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# Calculation of Emissions into Rivers in Germany using the MONERIS Model

Nutrients, heavy metals and polycyclic aromatic  
hydrocarbons



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## **Calculation of Emissions into Rivers in Germany using the MONERIS Model**

**Nutrients, heavy metals and polycyclic aromatic  
hydrocarbons**

by

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Dessau-Roßlau, September 2010



This report is dedicated to Dr. Horst Behrendt.

Dr. Behrendt worked at the Leibniz Institute of Freshwater Ecology and Inland Fisheries (IGB) in Berlin. He developed the basics for river basin modelling and realized the model system MONERIS (MODelling Nutrient Emissions in River Systems). He motivated us and many others to do this work and supported us substantially with his great experience.

We mourn the untimely death of Dr. Horst Behrendt, who died unexpectedly in December 2008.



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## Abbreviations

If an abbreviation is used twice, the meaning is clear from the context.

ABAG	German version of the Universal Soil Loss Equation
AD	Atmospheric deposition
AU	Analytical unit
BA	Average long-term soil erosion (soil loss)
BAW	Federal Waterways Engineering and Research Institute, Germany
BfG	Federal Institute of Hydrology, Germany
BGR	Federal Institute for Geosciences and Natural Resources, Germany
BIAS	Systematic deviation/error
BImSchV	German Federal Immission Control Act
BKG	Federal Agency for Cartography and Geodesy, Germany
BMU	Federal Ministry for the Environment, Nature Conservation and Nuclear Safety, Germany
BÜK	Soil Map of Germany
Cd	Cadmium
CLC	Corine Land Cover
CO <sub>2</sub>	Carbon dioxide
CORINE	Coordinated Information on the European Environment
Cr	Chromium
Cu	Copper
DD	Monitoring of direct atmospheric deposition
DIN	Dissolved inorganic nitrogen
DIN	German Institute for Standardization
DON	Dissolved organic nitrogen
DR	Tile drainage
DY	Dry year conditions
E	Emission
EEA	European Environment Agency
EF	Model efficiency according to Nash-Suthcliffe
EMEP	Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe
EnR	Enrichment ratio
EPA	Environmental Protection Agency, USA
EPER	European Pollutant Emission Register
EP-TE	Epoxy-tar substitute
ER	Erosion
EU	European Union
EUROSTAT	Statistical Office of the European Communities
EWA	European Water Agency

FDZ	Research Data Centres of the Federal Statistical Office and the statistical offices of the federal states, Germany
FNE	Flow Net Equation
GDR	German Democratic Republic
GIS	Geographic information system
GK	German Geological Map
GLOWA	Global Change and the Hydrological Cycle
GPCC	Global Precipitation Climatology Centre
GTOPO30	Digital elevation model of the United States Geological Survey
GW	Groundwater
HELCOM	Helsinki Commission
Hg	Mercury
ICM	Integrated coastal management
ICPER	International Commission for the Protection of the Elbe River
ICPOR	International Commission for the Protection of the Odra River
ICPR	International Commission for the Protection of the Rhine
ID	Identifier
IGB	Institute of Freshwater Ecology and Inland Fisheries, Berlin
inh	Inhabitant
INK	International Conference on the Protection of the North Sea
ISI	Fraunhofer Institute for Systems and Innovation Research, Karlsruhe
IWG	Institute for Water and River Basin Management, Karlsruhe
k	Correction factor
LABO	The German Working Group on soil issues of the Federal States and the Federal Government
LAWA	The German Working Group on water issues of the Federal States and the Federal Government
LfU	State Agency for the Environment, Baden Wurttemberg (Germany)
LOQ	Limit of quantification
LT	Long-term conditions
LUWG	State agency for Environment, Water Management and Trade Control, Rhineland Palatinate (Germany)
MONERIS	Modeling Nutrient Emissions into River Systems
MR	Main river
MSC-East	Meteorological Synthesizing Center East, Moscow
MWWTP	Municipal wastewater treatment plant
N	Average catchment precipitation
N	Nitrogen
NASA	National Aeronautics and Space Administration, USA
NE	Not estimated
Ni	Nickel

NO <sub>x</sub>	Nitrogen oxide
OSPAR	Oslo-Paris-Commission
P	Phosphorus
PAH	Polycyclic aromatic hydrocarbons
Pb	Lead
PM <sub>10</sub>	Particulate matter < 10 µm
PM <sub>2.5</sub>	Particulate matter < 2.5µm
PRTR	Pollutant Release and Transfer Register
PT	Total number of inhabitants and population equivalents
Q	Runoff balance
R+D	Research and Development
R <sup>2</sup>	Coefficient of determination
RB	River basin
RBD	River basin district
RIVM	National Institute of Public Health and the Environment (NL)
RMSE	Root mean square deviation
SB	Sub-basin
SDR	Sediment delivery ratio
SED	Sediment delivery
SO <sub>2</sub>	Sulfur dioxide
SR	Surface runoff
SRP	Soluble reactive phosphorus
SRTM	Shuttle Radar Topography Mission
STKP	Coal tar pitch
SU	Sub-unit
SWAT	Soil and Water Assessment Tool
TE	Tar epoxy
TN	Total nitrogen
TP	Total phosphorus
TRIB	Tributary
TVO	German Drinking Water Ordinance
UBA	The Federal Environment Agency, Germany
UN-ECE-CLRTAP	Convention on Long Range Transboundary Air Pollution
US	Urban systems
V	Mean catchment evaporation
VG	Administrative boundaries
WA	Working area
WFD	Water Framework Directive
WHG	German Water Resources Act
WS	Water surface
WY	Wet year conditions
Zn	Zinc





## Summary

This report illustrates the results of two research projects of the German Federal Environmental Agency that ran parallel:

- FKZ 204 24 218: Model-based quantification and internet-based visualisation of emissions into Germany's rivers („Priority substances“)
- FKZ 205 24 204: Development of a management tool on emissions into surface waters within the framework of the international reporting commitment („Nutrients“)

The aim of both projects was a methodological development of the MONERIS model to quantify emissions from point and diffuse sources into Germany's surface waters. The following substances were considered: nitrogen (TN) and phosphorus (TP) as nutrients as well as the 7 heavy metals cadmium (Cd), chrome (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn) as well as polycyclic aromatic hydrocarbons (PAH). Both projects are based on consistent sub-basins and the according basic data as well as homogenous calculation algorithms that are adapted to the specifications of each substance group.

The research encompasses Germany's large river basins as well as their catchment areas outside Germany and in total covers an area of 650,000 km<sup>2</sup>. This was divided into 3456 analytical units (2759 of those in Germany), the average catchment areas being 190 km<sup>2</sup> (135 km<sup>2</sup> in Germany). The modelling was performed in individual annual steps for the period between 1983-2005. For the evaluation of the temporal trends the data was aggregated for the periods 1983-1987 („1985“), 1993-1997 („1995“), 1998-2002 („2000“) and 2003-2005 („2005“) to soften the impact of hydrological influences.

All input data was collected and preprocessed with the highest spatial and temporal resolution possible based on the detailed topology. This especially concerns the point sources of communal wastewater treatment plants, mining activities and direct industrial discharges, the use of statistical microdata on wastewater discharge and its treatment, population density, the residence time of groundwater, the calculation of soil erosion based on a de-tailed elevation model, after setting a standard procedure for the complete area as well as river discharge and quality data to calibrate the runoff and test the plausibility of the calculated emissions.

The basic data and model results for all sub-basins, years and substance groups of both projects were merged into one database. Additionally, a web-based graphical user interface was developed to visualise the emissions for any area aggregation can be visualised.

The completion of both R+D-projects „Nutrients“ and „Priority substances“ delivered for the first time ever homogenous instruments that can identify the most important sources and contamination hotspots for different relevant substance groups in larger river basins which can then serve as a basis for further analyses to achieve efficient measures to reduce pollution. Apart from further developing the quantification approaches both projects significantly contributed towards improving the database, which now has a much higher spatial and temporal resolution. According to the detailed topology the emissions are available for each individual sub-basin. However, not all local characteristics of smaller river basins could be integrated adequately when quantifying the emissions due to an incomplete data basis as well as the scale that had to be covered. Furthermore, emissions from sub-basins are subject to a

much higher temporal variability, which cannot be adequately retrieved with the help of balance equations for every single year. Accordingly, small areas have a higher deviation between the calculated emissions and the balanced river loads. The model results for small sub-basins are therefore not to be seen as absolute values and predominantly serve as a visualisation of the regional distribution of emissions. These restrictions even more apply to heavy metals and PAH as opposed to nutrients there is only a limited amount of valid measurement data in an adequate resolution for these substances. Pollutants are measured to monitor the compliance to threshold values. In the process some analysis methods use limits of quantification that are too high which is why this data is useless for calculating emissions and immissions.

The target-oriented monitoring programmes have to be adapted to current issues under defined and comparable conditions, allowing the monitoring results to be transferred. These programmes are essential for monitoring the effectiveness of the measures taken as well as for emission calculations in medium-sized to small sub-basins.

# 1 Introduction

## 1.1 Background and aim

The aim of this project was the methodological development of the quantification of heavy metals and other priority substances from point and diffuse sources into Germany's surface waters. The approach was to be done river basin by river basin, in methodological accordance to the international requirements of the survey for the EU-Water Framework Directive as well as of further international marine and river basin conventions (OSPAR, 1998, HELCOM, 1992, ICPR, 1999a). A further UBA project was carried out parallel to this project called „Development of a management tool for emissions into surface waters within the framework of the international reporting commitment“ (FKZ 205 24 204) which dealt with the nutrient emissions into surface waters. Both projects are based on the same analytical units and database as well as the homogenous calculation algorithms adapted to the substance specifications. Therefore, the methods, database and results of both projects are described as one in this report.

The model system MONERIS (**M**odelling **N**utrient **E**missions into **R**iver **S**ystems) was developed by BEHRENDT ET AL. (1999, 2002a) to quantify the nutrient emissions into Germany's surface waters. The model was extended by FUCHS ET AL. (2002) to be able to quantify heavy metal and lindane emissions. For this the model was appropriately adapted and a first input database was compiled.

A central task of both research projects was to model the emissions for Germany's river basins on the basis of a topology conform to the catchment areas declared by the federal states within the framework of the WRRL-report. Figure 1 depicts the complete area as well as the river basins and coordinating areas. The average area size for modelling was notably reduced compared to earlier projects (approx. 1,000 km<sup>2</sup>, comp. BEHRENDT ET AL., 1999, 2002a and FUCHS ET AL., 2002) and now lies around 135 km<sup>2</sup> in Germany. Due to this detailed topology all input data was collected and preprocessed with the highest spatial resolution possible.

Apart from further developing the method and database another aim was to use the model MONERIS for a longer period on an annual basis for the first time. The calculations were made in individual steps for each year between 1983 and 2005. For this task the specific data was collected and accordingly deduced for every year. In addition three synthetic years were generated to represent different hydrological situations:

- long-term average (long-term = LT),
- long-term minimum (dry situation: dry year = DY) and
- long-term maximum (wet situation: wet year = WY).

The current anthropological input data (2005) was used for the three synthetic years. Synthetic databases were produced based on the whole time series for the hydrological input parameter precipitation and runoff. For the long-term average the average precipitation and the average area runoff over all years were used for the calculation. For the long-term minimum and the long-term maximum the absolute minimum/maximum of the annual area runoff

and the according year precipitation values were chosen. The three synthetic years LT, DY, WY were used to calculate the scenarios to be able to estimate not just the impact of the measures taken but also the effect of hydrology on emissions and loads in general.

The emissions over the observation period were aggregated to longer periods to rate the emission development, as trends over time can only be derived from average hydrological situation. Because of the periodicity of meteorological and hydrological variables a period of 5 years seemed adequate. According to the procedure in the past projects the periods 1983-1987 („1985“), 1993-1997 („1995“), 1998-2002 („2000“) and 2003-2005 („2005“) were chosen. A complete processing of the period “2005” (2003-2008) was not possible as the required input data (statistical data, administrative borders, model calculations, environment monitoring) needed a lot of editing by the according authorities and was therefore slowed down (approx. 1-3 years). The period 1988-1992 wasn't disclosed as the database was strongly affected by Germany's reunification and is tainted with large uncertainties.

The database and model results for all analytical units, years and substance groups from both projects were merged into one database. Additionally, a web-based graphical user interface was developed with which the emissions for any area aggregation can be visualised.

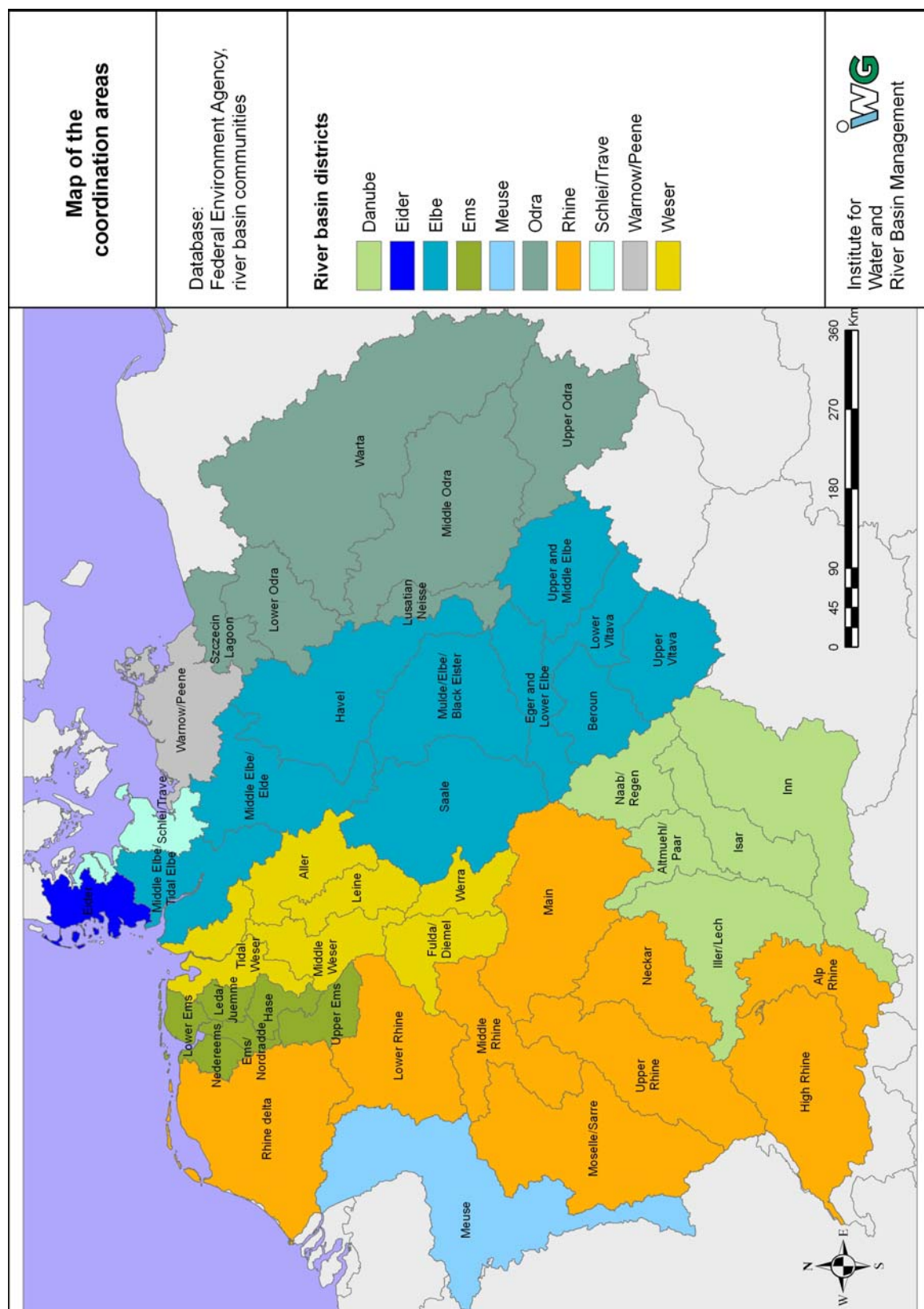
## **1.2 The structure of the report**

The „Priority substances“ as in the research project FKZ 204 24 218 and „Nutrients“ as in FKZ 205 24 204 are two very different substance groups and are therefore divided in this report as follows:

Chapter 2 gives all the information on the subdivision of the areas and the runoff model. This is followed by a description of the method used to quantify the emissions from point sources and diffuse pathways.

