aerosol form. The deposition fluxes based on these methods vary by about a factor of 3.

It should be noted that the measurement correction factor is large for a number of metals (table 1). This significant difference suggests that emissions of these substances are severely underestimated. That is surprising in view of the considerable amount of attention paid to emissions of these substances in a recent study [3]. However, it appears that the results of measurements obtained from two different methods are not clear. Measurements from the national rainwater measurement network [7] were used in this study and in previous studies on load in the Netherlands and on the NCP. Since 1990, RIVM has

<sup>4</sup> CORINAIR is a compound word made up of CORINE and AIR. CORINE: CO-ordination d'INformation Environnementale is a software program used to collect environmental data. Part of it, called CORINAIR, deals with emissions into atmosphere.



been measuring aerosol concentrations of four metals (arsenic, cadmium, mercury and zinc). There is a well-established correlation between the annual deposition flux based on concentrations of metals in the atmosphere and that based on concentrations in rainwater. The annual deposition flux based on concentrations in rainwater appears to be about three times higher than that based on concentrations of metals in the atmosphere. It is assumed that the two sets of measurements are not related. We recommend asking RIVM/PBL to clarify the matter and indicate which set of data should be used.

Emissions are distributed among the various compartments via a number of pathways (see chapter 8), and this distribution is determined by the quality of GIS data. This data is relatively robust, and is classed as B. The spatial allocation of emissions is reasonably reliable, as the principal sources are well known and the model is quite reliable in this respect. We therefore assign reliability class B.

The main areas where improvements could be made are:

- achieving consistency between measurements of metals from the rainwater network and those from aerosol measurements
- improving emission data for heavy metals and PAH
- improving estimates of the deposition velocity of gaseous organic compounds
- improving the verification of model calculations by performing measurements. This applies particularly to estimates on the NCP
- if we are to introduce more substances, it is vital that better emissions data is available for these substances and that measurement programmes are conducted.

### 13 Request for reactions

Any questions or comments on this working document should be addressed to: Richard van Hoorn, Centre for Water Management, +31 (0)320 298491, email [richard.van.hoorn@rws.nl](mailto:richard.van.hoorn@rws.nl) or Joost van den Roovaart, Deltares, +31 (0)6 57315874, email [joostvandenroovaart@deltares.nl](mailto:joostvandenroovaart@deltares.nl).

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## Appendix A Summary of substances