In chapter 3 the „general input data“ such as land use, population, geology, pedology and data on urban drainage systems are described. Chapter 4 charts all substance specific input data sorted by the substance groups “nutrients”, “heavy metals” and “polycyclic aromatic hydrocarbons” (PAH). An elaborate preprocessing of the input data is needed to regionalise and consider the temporal variability of the input data. The method as well as the results of the preprocessing are described in chapters 3 and 4, too.

The results are also presented individually for “nutrients”, “heavy metals” and “PAH” in chapter 5. In addition, all results for the analytical units are available in the database configured within the framework of the projects and can be visualised in a graphical user interface.



## 2 The method

To quantify the emissions it was distinguished between point source and diffuse pathways. Point sources are defined by being discrete, having distinct locations and quasi-continuous discharge, e.g. the discharge of municipal wastewater treatment plants and industrial plants. The emissions from diffuse sources influence different pathways and are discharged via different runoff components into the surface waters (see Figure 2). A differentiation of the runoff components is necessary as substance concentrations as well as the underlying processes differ significantly (BEHRENDT ET AL., 1999). Apart from anthropogenic sources which are generally the main source of pollution, diffuse emissions from natural geogenous sources do occur. A schematic summary of the considered emission sources and pathways is given in Figure 2.

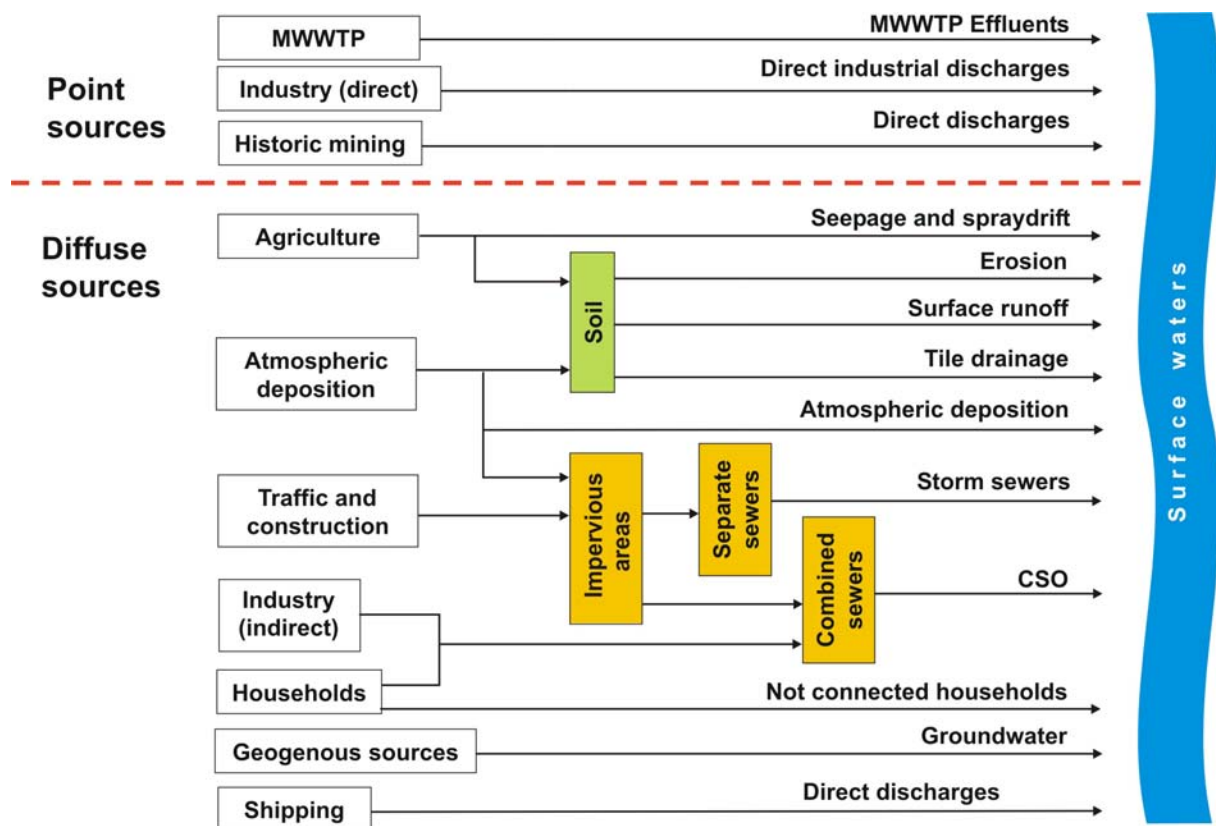


Figure 2: Considered emission sources and pathways

The MONERIS model was originally developed for nutrients and was then adapted for heavy metals and PAH. The methodology for heavy metals and PAH is based on the procedure for the nutrient phosphorus as the transport characteristics of these substances are similar.

### 2.1 Hierarchical structuring of the analytical units

The hierarchical structuring of the river basins in MONERIS starts with the “analytical unit” as smallest model unit. The grouping of all analytical units belonging to a river or river system (including the coastal waters) leads to “river basin district” according to the EU-Water Framework Directive (WFD). As set in the WFD (WFD, Art. 3.1, European Commission,

2000), the EU member states must identify and allocate each individual river basin district within their borders. The hierarchical structure of the WFD was considered in the topology of MONERIS. The following definitions of different catchment area types are represented:

analytical unit: the smallest model unit in MONERIS.

sub-unit: part of an analytical unit or river basin in an administrative unit (federal state, country).

sub-basin: combines larger parts of a river system or tributaries. This is the equivalent, boundary-wise, to the 1000 km<sup>2</sup> areas of the former MONERIS grouping and can, according to the WFD, be defined as the area of land from which all surface runoff flows through a series of streams, rivers and, possibly, lakes to a particular point in a water course (normally a lake or a river confluence) (WFD Art. 2, 14, European Commission, 2000).

coordination area / working area: organisational division of river basin districts to implement the WFD. Data is collected and aggregated and measures are planned within the coordination areas. These coordination areas are subdivided into working areas (they are not systematically included in MONERIS). Figure 1 shows the coordination areas considered in MONERIS.

river basin / river system: area of land from which all surface runoff flows through a sequence of streams, rivers and, possibly, lakes into the sea at a single river mouth, estuary or delta (WFD Art. 2, 13, European Commission, 2000). In MONERIS the term 'river system' is used synonymously for 'river basin'.

river basin district: according to article 2 paragraph 15 of the WFD (European Commission, 2000) the area of land and sea, made up of one or more neighbouring river basins together with their associated groundwaters and coastal waters (Figure 3).

Very small analytical units were merged with larger analytical units to achieve a minimum area of 100 km<sup>2</sup>. Coastal waters were identified and allocated to according river basin districts. Figure 3 shows the hierarchical structure of the topology in MONERIS.



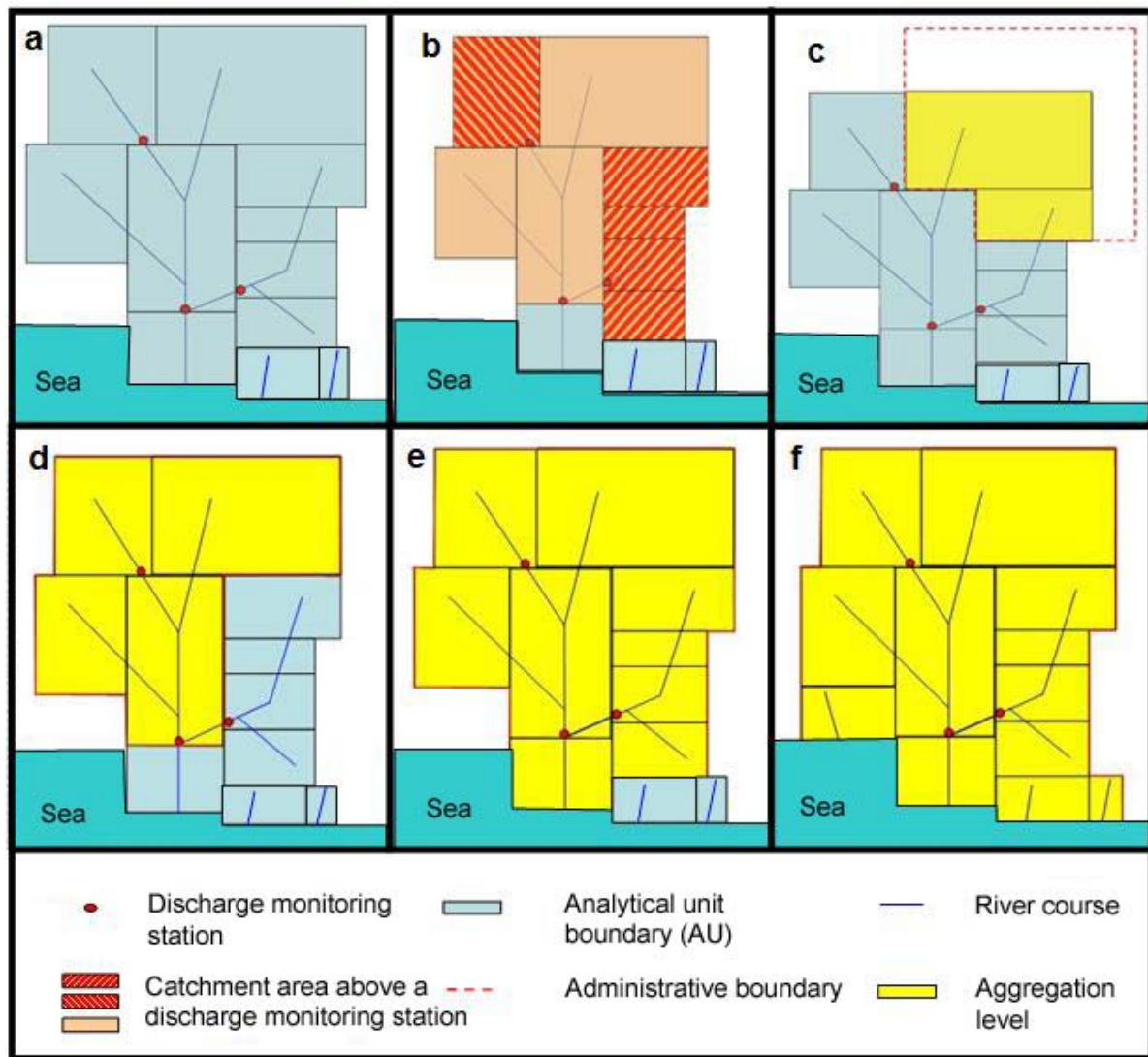


Figure 3: Hierarchical structuring of the model units

a = analytical unit; b = sub-basin; c = sub-unit; d = coordination area;  
e = river basin; f = river basin district

## 2.2 Runoff model and runoff equation

Calculations for emission and retention were made on the level of the analytical units. A runoff model that defines the direction of flow for each analytical unit was generated to aggregate loads on the watercourse system on the basis of river maps. The analytical units were assigned to the runoff model by using identifiers (ID). While „From\_ID“ defines the identifier of an analytical unit, the „To\_ID“ refers to the downstream analytical unit into which the defined analytical unit drains. The identifiers were assigned so that an area with a low „From\_ID“ would always drain to an area with a higher „To\_ID“ so that the identifiers could also be used as sorting criteria along the runoff model (Figure 4). The outlet of a river basin district is characterised by the highest ID. The first digit of an analytical unit-ID describes the appropriate river system (1 = Danube, 2 = Rhine, 3 = Ems, 4 = Weser, 5 = Elbe, 6 = Odra, 7 = direct catchment areas of the North Sea; 8 = direct catchment areas of the Baltic Sea). The

coastal areas of river basins are represented by the number of the river system and the code „9999“ (e.g. Rhine = 29999).

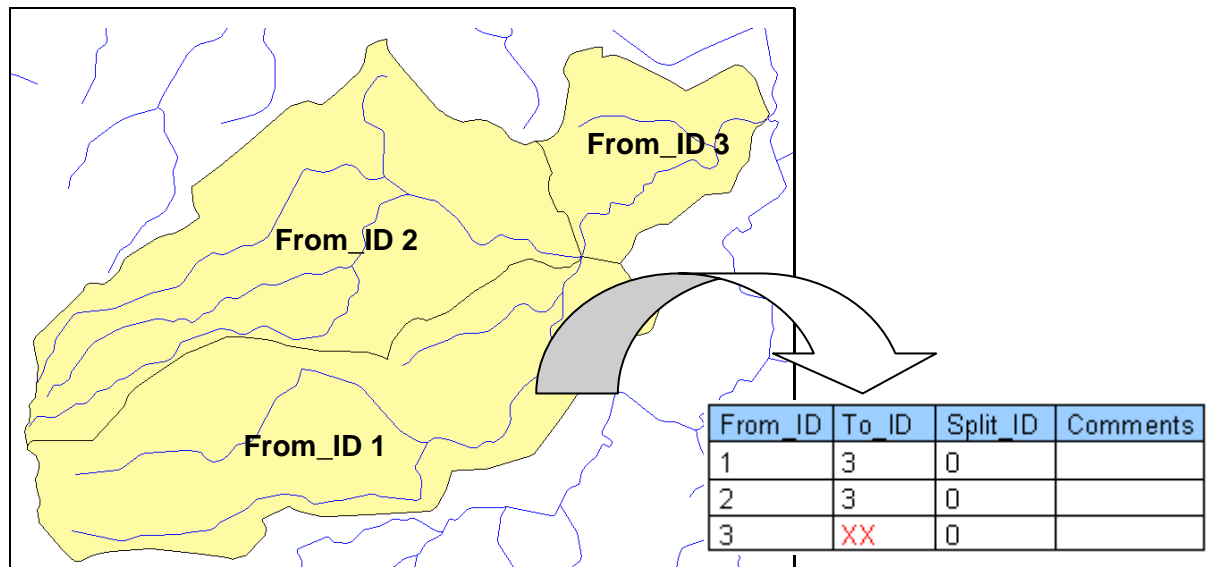


Figure 4: GIS-based generation of a runoff model

Should an analytical unit drain into two different downstream analytical units (splitting; for example due to a canal or forking) a „Split\_ID“ is inserted. Figure 5 shows the example of an analytical unit that drains through a canal into an analytical unit (green) as well as into a downstream analytical unit. The „Split\_ID“ divides the two drainages.

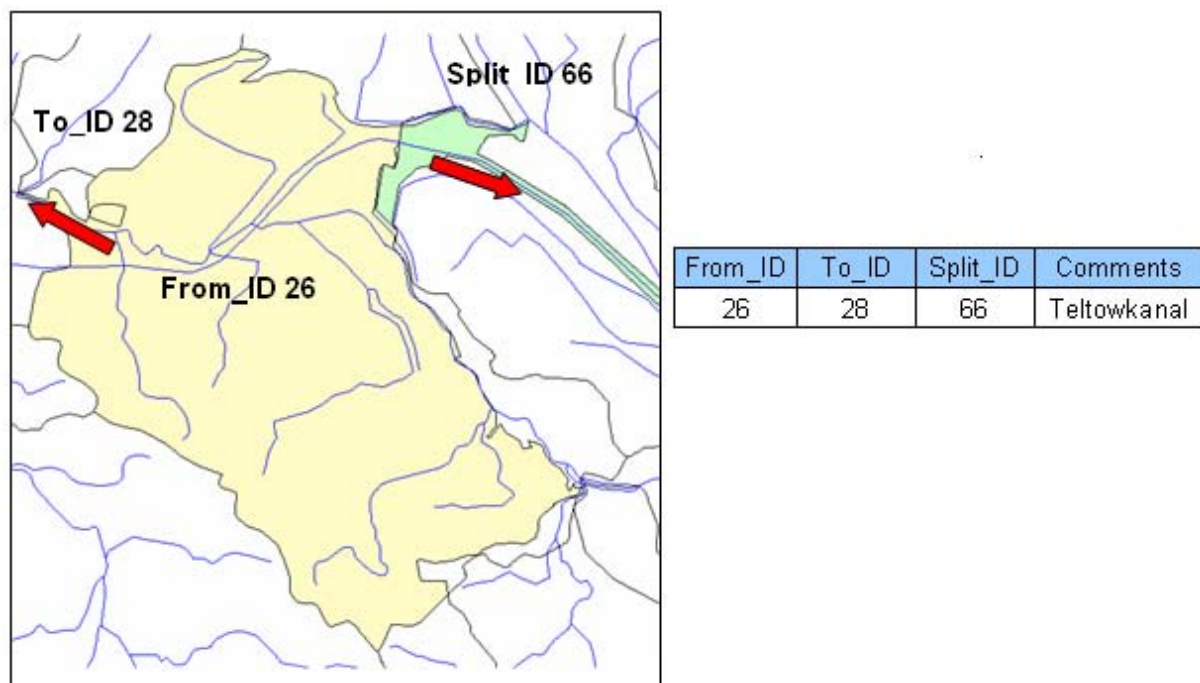


Figure 5: Splitting of the course of a river

If the course of the river is identical with the border between two analytical units, for example due to country borders (Figure 6) then the runoff model must define a main discharge area (generally the larger of the two analytical areas). The remaining area therefore drains into the

main discharge area. In addition the designated main courses were all allocated to a main discharge area.

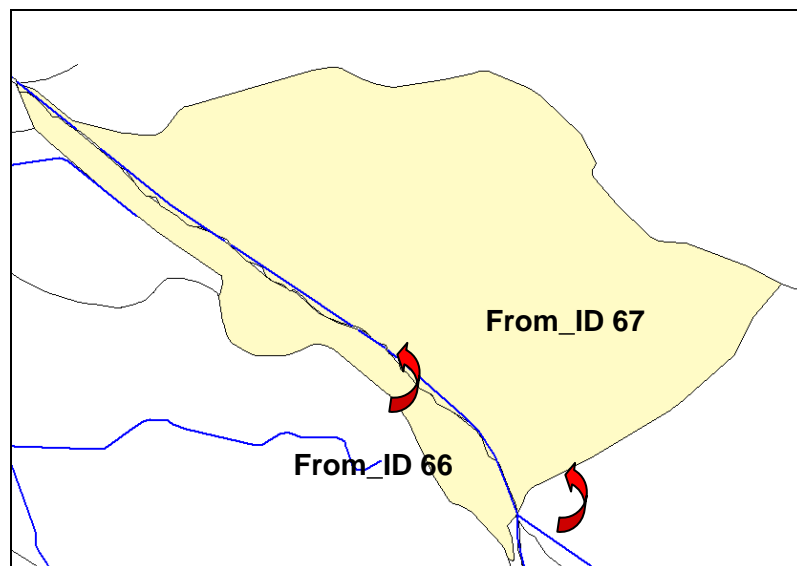


Figure 6: Runoff model definition for rivers along borders

The runoff equation (Flow Net Equation, FNE) is the description of the runoff model for the use with MONERIS and describes the topology of the river network. The runoff equation was compiled for three versions:

- FNE without splitting: This version is used e.g. to determine the size of the complete river basin.
- FNE with splitting: When calculating loads this version considers that certain parts of the load can be lead away, e.g. via a canal.
- FNE in headwaters: this version determines e.g. the runoff which drains from upstream analytical units into the observed analytical unit, without considering the runoff of the particular analytical unit. This version also considers splitting.

## 2.3 Runoff calibration

Average annual runoff from all analytical units is needed as a basis for the emission calculation with MONERIS. A runoff calibration was made for each individual year.

First of all a dataset was set up with discharge monitoring stations for which the discharge was available for at least 20 of the 23 years of the observation period. The missing years were extended by the correlation with the annual discharges of neighbouring stations. Additionally, the stations were chosen so that the complete examined area was covered evenly. Altogether the discharge values for 155 discharge monitoring stations were taken as a basis for calibrating the data (comp. paragraph 5.1).

The medium area evaporation (V) (1961 - 1990) and the average area precipitation (N) (1983 - 2005) were used to calculate the long-term average area runoff. The balance of precipitation and evaporation are then used to calculate the uncorrected area runoff. As precipitation

and evaporation do not cover the same period an additional correction factor (k) for the runoff balance (Q) was introduced (Equation 2-1).

Equation 2-1 
$$Q = N - k \cdot V$$

Q	runoff balance
N	average area precipitation
k	correction factor
V	medium area evaporation

The correction factor was assigned to all analytical units in the catchment area of a monitoring station as long as these had not already been assigned to another one further upstream. By using the runoff equation with splitting, the intermediate region runoff was summed up to a total area runoff. By adjusting the correction factors (k) the summed up area runoff was calibrated with the help of the measured river discharges. Finally, the calculated area runoff values were compared to those of a hydrological atlas (BMU, 2003). Orographically caused deviations to the runoff in the hydrological atlas, e.g. due to orographical rainfall or rain shadow, were able to be minimised by manually allocating the correction factors from neighbouring model areas.

## 2.4 Calculation of the water surface

The water surface (WS) in the analytical units is important for calculating the nutrient retention in rivers and seas as well as the emissions caused by atmospheric deposition. To estimate the water surface of the rivers the approach by VENOHR ET AL. (2005) was used to estimate a river's width. This approach estimates water surface as product of the mean river width and the flow length of a river stretch, where it is distinguished between main river (MR) and tributaries (TRIB) (Figure 7).

500 river systems in Europe with different hydromorphological characteristics were used for the calibration and validation. The calibration is based on measurements of the width of the rivers as well as detailed hydrological maps which had detailed information on the width of the rivers. The calculated water surface was then verified by statistical data of the appropriate federal state. The width of the river is calculated as dependant of the complete river basin size, the specific discharge and the average slope of the particular analytical unit (flow chart "water surfaces"). The river's water surface is, differentiated by main and tributary, added to the area of the lakes. The flow length of the tributaries is determined for each analytical unit on the basis of the difference between all river lengths of the map and the lengths of the main rivers.

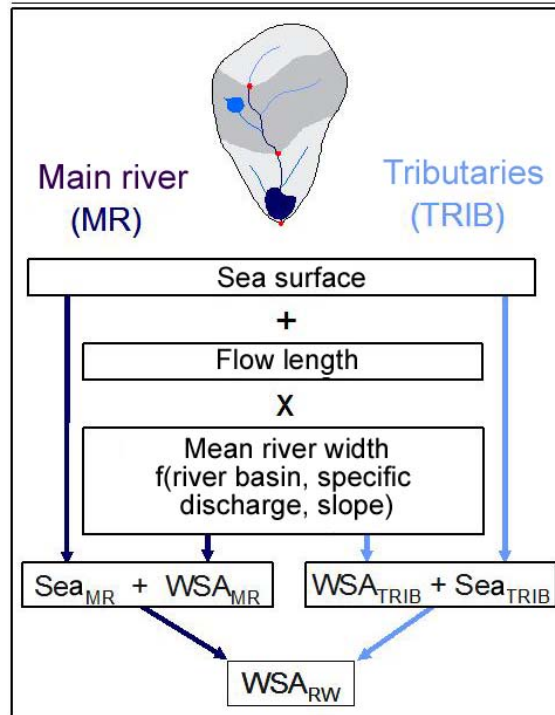


Figure 7: Calculation of the water surface (WS) of the main rivers (MR) and tributaries (TRIB) by VENOHR ET AL. (2005)

When calculating the water surface it is important to be aware that for smaller scaled maps the generalisation does increase and therefore smaller rivers and meanders go missing. This means that the real flow lengths and the water areas are underestimated. This is why the length of the rivers was taken from different maps with different scales and compared to derive scaling factors for main rivers and tributaries. For this comparison different maps with scales from 1:25,000, 1:100,000, 1:250,000 and 1:1,000,000 were used. The scaling factors (Table 1) were defined by using 87 German catchments.

Table 1: Scaling factors for maps with different scales for main rivers (MR) and tributaries (TRIB)

Map	Scale	Scaling faktor	
		Tributaries	Main river
DTK25	25,000	1.00	1.00
UBA1000	100,000	1.83	1.11
UBA-OSU1000	100,000	2.10	1.11
DLM250	250,000	3.23	1.11
DLM1000	1,000,000 (250,000)	2.99	1.13
BART1000	1,000,000	8.40	1.18
DCW1000	1,000,000	6.28	1.17

It was assumed that the complete flow length is depicted in the maps with a scale of 1:25,000.

## **2.5 Quantification of the emissions**

The following paragraphs will give an overview over the method used to quantify the emissions from point and diffuse pathways. In the following chapters 3 „General input data“ and 4 „Substance specific input data“ the basis of the data is explained. The calculation approaches for the different substances can differ. The substance related differences are explained in chapter 4. The annex has an elaborate documentation of the approach that the MONERIS model takes on the example of nutrients.

### **2.5.1 Point pathways**

To record all emissions from municipal wastewater treatment plants, direct industrial dischargers and mining activities a considerable data collection was conducted thereby trying to cover all discharged loads as exactly as possible.

#### **2.5.1.1 Municipal wastewater treatment plants**

Emissions from municipal wastewater treatment plants with a design capacity  $\geq 2,000$  PT (total number of inhabitants and population equivalents) are recorded with the help of the coordinates of the discharge point and are allocated to the appropriate analytical unit (paragraph 3.4). For the year 2005 there is exact data for the nutrient emissions of each plant (paragraph 4.1.1.1). For heavy metals and PAH the emissions were calculated on the basis of the treated annual wastewater amount (paragraph 3.4) and the average effluent concentration (paragraph 4.2.1.1 and 4.3.1.1).

Data for MONERIS on wastewater treatment with a desing capacity  $< 2,000$  PT as well as plants outside Germany is collected in an aggregated form for the analytical units.

The complete emissions from municipal wastewater treatment plants is subsequently calculated as sum of the plants  $<$  and  $\geq 2,000$  PT for the analytical units. Both datasets (exact data of the plants  $\geq 2,000$  PT and the aggregated data from the plants  $< 2,000$  PT) refer to the period 2004/2005. The consideration of the temporal development of emissions is carried out on the basis of changing factors and is described for each group of elements in chapter 4.

#### **2.5.1.2 Direct industrial discharges**

The emissions from direct industrial dischargers are recorded by data enquiries and research. Chapter 4 presents the basic data for all substance groups. In MONERIS emissions from direct industrial dischargers are recorded as sum for each analytical unit and year.

#### **2.5.1.3 Historic mining activities**

Emissions from historical mine locations are only interesting for heavy metals. Emissions into surface waters from historical mines are comprised on the basis of the data collection. The sum of all emissions for the analytical unit is calculated on the basis of the coordinates of the discharge points. The database is described in paragraph 4.2.1.3.

## 2.5.2 Diffuse pathways

The model system MONERIS considers 6 diffuse pathways: atmospheric deposition onto water surface, erosion, surface runoff, drainage, groundwater and sewer systems.

### 2.5.2.1 Atmospheric deposition onto water surface

The emission quantification through atmospheric deposition is based on the multiplication of the water surface (paragraph 2.4) with the area-specific deposition rate of the specific substance (chapter 4).

### 2.5.2.2 Erosion

The loads from unsealed areas reach the surface waters due to heavy rainfall via two pathways. Pathway „erosion“ comprises the particulate share, whereas the share of dissolved load is considered in the pathway „surface runoff“ (chapter 2.5.2.3).