Table A1: List of substances

Substances (substance number)	NL NEI <sup>1</sup> DvdG-2000	Abroad DvdG-2000 <sup>2</sup>	Outside <sup>3</sup> atmosphere msmt.	Physical / chemical charact. <sup>4</sup>	Status <sup>5</sup>
<b>'Yes' substances<sup>6</sup></b>					
Cadmium (104)	2003	DvdG-2000	RIVM	Yes	1
Mercury (110)	2003	DvdG-2000	-	Yes	2
Lead (111)	2003	DvdG-2000	RIVM	Yes	1
Arsenic (103)	2003	DvdG-2000	RIVM	Yes	1
Chromium (105)	2003	DvdG-2000	RIVM	Yes	1
Copper (109)	2003	DvdG-2000	RIVM	Yes	1
Nickel (114)	2003	DvdG-2000	RIVM	Yes	1
Selenium (115)	2003	DvdG-2000	-	Yes	2
Zinc (120)	2003	DvdG-2000	RIVM	Yes	1
Hexachlorobenzene	DvdG-2000	DvdG-2000	TNO	Yes	1
Polychlorobiphenyl	DvdG-2000	DvdG-2000	TNO	Yes	1
Hexachlorohexane	DvdG-2000	DvdG-2000	TNO	Yes	1
Benzo(a)pyrene (539)	2000	DvdG-2000	TNO	Yes	1
Benzo(b)fluoranthene (540)	2000	DvdG-2000	TNO	Yes	1
Benzo(k)fluoranthene (541)	2000	DvdG-2000	TNO	Yes	1
Indeno(1,2,3-cd)pyrene (543)	2000	DvdG-2000	TNO	Yes	1
Benzene (515)	2000	Berdw-1990 <sup>7</sup>	RIVM	Yes	3
Benzo(g,h,i)perylene (542)	2000	Berdw-1990	TNO	Yes	3
Endosulfan	DvdG-2000	DvdG-2000	TNO	Yes	1
Fluoranthene (536)	2005	Berdw-1990	TNO	Yes	1
polybromide diphenyl ethers	DvdG-2000	DvdG-2000	-	yes	2
Hexachlorobutadiene	DvdG-2000	DvdG-2000	TNO	yes	1
Pentachlorophenol (679)	2005	DvdG-2000	TNO	yes	1
Tetrachloroethene (619)	2005	Berdw-1990	-	Yes	4
Trichloroethene (621)	2005	Berdw-1990	-	Yes	4
NOx (305)	2005	DvdG-Vissch <sup>8</sup>	RIVM	yes	3
NH3 (301)	2000	DvdG-Vissch	RIVM	yes	3
Total N					
Naphthalene (545)	2005	Via benzene <sup>9</sup>	TNO	yes	3
<b>'No' substances<sup>6</sup></b>					
Di2-ethylhexyl phthalate	2005	-	-	yes	N
Dichloroethane,1,2	2005	??	-	Yes	N
Dichloromethane	2005	??	-	yes	N
Anthracene <sup>10</sup>	NEI <sup>11</sup>	UBA <sup>11</sup>	TNO	Yes	N
Atrazine	RIALT <sup>12</sup>	RIALT	TNO	Yes	N
Chlorfenvinphos	2005	RIALT	TNO	yes	N
Chlorpyrifos	2005	RIALT	TNO	Yes	N
Trichlorobenzenes	2005	??	-	Yes?	N
Trichloromethane	2005	-	-	Yes	N
Tetrachloromethane (620)	2005	??	-	Yes	N
Dichlorvos	RIALT	RIALT	TNO	yes	N
Mecoprop	not	RIALT	-	yes	N
SCCP (short chain chlorinated paraffins)	DvdG-2000	??	-	Yes?	N
Vanadium	??	??	-		N
Dieldrin	-	-	-	partly	N
Diuron	-	-	-	Yes	N
Isodrin	-	-	-		N
Isoproturon	2005	-	-		N
Alachlor	-	-	-	partly	N
Nonylphenol	-	-	-		N
Octylphenols	-	-	-		N
Simazine	RIALT	RIALT	TNO	Yes	N

Substances (substance number)	NL NEI <sup>1</sup> DvdG-2000	abroad DvdG-2000 <sup>2</sup>	Outside <sup>3</sup> atmosphere msmt.	Physical / chemical charact. <sup>4</sup>	Status <sup>5</sup>
Tributyl compounds	-	-	-		N
Trifluralin	-	-	-	Yes	N
Triphenyltin	RIVM	-	-		N
MCPA (2-methyl-4-chlorophenoxyacetic acid)	2005	-	TNO	partly	N
Heptachlor	RIALT	RIALT	TNO	partly	N
Bentazon	2005	RIALT	TNO	yes	N
Chloridazon	2005	RIALT	TNO	yes	N
Dichlorvos	RIALT	RIALT	TNO	yes	N
Dichlorprop	-	-	-		N
Dimethoate	RIALT	RIALT	TNO	yes	N
Pyrazon	RIALT	RIALT	TNO	yes	N
Boron	-	-	-		N
Uranium	-	-	-		N
Propoxur	RIALT	RIALT	TNO	yes	N
Diazinon	RIALT	RIALT	TNO	yes	N
Heptenophos	-	-	-		N
Aldrin	0 <sup>3</sup>	0	TNO	partly	N
Chlordane	0	0	-		N
Chlordecone	0	0	-		N
DDT (Dichlorodiphenyltrichloroethane)	0	0	-		N
Endrin	0	0	0	partly	N
Hexabromobiphenyl	0	0	-	partly	N
Mirex	0	0	-		N
Toxaphene	0	0	-	partly	N
Heptachlor	0	0	-0	partly	N