Emissions via erosion from arable areas is calculated from the sediment input, the topsoil content and an enrichment factor due to the preferential transport of fine particles (Enrichment Ratio, ER) . Figure 8 shows the input data needed.

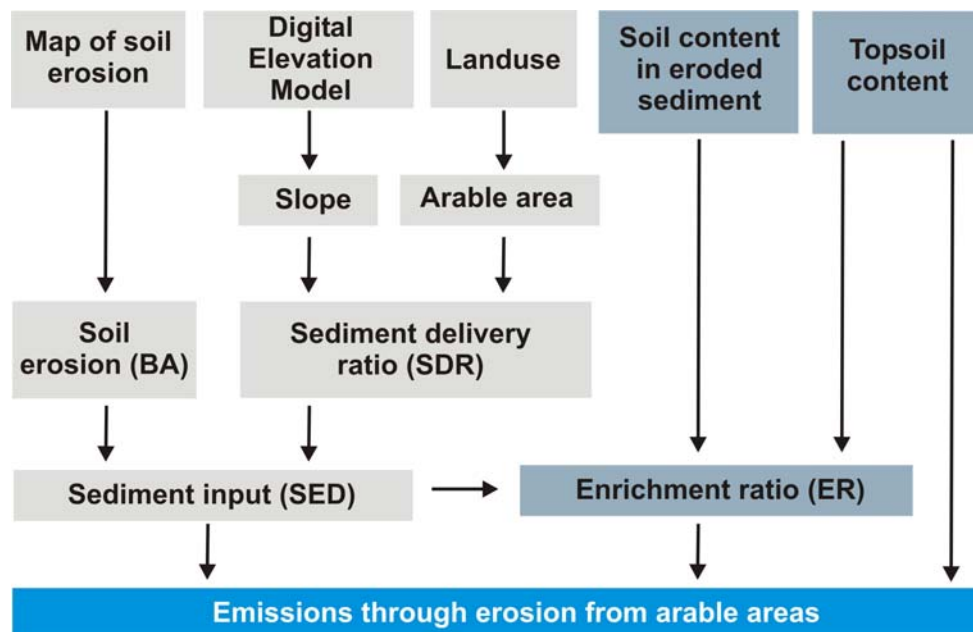


Figure 8: Input data to quantify the emissions from erosion

The quantification of sediment input (SED) is based on the soil erosion from arable areas which is dependent on the slope. The Institute of Freshwater Ecology and Inland Fisheries of Berlin (IGB) has created a soil erosion map on the basis of the digital elevation model of NASA-SRTM (NASA-SRTM, 2005), the land use data of CORINE Landcover and the European Soil Map of the European Soil Bureau (2007) and on the basis of the general soil erosion equation (ABAG) by SCHWERTMANN (1987). With the help of ABAG a long-term average soil erosion in t/(ha·a) can be calculated on the basis of 6 factors (Equation 2-2).



Equation 2-2

$$BA = S \cdot L \cdot R \cdot K \cdot C \cdot P$$

BA	average, long-term soil erosion (t/(ha·a))
S	slope factor, to be considered if the decline differs from the standard slope (9 %)
L	length of slope factor, to be considered if the length of the slope differs from a standard slope (22 m)
R	rain and surface runoff factor as a measure for the region specific erosivity of precipitation (expressed by kinetic energy, intensity and amount of rainfall)
K	soil erodibility factor, which describes the soil erosion on a standard slope with continuous bare fallow and is a measure for the soil erodibility
C	land cover and cultivation factor to be considered for all horticultural and cultivational measures
P	erosion protection factor as reducing factor for already existing protection measures (contour farming, strip farming, terracing etc.)

To define the S-factor the slope was calculated for each grid on the basis of the 100 x 100 m elevation model (SRTM, comp. paragraph 3.3) available for Europe. Then the S-factor was derived on the basis of the elevation model according to the approach by NEARING (1997) (Equation 2-3).

Equation 2-3

$$S = 1.5 + \frac{17}{1 + e^{2.3 - 6.1 \sin(\text{Slope})}}$$

Slope    Slope [%]

The L-factor is defined with the help of Equation 2-4. For slope above 17.29° (minimum of the function) the L-Factor is set to a constant value of 0.37. The equation is based on the data from the soil erosion atlas of Baden-Württemberg (GÜNDRA ET AL., 1995).

Equation 2-4

$$L = 0.0028 \cdot \text{Slopg}^3 - 0.0937 \cdot \text{Slopg}^2 + 0.729 \cdot \text{Slopg} + 1.3038$$

Slopg    Slope [°]

The K-factor is the correlation described by STRAUSS & WOLKERSDORFER (2004) on the basis of the soil's silt content (Equation 2-5). The silt content was calculated according to the data of the general soil map (BÜK 1000) or the European Soil Map respectively for each soil texture (comp. paragraph 3.3).

Equation 2-5

$$K = 0.0086 \cdot \text{silt}$$

silt        silt content of the soil [%]

The calculation of the R-factor is based on the average long-term summer rainfall according to the approach by DEUMLICH & FRIELINGHAUS (1993) (Equation 2-6).

Equation 2-6

$$R = ([P6190\_su] \cdot 0.152) - 6.88$$

P6190\_su        average long-term summer rainfall [mm/a]

The conversion of the long-term average soil erosion into soil erosion for each individual year is based on including a rainfall-dependant weighting coefficient (ER\_PRECcorr, current period compared to a long-term average). This weighting coefficient is deduced on the basis of the relation between the number of intense rain days (according to ROGLER & SCHWERTMANN, 1981) and the number of intense rain days in the total observation period.

The C-factor was calculated from the values which AUERSWALD & SCHMIDT (1986), STRAUSS & WOLKERSDORFER (2004), DEUMLICH & FRIELINGHAUS (1993) as well as SCHWERTMANN ET AL. (1987) specified as average. This yields the land use specific C-factors between 0.004 and 0.38 (Table 2).

Table 2: Land use specific average C-factors

Land use	C-factor
Corn	0.380
Potatoes	0.280
Sugar beets	0.280
Rapeseed	0.100
Winter barley	0.100
Grains	0.100
Vegetables	0.250
Pulses	0.250
Grassland / pasture	0.010
Deciduous forest	0.004
Coniferous forest	0.008
Mixed forest	0.004
Shrub- and bushland	0.010

The percentage of soil erosion from arable areas that ends up in surface waters is calculated by the sediment delivery ratio (ER\_SDR) (WALLING, 1983; 1996). Based on a GIS-supported model of loads caused by erosion, those individual areas within a catchment area can be identified where soil erosion reaches the waterbody. This analysis was done for different catchment areas where precise data was given (BEHRENDT ET AL., 1999). Subsequently, the relation between the sediment delivery ratio and certain catchment area attributes were identified. Non-linear multiple regression analyses showed that the slope and the share of arable area had the largest influence on the sediment delivery ratio. The sediment delivery ratio for all analytical units is calculated with the help of the relation shown in Equation 2-7.

$$\text{Equation 2-7} \quad \text{ER\_SDR} = 0.012 \cdot (\text{Slope} - 0.25)^{0.3} \cdot A_{\text{arable area}}^{1.5}$$

ER\_SDR      sediment delivery ratio [%]

Slope      slope [%]

A<sub>arable area</sub>      share of arable area [%]

During the erosion process fine particles accumulate in the eroded sediment due to the favoured transport of fine soil particles. As pollutants are bound to finer grains due to their

higher specific surface these also accumulate during the transport process. The enrichment of a substance in the erosion material is described by the enrichment ration (EnR) compared to the substance content in arable land. This enrichment ratio is calculated according to BEHRENDT ET AL. (1999).

Apart from the sediment input from arable areas the natural erosion from open mountain rock (mountain rock above 1,000 m above sea level) must be considered.

### 2.5.2.3 Surface runoff

Emissions of solutes via surface runoff from unsealed areas are calculated according to the scheme on Figure 9.

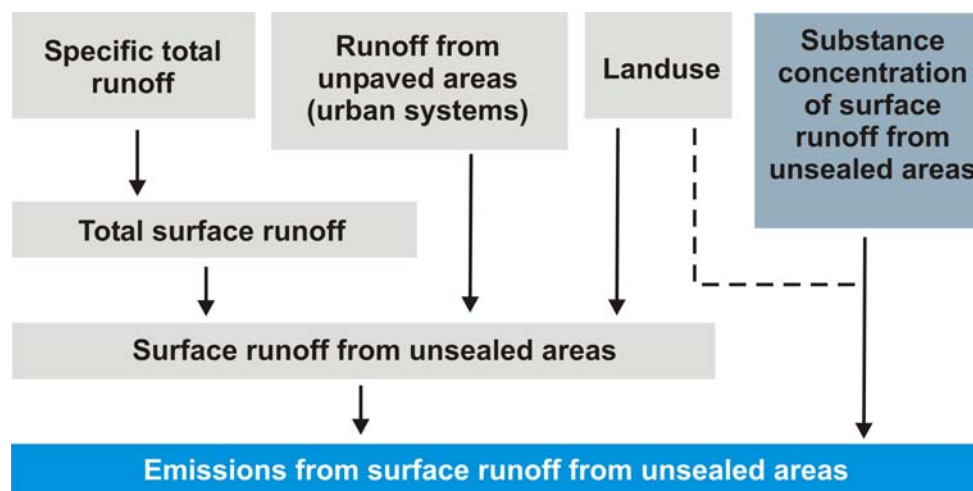


Figure 9: Input data to quantify emissions via surface runoff from unsealed areas

The surface runoff is calculated with an approach developed by CARL ET AL. (2008) and CARL & BEHRENDT (2008) as a function of the total runoff (Equation 2-8). This approach was used by SCHREIBER ET AL. (2005) for the river basin of the Danube. The results of this conceptional time series model are comparable to those of the hydrological models such as SWAT (Soil and Water Assessment Tool) for chosen example areas of the Danube river basin (SCHREIBER ET AL., 2005).

Equation 2-8 
$$Q_{SR\_pre} = 0.0426 \cdot Q_{calc}^{1.2461}$$

$Q_{SR\_pre}$  total surface runoff [mm/a]

$Q_{calc}$  total runoff [mm/a]

According to Equation 2-8 the complete surface runoff of the analytical units can be calculated. It must be considered that also impervious urban areas cause surface runoff. Runoff from impervious surfaces is comprised in the pathway „sewer systems“ (paragraph 2.5.2.6) and must therefore be deducted. The surface runoff for the different land use categories can be calculated from the resulting surface runoff for unsealed areas in the analytical units and the land use.

#### 2.5.2.4 Tile drainage

The quantification of emissions via tile drainage results from the size of the drained area, the specific drainage rate and the substance concentration in the drainage water (Figure 10). The estimation of the drained area is described in paragraph 3.3. The drain discharge is calculated according to KRETSCHMAR (1977) on the basis of the assumption that 50 % of precipitation in winter and 10 % of precipitation in summer drain away. This approach considers the regional differences in precipitation distribution. The procedure for calculating substance concentrations is described in chapter 4.

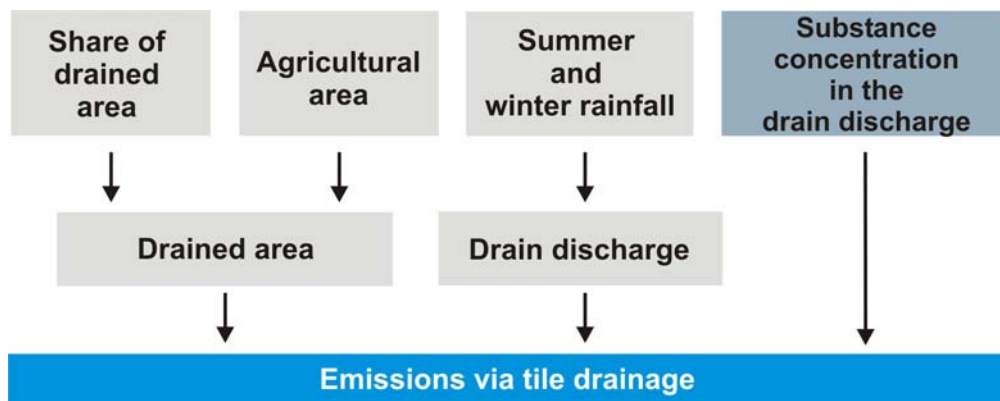


Figure 10: Input data for quantifying emissions via tile drainage

#### 2.5.2.5 Groundwater inflow

Emissions through groundwater inflow are calculated as product of groundwater discharge and substance concentration in the groundwater and includes the natural interflow and base flow. The groundwater runoff is calculated for each analytical unit as the difference between the total runoff and the sum of other runoff components (drain discharge, runoff from pervious and impervious areas, runoff from point sources and rainfall onto the water surface).

#### 2.5.2.6 Sewer systems

The emissions from sewer systems are composed of four different pathways (Figure 11):

- emissions from impervious urban areas via storm sewers,
- emissions from households and impervious urban areas via combined sewer overflows,
- emissions from households and impervious urban areas that are connected to a sewer system but not to a wastewater treatment plant and
- emissions from households and impervious urban areas that are not connected to a sewer system.

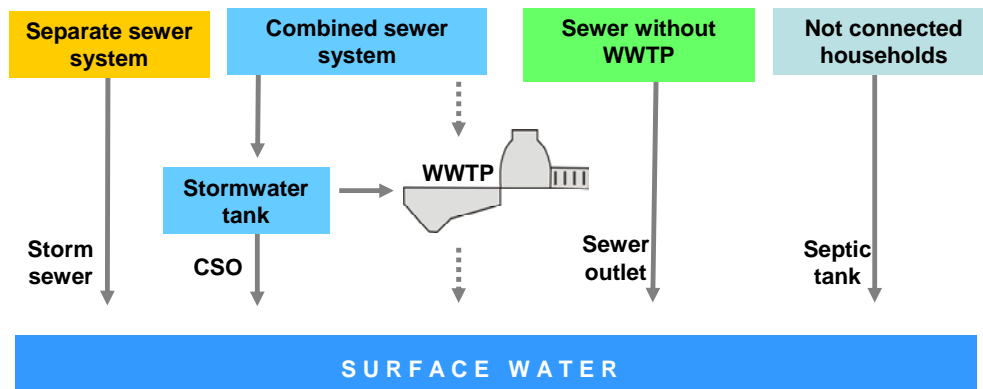


Figure 11: Pathways of sewer systems (without WWTP)

In Figure 12 the necessary input data to quantify emissions from sewer systems is shown.

The complete urban areas were taken from CORINE Land Cover (CLC 2000) (paragraph 3.3.1). The impervious urban area ( $US_{IUA\_tot}$ ) was calculated according to HEANEY ET AL. (1976) considering the population density. (paragraph 3.3.7) (Equation 2-9). Subsequently the impervious urban areas are allocated to the sewer systems in the analytical units according to the proportion of connected population (paragraph 3.3.8) and the proportion of combined and separate sewer systems (paragraph 3.3.9.1).

$$\text{Equation 2-9} \quad US_{IUA\_tot} = Cus_1 \cdot (Cus_2 \cdot E_{Dichte})^{Cus_3 - Cus_4 \cdot \log(Cus_2 \cdot E_{Dichte})} \cdot \frac{BI_{LU\_urb}}{100}$$

$US_{IUA\_tot}$  impervious urban area [ha]

$BI_{LU\_urb}$  urban area [ha]

$E_{Dichte}$  population density [E/ha]

$Cus_1 - Cus_4$  coefficients:  $Cus_1=9.6$ ;  $Cus_2=0.4047$ ;  $Cus_3=0.573$ ;  $Cus_4=0.0391$

The surface runoff from impervious areas needs to be calculated to determine the total runoff from the different sewer systems. The proportion of the drained precipitation is calculated with the help of the runoff coefficient, which according to HEANEY ET AL. (1976) is defined on the basis of the degree of surface sealing in each analytical unit. The precipitation and runoff coefficient then lead to the surface runoff for the areas connected to the different sewer systems.

#### *Emissions from storm sewers (separate sewer system)*

Emissions from storm sewers are calculated with the help of area-specific surface load from the impervious surfaces. The emission into surface waters is derived from the multiplication of the area-specific surface load with the area of the impervious surface connected to the separate sewer system.

#### *Emissions from combined sewer overflows*

Combined sewage comprises wastewater from both households and indirect dischargers together with stormwater runoff in one sewer and is lead to a wastewater treatment plant. During heavy rainfall the system is only able to retain a fraction of the total water volume.

The water quantity exceeding the storage volume is discharged into surface waters by means of combined sewer overflows (Figure 11). The extent of the water pollution from combined sewer systems depends especially on the effective annual overflow duration. This parameter is controlled by the retention volume, i.e. the available storage volume of the stormwater overflow tanks. The amount of water discharged (overflow rate;  $US_{CS\_Q}$ ) is estimated according to MEIßNER (1991) depending on the annual precipitation amount and the storage volume of the stormwater overflow tanks (paragraph 3.3.9.2) (Equation 2-10).

$$\text{Equation 2-10} \quad US_{CS\_Q} = \frac{\frac{4000 + 25 \cdot q_R}{0.551 + q_R}}{V_S + \frac{36.8 + 13.5 \cdot q_R}{0.5 + q_R}} - 6 + \frac{PD_{PREC\_yr} \cdot 800}{40}$$

$US_{CS\_Q}$	overflow rate of the combined sewer system [%]
$q_R$	rainfall runoff rate [l/(ha·s)]
$V_S$	specific storage volume [m <sup>3</sup> /ha]
$PD_{PREC\_yr}$	annual precipitation [l/(m <sup>2</sup> ·a)]

Apart from the stormwater, the discharged combined sewage also contains wastewater from households and indirect discharges from small industries. The emissions are estimated on the basis of the effective annual overflow duration. Emissions from households are calculated on the basis of the amount of inhabitants connected to the combined sewer as well as a substance specific inhabitant load. Small industries are given an average concentration in the effluent. It is assumed that industrial areas cover 8% of urban areas. The amount of water is determined on the basis of a specific runoff for commercial areas and the assumption that this is effective 10 hours a day.

#### *Emissions from sewer systems that are not connected to a wastewater treatment plant*

Furthermore, loads from impervious areas and from inhabitants that are connected to a sewer system but not to a wastewater treatment plant have to be considered. The share of these areas to the complete impervious area is determined by the part of the population that is only connected to sewer system (paragraph 3.3.8). In regard to the emissions these areas can be seen as the areas connected to a separate sewer system. Additionally the complete sewage from households and small industries is discharged into surface waters. The same assumptions are made to determine the emissions as for the combined sewer system.

#### *Emissions from impervious areas and inhabitants with no connection to a sewer system*

For households and impervious areas without a connection to a sewer system it is assumed that part of the solid matter is pumped out of the septic tanks and is taken to wastewater treatment plants. This is not added to the calculation as it is already included in the emissions from the wastewater treatment plants. Therefore only a part of the dissolved share of pollutants from the septic pools reaches the waterbodies after percolation through the soil. Loads from small industries are not expected for this pathway.

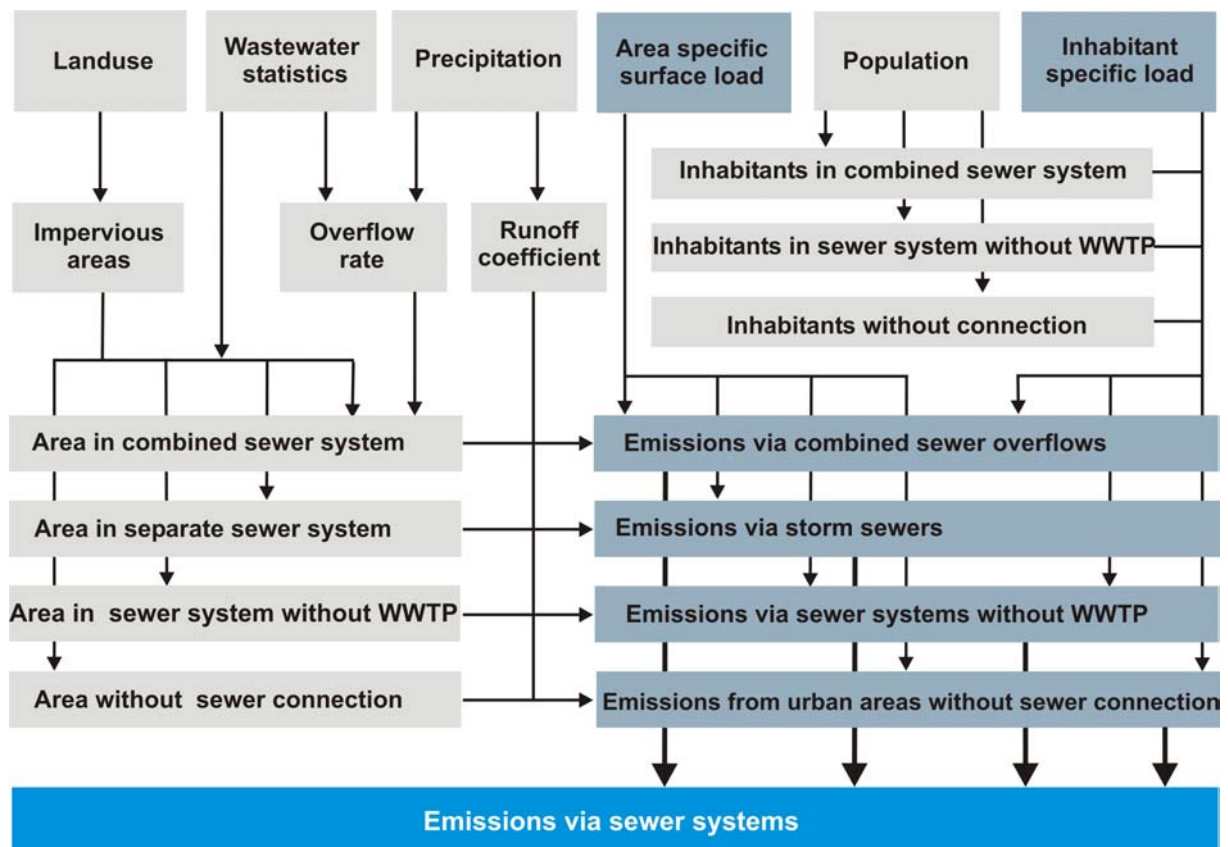


Figure 12: Input data for quantifying emissions via sewer systems

### 3 General input data

This chapter gives an overview over the general input data that was used. Part of the input data used has been described in detail in earlier reports (BEHRENDT ET AL., 1999; BEHRENDT ET AL., 2002a, FUCHS ET AL., 2002). Therefore, only recently collected and preprocessed data within the framework of the current project will be illustrated.

#### 3.1 Analytical units

The analytical units (paragraph 2.1) were compiled by accumulating the partially very detailed catchment area borders given by the federal states, whereby the border for the accumulation was set at an areas size of 100 km<sup>2</sup>. The analytical units of the Oder and Elbe that are outside of Germany were taken from the ICM-Oder project (BMBF, 03F0403A-H) resp. the GLOWA-ELBE (BMBF, 01LW0304A).

For the areas of the Rhine outside Germany the borders set in the UBA project 29922285 (BEHRENDT ET AL., 2003b) were used. Figure 13 shows the analytical units used. The whole area encompasses 3.456 analytical units with an average size of 190 km<sup>2</sup>. 2.759 of these areas are in Germany and have an average size of 135 km<sup>2</sup>. The analytical units were split along the borders (states and federal states) so that they could be allotted to an appropriate administrative unit.

#### 3.2 River discharge and quality data

Comprehensive river discharge and quality data was collected from the responsible federal and state authorities to create a runoff balance and to calculate loads in the surface waters. As far as available the BfG supplied the data for the federal waterways. The total daily discharge data from 1273 discharge monitoring stations was provided as well as substance concentrations for 1591 quality monitoring stations for the current period (1998 to 2005).

Not all measurement series could be used for the model application and respectively the validation later as

- an analytical unit often comprises several monitoring stations. In this case only one of the monitoring stations was considered.
- some of the monitoring stations were located so awkwardly that they couldn't be clearly allocated to an analytical unit.
- some of the stations only had a series of measurements available for less than a year or there was no spatial allocation between the discharge and quality monitoring station possible. Therefore these stations were excluded.
- only quality stations with more than 10 measurement values per year were considered for calculating the loads.



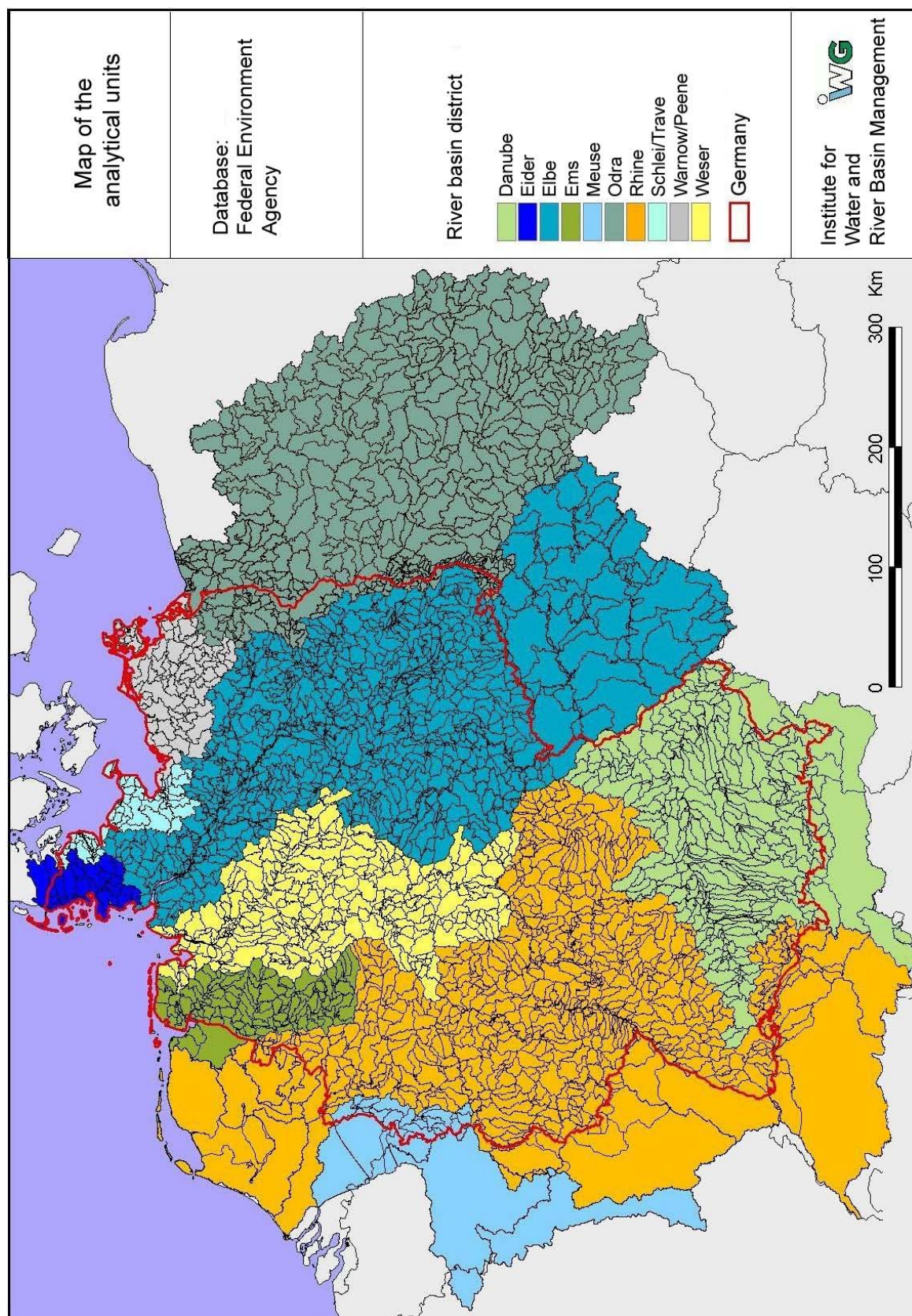


Figure 13: Map of the river basin districts and the analytical units

When calculating the river loads (BEHRENDT ET AL., 1999) the following sources for errors should be minded:

- If the river discharge and the water quality are measured at different points the load is calculated with the help of correcting factors which are defined by the ratio between the size of the catchment area of the discharge monitoring station and the corresponding water quality monitoring station. Due to the lack of monitoring stations the correcting factors are sometimes larger than 2. It is not clear to which extent such large correction factors actually represent the real runoff situation in those catchment areas.
- As most parameters are not read continuously the river loads from immission monitoring stations are generally underestimated. During flood events the emissions into the water bodies are higher and substances are mobilised from the sediment. Readings from quality monitoring stations are very scarce in those cases.

After processing and critically reviewing the data on its adequacy 155 discharge monitoring stations were chosen to calibrate the runoff balance. An additional 513 monitoring stations were used for validation. A map of the monitoring stations used for the calibration and validation is depicted in chapter 5.1 (Figure 40).