- 1) If a number appears after the emission, the source is the National Emission Inventory (NEI) or Denier van der Gon et al., [3a/b]; if not, the source is either Berdowski et al., (1995) [4] or RIVM/ALTERRA
- 2) If a number appears after the emission, the source is Denier van de Gon (2005), [3a/b]
- 3) RIVM measurement network, precipitation covering the period 1992-2004/5 in four-weekly periods. Stations: Beek, Biddinghuizen, de Bilt, de Zilk/Leiduin, Eibergen, Gilze-Rijen, Huijbergen, Kollumerwaard, Philippine, Rotterdam, Speulderbos, Valthermond/Witteveen, Vredepeel, Wageningen, Wieringerwerf; the figures for total N and benzene relate to concentrations in the atmosphere; TNO measurement network covers atmospheric samples and precipitation samples in the period 2000-2001 in four-weekly periods; 19 stations throughout the country.
- 4) Partly: figures are available for vapour tension, solubility and molecular mass, but not for deposition rates.
- 5) Status; see table below.
- 6) 'Yes' substances are substances which are reported; 'no' substances are not reported because insufficient information is available or because the data that is available is insufficiently reliable
- 7) Based on Berdowski et al., (1995) [4]
- 8) Denier van der Gon and Visschedijk [10]
- 9) Naphthalene emissions are assumed to be 10% of benzene emissions.
- 10) Anthracene is "promoted" to the 'yes' group. The figures are not calculated, but derived from those for fluoranthene. (anthracene = 0.11x fluoranthene.)
- 11) Information from Hulskotte and Duyzer (2004), [17]
- 12) RIALT: emission data as used in Duyzer and Vonk (2003), [9].

Status table

	1	2	3	4	N*
Emissions in the Netherlands	NEI/DvdG	NEI/DvdG	NEI/DvdG	NEI/DvdG	
Emissions in other countries	DvdG	DvdG	Berd.	Berd.	
Trend information	yes	yes	no	no	
Measurement network	yes	no	yes	no	
Substance characteristics	yes	yes	yes	yes	

\* 'No' substances are those for which the sources for emissions in the Netherlands and abroad are other than the NEI, DvdG or Berdw.

## Appendix B Comparison of fluxes

Table B1 compares the deposition flux of a number of substances to the four different destinations with the deposition flux as shown by measurements. The deposition flux for each unit of time and area varies for the four different destinations. The differences between the destinations in relation to deposition flux are caused by the characteristics of the substances. The deposition flux "direct to water, NCP" is about half that of "direct to water, NL" because the NCP is much further away than surface waters in the Netherlands from Dutch sources in particular.

The deposition flux from measurements corresponds closely to the deposition flux figures resulting from calculations; this is not unexpected given the use of measurement correction factors.

Table B1: Deposition flux (grammes/ha/y) compared with figures from measurements [8,9]