The amount and quality of the data for the different substance groups varies widely. Especially the large amount of data below the limit of quantification for heavy metals and PAH are a problem when calculating loads. The results of these load calculations are shown for each substance group in chapter 5.2 (nutrients), 5.3 (heavy metals) and 5.4 (PAH).

### **3.3 Spatial input data**

#### **3.3.1 Land use**

Data of the CORINE Land Cover (CLC 2000, European Environment Agency, EEA, 2005) was used for the classification of land use. Data is collected from a grid of 500 x 500 m which is then calculated down to a 100 x 100 m resolution (Figure 14). Due to the coarse grid small partially used areas such as water surfaces are not detected properly. CORINE land use data for Switzerland for the reference year 2000 was not available in time for the data processing. This is why Switzerland's data from 1990 (European Environment Agency, EEA) was used as basic data. Classes for the CORINE Land Cover were aggregated according to tab.-ann. 8 for modelling.



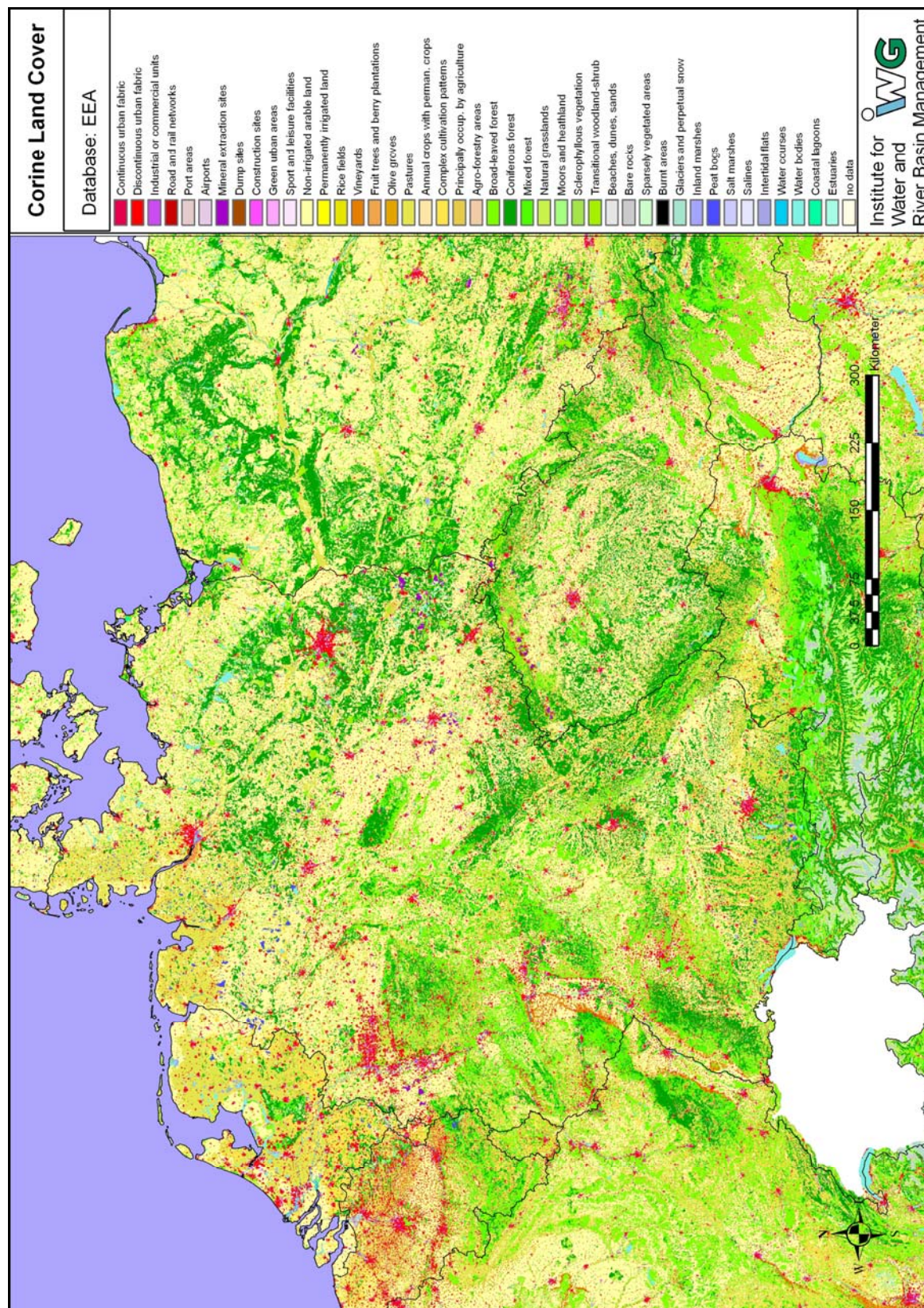


Figure 14: Map of land use (CORINE Land Cover 2000, EEA 2005)

### **3.3.2 Digitale Höhenmodelle**

The worldwide available digital elevation model GTOPO30 of the U.S. Geological Survey (USGS, 1996) with a grid size of 1 x 1 km was used to derive the average slope and the average height of the catchment areas. The soil erosion map was generated with the digital elevation models of NASA-SRTM (2005).

### **3.3.3 Precipitation data**

Annual and summer precipitation sums were deduced from the interpolated rainfall data (monthly values from 1983-2005) of the Global Precipitation Climatology Centre (GPCC, 2006) and were clipped to fit the catchment area borders.

### **3.3.4 Soil maps**

The following digital ground maps were available: the general soil map (BÜK 1000; on a scale of 1:1,000,000) of the Federal Institute for Geosciences and Natural Resources (BGR, 1998) and the European Soil Map of the European Soil Bureau (2007). Information was extracted for the share of sand, clay, loam and silt dominated soils as well as fens and bogs, their permeability and the nitrogen content of the topsoil.

### **3.3.5 Geological maps**

The differentiation between loose rock and bedrock areas within the German river basins was made on the basis of the geological map of Germany (GK 1000; on a scale of 1:1,000,000) of the Federal Institute for Geosciences and Natural Resources (FISA/BGR, 1993) and for the analytical units outside Germany from the hydrogeological map of Europe of the National Institute for Public Health and Environment (RIVM, 2007).

### **3.3.6 Share of drained arable land**

The calculation of the share of drained areas of the complete arable area is based on the work of BEHRENDT ET AL. (1999), whereas the drained areas of the river basins in the new states of Germany was estimated with a weighting of the soil types (Figure 15). For this the drainage area maps were merged with the agricultural mapping to determine the soil types of the drained areas and river basins (BEHRENDT ET AL., 1999).

It was deduced that most of the areas (in average 78 %) of the moist soil types are drained, the water logged deep loam and loam locations with about 41 % taking up the largest part of that area (BEHRENDT ET AL., 1999). HIRT ET AL. (2005a, b) deduced similar results for drained areas in the Mulde basin with a different approach.



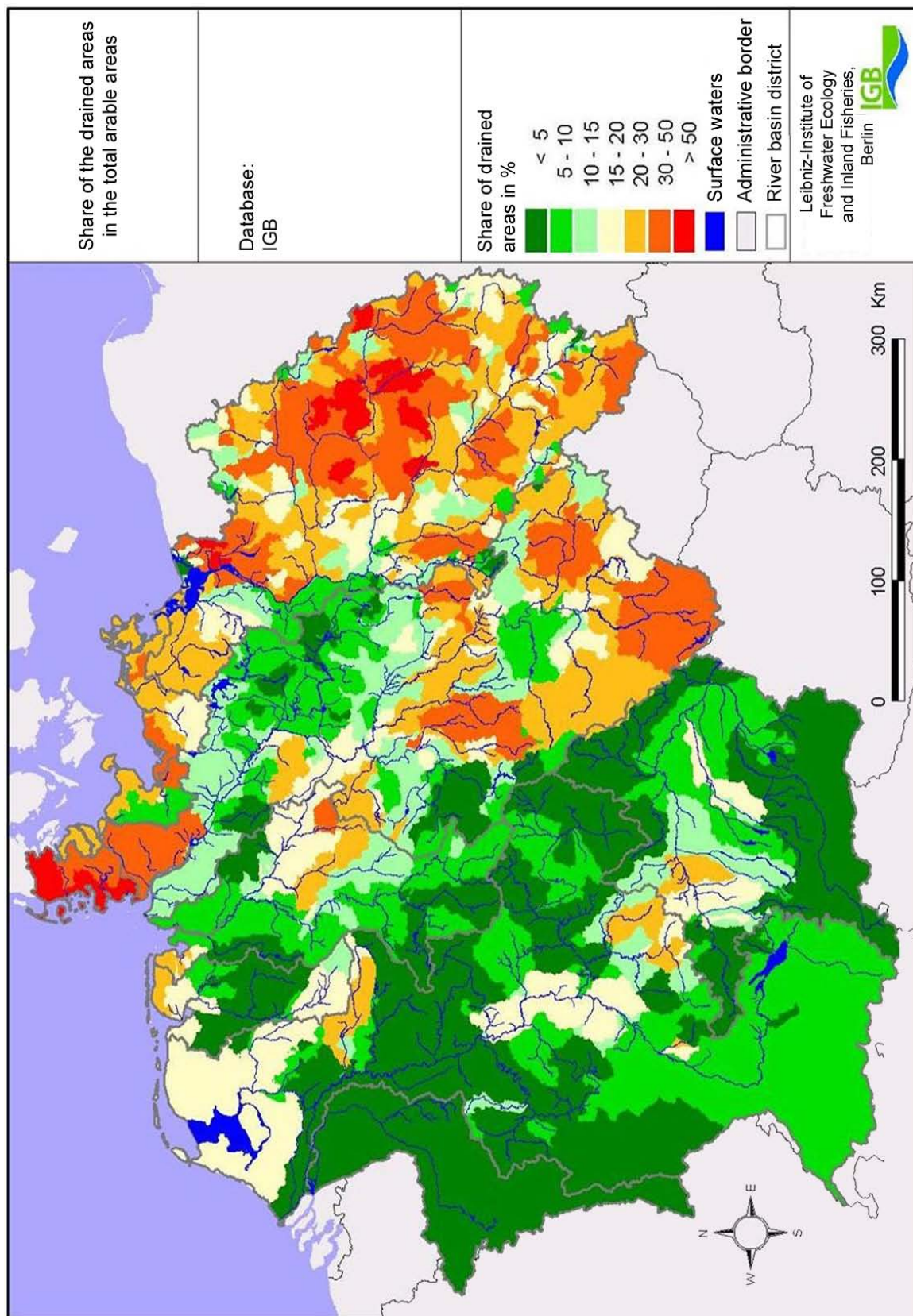


Figure 15: Share of drained arable land in the total arable areas

### 3.3.7 Population

The European Environment Agency (EEA) provided a digital map of the population density for the year 2001 (EEA, 2007). This map is based on statistical municipal data on population, which was regionalised with the help of CORINE land use classes (CLC 2000, EEA, 2005). Before this map was used the population in several municipalities of different federal states was determined and compared to the statistical data (STATISTIK LOKAL, 2004). In total it showed a good concordance to the local level. Figure 16 shows the population density for the river systems.

The population for the year 2001 was calculated for the analytical units. The areas of the Swiss Rhine basin are not included in the EEA map (comp. Figure 16). The missing data was supplemented by a municipal map on population compiled by the IGB (BEHRENDT ET AL., 2003a). There is no further spatial high-resolution data for the population for the complete period between 1983 and 2005. The population development was therefore considered on the basis of statistical data on state and federal state level. For this the population data of the Statistical Office of the European Communities (EUROSTAT, 2007a), the Statistical State Offices (Brandenburg, Saxony, Saxony-Anhalt, Thuringia) as well as the Statistical Yearbook of the GDR (1986) (comp. annex tab.-ann. 1) were used. Then detailed data for the year 2001 was used as a basis to chart the changes in population on a federal state and state level for all calculation years in the analytical units. In doing so it was assumed the population development in the analytical units would be the same as in the administrative borders.

### 3.3.8 The populations connection rate to sewer systems

Statistical microdata on a municipal level from the research data centre of the statistical offices of the state and federal states (FDZ, 2007) was used to determine the populations connection rate to sewer systems in the analytical units in Germany. Data is collected every three years for the attribute 'municipality under service' and is available from the year 1998 onwards. The total population as well as the connection rate to the public sewer systems and wastewater treatment plants is registered.

The most recent dataset at the time of research was that of the year 2004, which was then used to determine the connection rate within the river basins. The number of inhabitants connected on a local level was transferred area-weighted to the analytical unit. The map with the administrative boundaries (VG250) needed for the reference date of the statistical data collection (31<sup>st</sup> December) was supplied by the Federal Agency for Cartography and Geodesy (BKG, 2007). By intersecting the municipal boundaries with the analytical units the connected inhabitants for the intersecting areas are calculated. So the connected inhabitants in an analytical unit result from the sum of the share of connected inhabitants of all municipal areas that lie within the corresponding analytical unit. Finally, on this base the connection rate of the population to the public sewer system and wastewater treatment plants is determined. The resulting connection rate for the year 2004 is depicted at Figure 17.



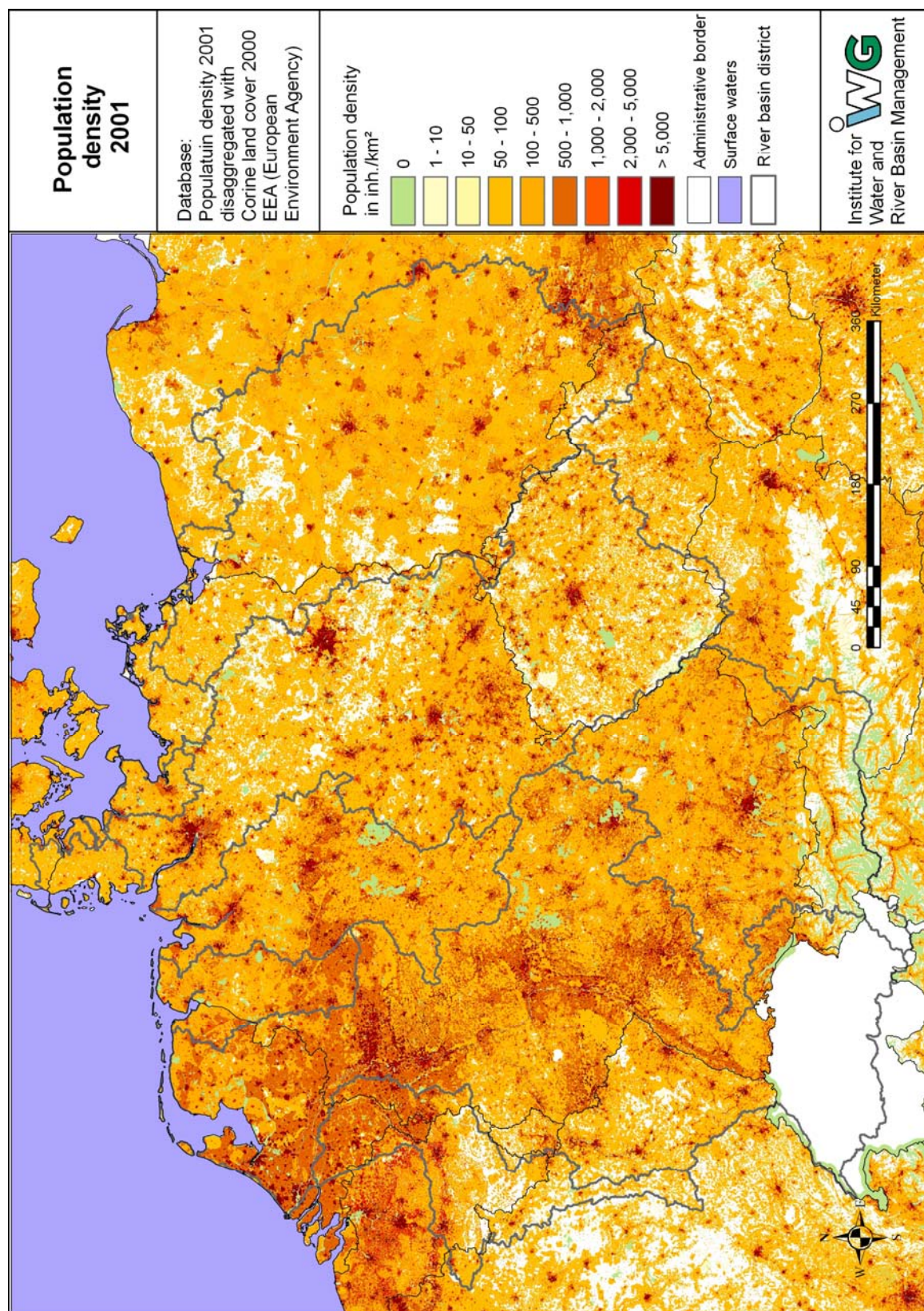


Figure 16: Population density in 2001 (European Environment Agency)



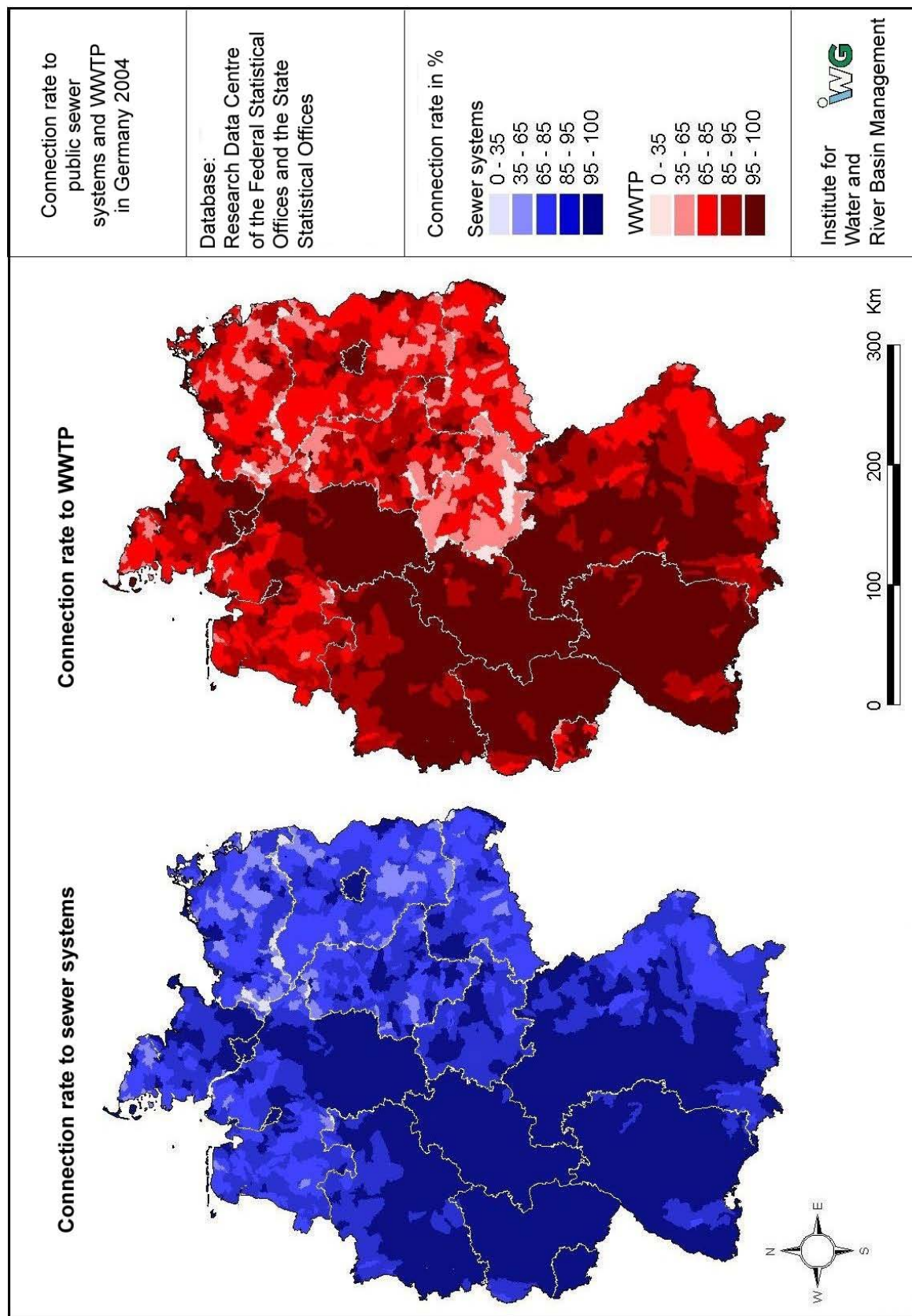


Figure 17: The populations connection rate to public sewer systems and MWWTP in Germany



As there is no high-resolution data for the complete period a temporal change from 1983 to 2004 at federal state level was included. For this the data of the Federal Statistical Office (1983, 1987, 1991, 1995, 1998a, 2001 and 2004) was used. The Federal Statistical Office only has data for the new states of Germany for the years 1991 and following. But it can be presumed that the changes of the connection rate between 1983 and 1991 were minimal in the former GDR. The connection rates taken into account for the federal states are in the annex (Tab.-Ann. 2, Tab.-Ann. 3).

First of all the change factors on a federal state level were determined for every individual year referring to the reference year 2004. For the years lacking statistical data the factors were determined by linear interpolation. Subsequently, based on the detailed data for the year 2004, the number of connected inhabitants in the analytical units was determined for each year with the help of the change factors. In the process the connection rate to the wastewater treatment plants in an analytical unit must not rise above the connection rate to the sewer system and the connection degree cannot exceed 100%.

From Figure 17 it is obvious that the connection rate in the less densely populated regions in the east as well as the north west of Germany is lower than in the more densely populated federal states. And it can also be seen that the connection rate in the new states of Germany still lies below the federal average in 2004 (Figure 17, tab.-Ann. 2, tab.-Ann. 3).

EUROSTAT has the data for other countries on their connection rate to public sewer systems and wastewater treatment plants (EUROSTAT, 2007b, c, tab.-ann. 2, tab.-ann. 3). The data is not available for all countries for the complete period which is why the data series had to be interpolated. For the other countries it is assumed that the connection rate within the analytical unit does not vary.

### **3.3.9 Sewer systems**

#### **3.3.9.1 Distribution of combined and separate sewer system**

The length of the wastewater, stormwater and combined sewers are known on municipal a level from the FDZ (2007). However, the data is only collected according to the address of the operating company. This means that data from larger sewage boards that operate sewer systems of different municipalities are assigned to the municipality where the headquarter of the operating company is located. Evaluating the federal statistics on public sewer systems has shown that this exposes problems, especially in the new states of Germany, as there are several municipalities not accounted for. So these statistics cannot be considered for questions on a local level in this project. This is why data was aggregated on a district level to reduce those kinds of errors.

For some federal states the length of the sewers is additionally charted for the attribute 'municipality under service' (state version of the statistic on public wastewater disposal) and therefore record the actual sewers in a municipal area. This data is available for Schleswig-Holstein, Lower Saxony, North Rhine-Westphalia, Rhineland-Palatinate, Baden-Württemberg, Saarland, Brandenburg, Mecklenburg-Western Pomerania (only 1998), Saxony, Saxony-Anhalt and Thuringia (only 2001 and 2004). Data from the federal version was included for all other federal states. The sewer lengths of the state versions also were aggregated on a district level to ensure that the spatial resolution within Germany is kept the

same for all federal states. The share of combined and separate sewer systems on the district level was calculated with the help of the length of the combined and wastewater sewers for 1998, 2001 and 2004. Figure 18 (left) shows the share of combined sewer systems on a district level for the year 2004. The share of separate sewer systems results from the difference of the combined sewer system share and 100 %. Subsequently, the shares in the analytical units were calculated as area-weighted mean. The development in time from 1983 to 2004 was calculated with the help of data from the Federal Statistical Office (1983, 1987, 1991, 1995, 1998, 2001, 2004) on a federal state level (comp. tab.-ann. 4). Southern Germany is mainly drained by combined sewer systems, whereas the northern federal states use separate sewer system drainage. From 1987 to 2004 the share of separate sewer systems increased continuously and has now reached 42% in Germany. (Figure 18, tab.-ann. 4).

There is no detailed data for the countries outside Germany on the distribution of storm sewage and separate-sewer system. Requests at the European Water Association (EWA, 2005) in Austria, Belgium, Luxembourg, France, the Czech Republic, the Netherlands and Poland only yielded little usable information. The percentage of storm sewage and separate-sewer systems was therefore estimated on the basis of the average height of the catchment area. This procedure is based on the assumption that separate sewer systems are used for low gradients.

### **3.3.9.2 Design capacity of combined sewage treatment**

In the statistic on public sewage disposal (federal version) the number and volume of stormwater overflow tanks in the combined sewer system are included. However, the municipal data is not usable as they are, as already described, registered by the location of the head office of the operating company and not by the actual location of the stormwater overflow tanks. The data was therefore aggregated on a district level. However, even after aggregating the resulting storage volume still doesn't match the actual disposed area for all districts. For example the towns of Magdeburg and Suhl have no data on their sewage disposal in the statistics. The district data on storage volumes therefore had to be corrected. For this the length of the sewers in the state version on sewage disposal was used. By comparing the combined sewer lengths in the federal and state versions the storage volume was adjusted according to the differences. It was furthermore found that especially in districts with unincorporated towns, there are inconsistencies between the tank volumes of the town and the neighbouring district. Some of the stormwater overflow tanks that take in the combined wastewater of the unincorporated town lie in the area of the district and are operated by a company residing in the rural district area. In those cases the storage volumes of the district and the unincorporated towns were balanced. These corrections were taken in Lower Saxony, Rhineland-Palatinate, Saarland, Brandenburg, Mecklenburg-Western Pomerania, Saxony, Saxony-Anhalt, Thuringia, Berlin and Mecklenburg-Western Pomerania. All other state data did not need to be corrected.

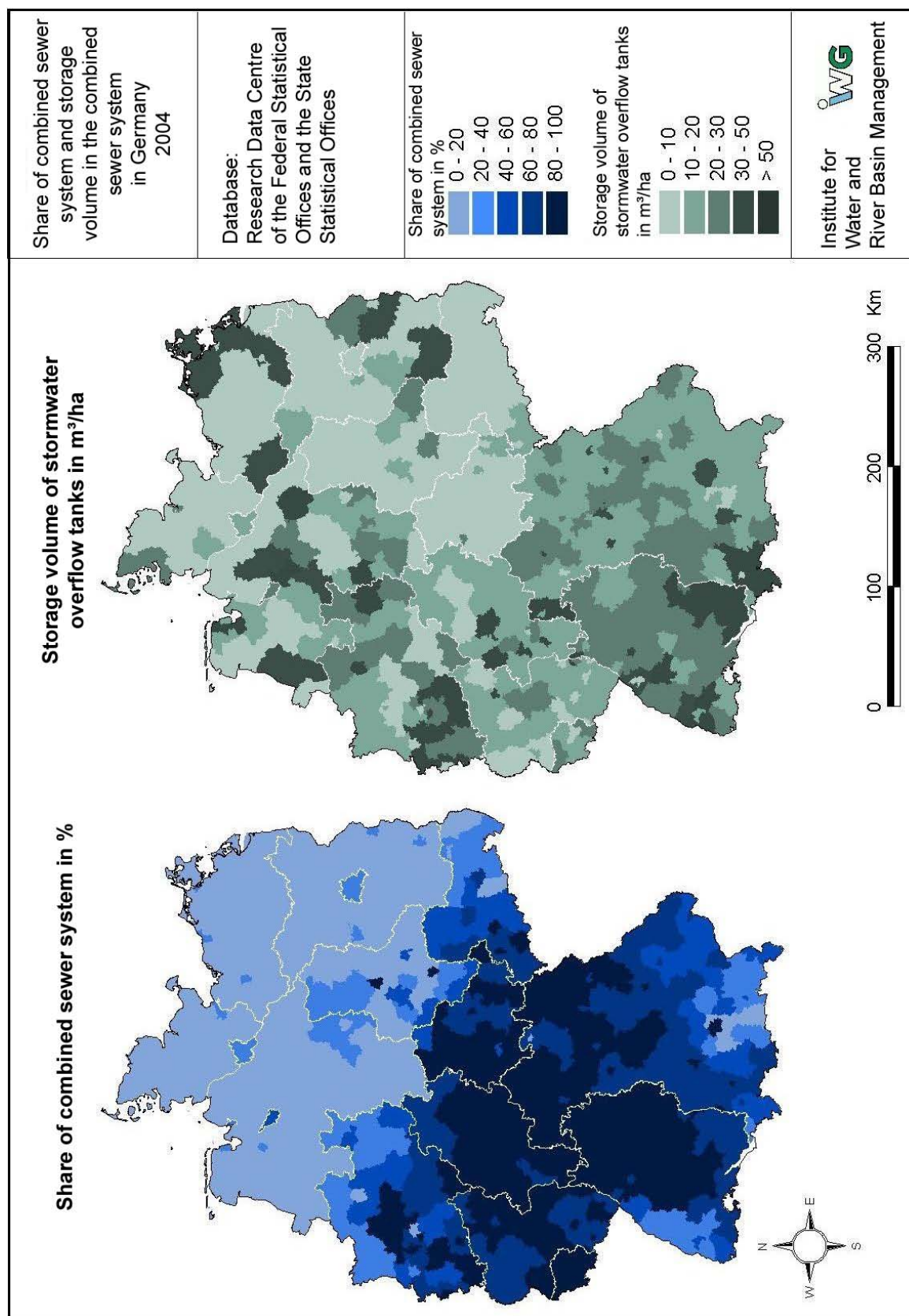


Figure 18: Share of combined sewer system and storage volume of stormwater tanks in the combined sewer system

The specific storage volumes based on the impervious areas and connected to the combined system are needed for the calculations in the model. The urban areas are charted in the local statistic on a district level (STATISTIK LOKAL (2004)). With the approach from HEANEY ET AL. (1976) the degree of impervious area was calculated on the basis of the population density (Statistik lokal) in the districts. Based on the occurrence of combined sewer system (comp. Figure 18 left) the specific storage volume was calculated for every district. The result for the specific storage volumes in the year 2004 on a district level is shown in Figure 18 (right). The temporal change was calculated on the basis of the information from the Federal Statistical Office (1987, 1991, 1995, 1998, 2001, 2004) on a federal state level (comp. Annex Tab.-Ann. 5).