substance name	Destination. Flux (g/ha/year)				Flux (g/ha/year) Measurements
	Direct to soil NL	Paved surface with sewer connection NL	Direct to water NCP	Direct to water NL	
Chromium <sup>1</sup>	1.3004	1.4345	0.6787	1.1025	1.204
Nickel	4.6493	6.0664	2.5775	4.2453	3.853
Copper	34.9256	47.8254	6.9986	22.3991	20.298
Zinc	119.5889	134.3886	44.8512	85.4753	90.900
Arsenic	1.7195	1.8364	1.0566	1.4103	1.442
Cadmium	1.2200	1.3533	0.4373	0.9848	0.881
Lead	40.3790	44.7226	14.0677	29.3094	31.448
Benzo(a)anthracene <sup>2</sup>	0.1772	0.1951	0.1096	0.2211	0.2
Benzo(b)fluoranthene	0.3039	0.3196	0.4557	0.8139	0.48
Benzo(k)fluoranthene	0.3193	0.4965	0.1700	0.2901	0.29
Indeno(1,2,3-cd)pyrene	0.1562	0.1700	0.1034	0.1989	0.18
Benzo(g,h,i)perylene	0.3362	0.4472	0.0522	0.1154	0.19
Hexachlorobenzene	0.004187	0.005233	0.00028	0.00094	0.006
Hexachlorohexane	0.0662	0.0831	0.0277	0.0283	0.023
Polychlorobiphenyl	0.0130	0.0164	0.00018	0.0025	0.05
Endosulfan	0.2115	0.2460	0.0657	0.1386	0.006
Hexachlorobutadiene	0.00063	0.00064	0.00043	0.00047	0.0006
Pentachlorophenol	0.2276	0.4434	0.1063	0.1689	0.14
Naphthalene	0.8169	1.2105	3.9043	11.4473	5.79
Fluoranthene	5.1429	8.9752	0.4086	1.0696	2.97

1: measurements of all heavy metals from the national rainwater measurement network [7]. Flux is twice the wet deposition flux

2: measurements of benzo(a)pyrene and all substances listed thereafter from the TNO measurement network [8, 9]

## Comparison with previous studies

Comparison with the results of earlier studies is difficult because the areas for which the calculations were performed (including catchments) vary. The only exception to this is the study "Atmospheric deposition in saltwater in 2005-2006 [6]." Table C1 contains a summary of the emissions, measurement correction factors and load on the NCP in both studies.

The measurement correction factors are slightly different for each study. The present study includes data from all regional stations spread throughout the Netherlands, while the previous study included only data from three coastal stations. That study was focused on saltwater, not on the Netherlands as a whole.

Deposition rates at sea (table 1) were identical in both studies.

It is also interesting to note that the emissions for the same reference year were different in the two studies. Major differences exist for copper and for domestic emissions of cadmium and lead. Copper emissions have been adjusted in the light of new estimates of the contribution of brake linings. The reason for the significant differences in emission statistics for cadmium and lead is unclear.

*Table C1: Comparison of emissions and NCP load in this study and the Saltwater Atmospheric Deposition study. Reference year 2000*

Substance	Emissions into atmosphere (tonnes/year)		Msmt. correction factor	Load on NCP (kg/year)	
	NEI	abroad		from domestic sources	from foreign sources
<b>This study</b>					
Cadmium	2.5	326	3.418	434	2070
Copper	87.4	4,990	2.658	7,105	56,849
Mercury	0.6	315	1	31	869
Lead	47.2	12,995	3.823	9,185	71,387
Nickel	19.1	3,730	1.018	935	9,618
Zinc	99.4	16,881	5.075	18,061	238,824
NO <sub>x</sub>	420 ktonnes	16,430 ktonnes	1	3.590 tonnes N	19.529 tonnes N
<b>Saltwater study</b>					
Cadmium	1.0	376	1.85	102	2,165
Copper	19.6	2,826	9.46	8,880	65,088
Mercury	0.8	343	1	43	917
Lead	29.2	35,344	1.17	2,010	94,677
Nickel	24.9	4,052	0.63	1,006	9,883
Zinc	94.1	19,409	2.77	11,277	217,896
NO <sub>x</sub>	451 ktonnes	17,820 ktonnes	1	54.78 tonnes N	23.435 tonnes N

Another difference is that the present study was conducted using meteorology data from 2000, while the previous study used long-range average meteorology figures for 1990 to 2000. This difference explains deviations of up to 10 to 20%. Finally, it should be pointed out that comparing emissions alone does not explain everything. Other important factors are the height of the source: the height at which substances enter the atmosphere. Source height is important for NO<sub>x</sub> emission from sources such as power stations. This comparison has not been made.

Taking the emissions and the measurement correction factors into account, it can be concluded that for some substances there is a reasonable correlation in respect of their contribution to NCP load. For other substances there are significant differences, largely due to the differences in emissions.