There is no specific information for countries outside Germany on exact area-specific volumes for stormwater overflow tanks. In Austria an average of 15 m<sup>3</sup>/ha is estimated (ZESSNER-SPITZENBERG, 2007). Switzerland and the Netherlands are expected to have a high extension degree (SIEKER, 2004) which is why a specific volume of 23 m<sup>3</sup>/ha is estimated which corresponds to an extension degree of 100 % according to BROMBACH & MICHELBACH (1998). All other countries are estimated to have a 5% extension degree.

### **3.4 Input data from municipal wastewater treatment plants**

For municipal wastewater treatment plants with a design capacity of  $\geq 2,000$  PT data was requested from the federal state authorities by the German Federal Environment Agency within the framework of the duty to report on implementing the Urban Wastewater Treatment Directive. The coordinates of the location of the MWWTP and discharge point, the design capacity and degree of capacity utilisation in PT, the treated annual wastewater amount and the nutrient effluent loads (comp. paragraph 4.1.1.1) for the reporting date 31<sup>st</sup> December 2005 were collected. Subsequently, the wastewater treatment plants were allocated with the help of the coordinates of the discharge points to the analytical units. In Figure 19 the discharge points for the considered units are shown in variable size classes. In total 4614 units  $\geq 2,000$  PT were recorded in the year 2005.

Data on all municipal wastewater treatment plants in Germany is in the statistics of the public wastewater treatment at the FDZ. This statistic was used to supplement the water amounts and nutrient loads (comp. paragraph 4.1.1.1) from WWTP < 2,000 PT. For reasons of data protection the data can only be published as aggregated data (at least three units). Furthermore the statistic does not include the coordinates of the discharge points but only the municipality of the location of the MWWTP and discharge point, thus making it impossible to pinpoint them to an analytical unit. Data of units < 2,000 PT was therefore aggregated at a district level. In total 5334 units were considered for the year 2004.



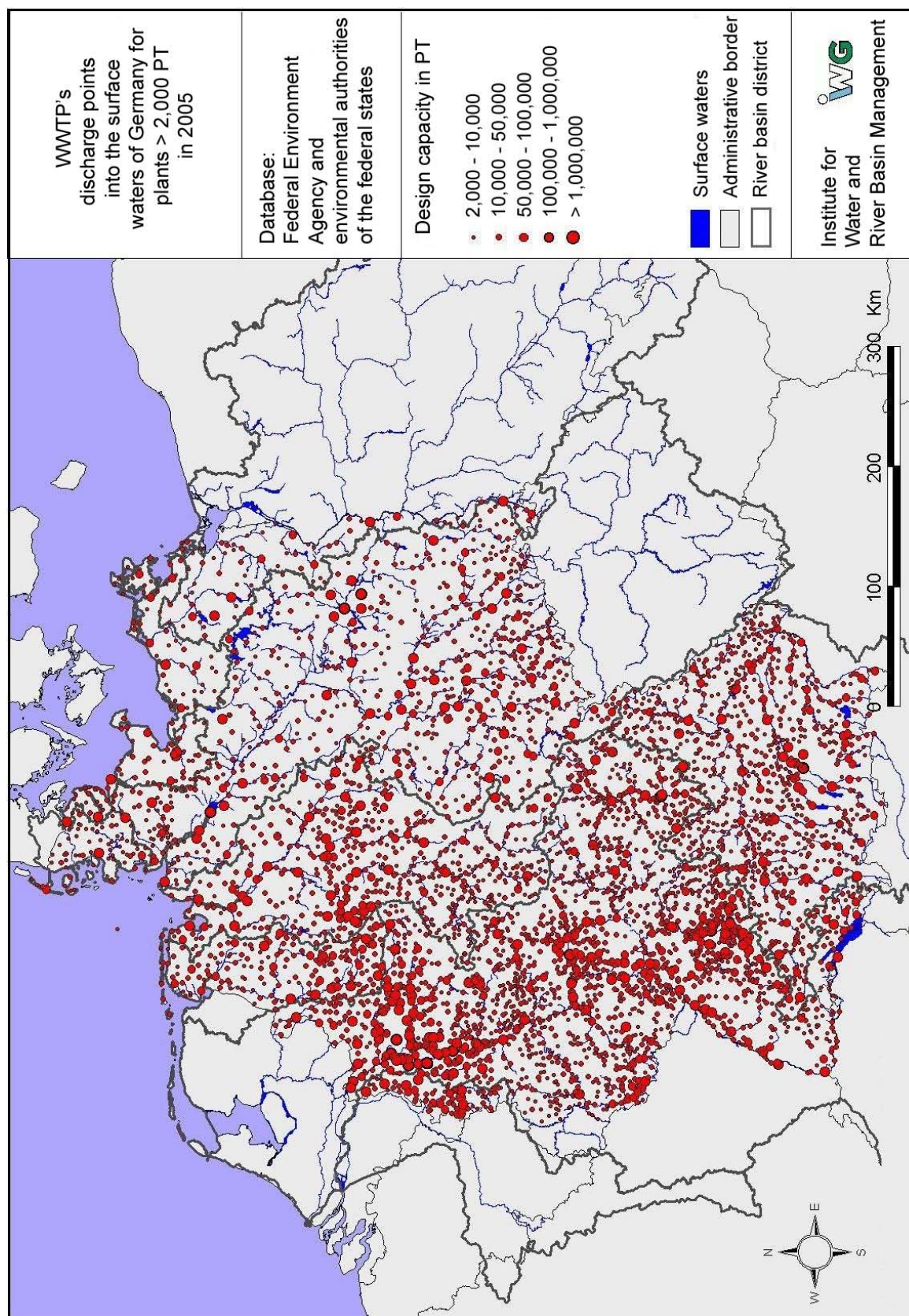


Figure 19: Discharge points of WWTP  $\geq 2,000$  PT in Germany in 2005

Table 3 shows the number, design capacity and the treated annual wastewater amount for MWWTP in different size classes. The wastewater treatment plants < 2,000 PT represent 54 % of all units in Germany, however only 2 % of the population are connected to them and only 2.5 % of all wastewater is treated there.

Table 3: Number, design capacity and treated annual wastewater amount of municipal wastewater treatment plants for different size classes in Germany

<b>WWTP size class (PT)</b>	<b>Number</b>	<b>Design capacity [1,000 PT]</b>	<b>Treated wastewater amount [1,000 m<sup>3</sup>/a]</b>
< 2,000	5,334 (53.6 %)	2,996 (2.0 %)	224,668 (2.5 %)
≥ 2,000 – 9,999	2,381 (23.9 %)	11,044 (7.3 %)	861,359 (7.3 %)
≥ 10,000 – 49,999	1,672 (16.8 %)	38,092 (25.3 %)	2,478,723 (25.3 %)
≥ 50,000 – 99,999	317 (3.2 %)	22,378 (14.9 %)	1,259,629 (14.9 %)
≥ 100,000	244 (2.5 %)	76,169 (50.6 %)	4,182,374 (50.6 %)

## 4 Substance specific input data

The following chapter will outline all substance specific input data for the model system MONERIS. The database is described sorted by pathways for each substance group (nutrients, heavy metals and PAH). The pathways as well as the procedures may differ from group to group.

### 4.1 Nutrients

The method to quantify emissions is best represented in detail with the model system MONERIS using the example of nutrients in flow charts in the annex to this report. Therefore the variable notation will be specified in the following chapters.

#### 4.1.1 Point pathways

##### 4.1.1.1 Municipal wastewater treatment plants

Within the framework of the data collection made by the German Federal Environment Agency on municipal wastewater treatment plants with a design capacity of  $\geq 2,000$  PT the nutrient effluent loads for all units were gathered for the reporting date 31<sup>st</sup> December 2005 (comp. paragraph 3.4). Only a few units delivered no information on their N and P runoff loads to the federal administrations. The missing data was therefore estimated on the basis of average emission factors. For this every federal state missing data had the average inhabitant nutrient loads calculated based on the total number of inhabitants and population equivalents, considering the size class of the units.

Nutrient loads from municipal wastewater treatment plants  $< 2,000$  PT were supplemented by the statistics for public wastewater treatment of the FDZ for the year 2004. For reasons of data protection the effluent loads of the units  $< 2,000$  PT was aggregated on a district level. Subsequently the loads for nitrogen and phosphorus from the districts were assigned to the analytical units area-weighted. The result based on the inhabitants connected to municipal wastewater treatment plants is depicted for the analytical units in Figure 20. High specific emissions per inhabitant mean that a relatively large number of units in these regions has a design capacity of  $< 2,000$  PT. This is especially the case in Schleswig-Holstein, Mecklenburg-Western Pomerania, in parts of Lower Saxony, Rhineland-Palatinate as well as in the north of Hesse, Baden-Wurttemberg and Bavaria.

Table 3 (paragraph 3.4) shows that only 2% of the population are connected to municipal wastewater treatment plants for  $< 2,000$  PT. But these plants still emit 4 % of the nitrogen and 9 % of the phosphorus loads from all municipal wastewater treatment plants. The specific phosphorus load is higher than the nitrogen load as the units  $< 2,000$  PT do not eliminate any phosphorus. Phosphorus elimination is only required for units  $\geq 10,000$  PT.



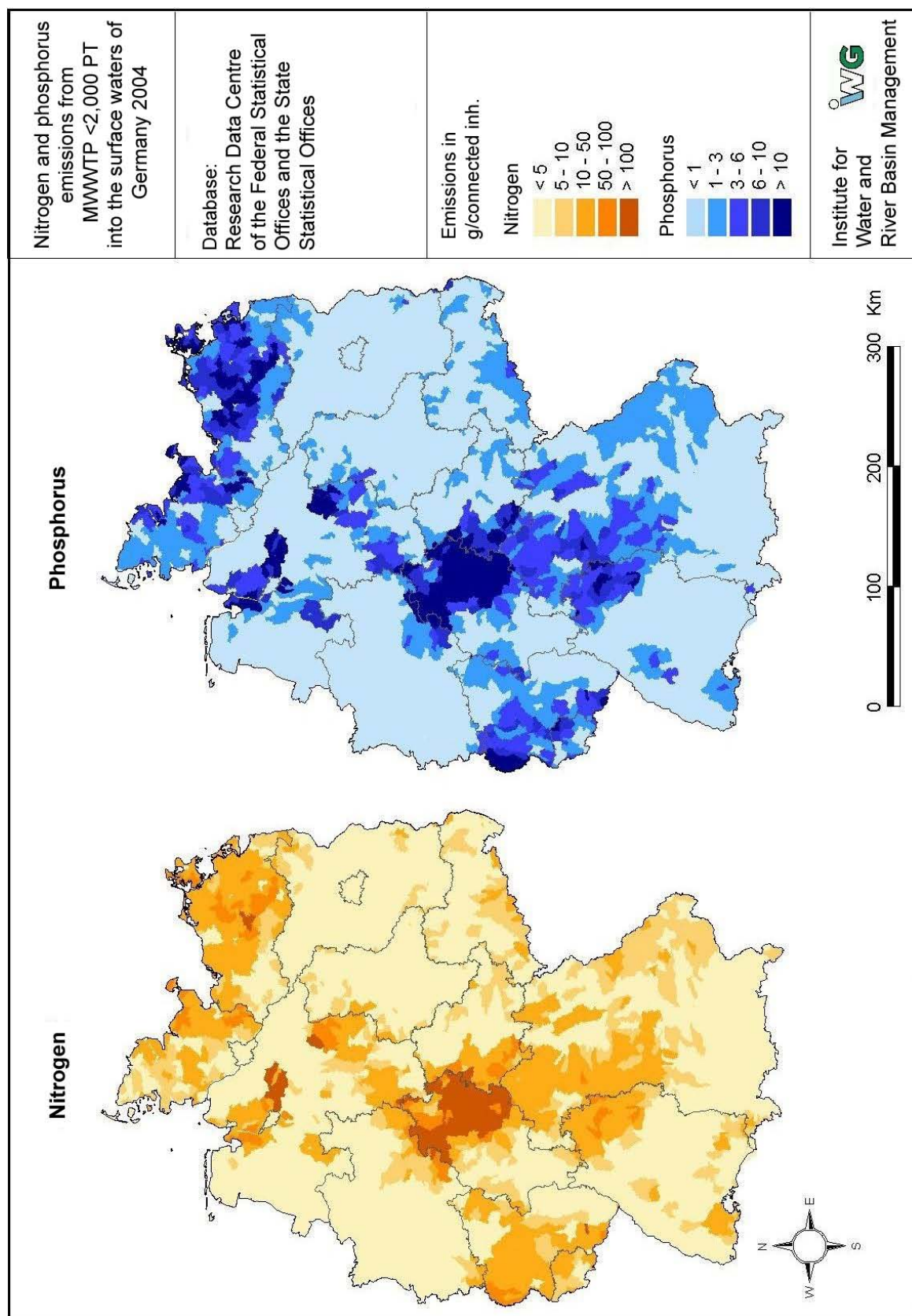


Figure 20: N and P emissions from municipal wastewater treatment plants < 2,000 PT into the surface waters



Data from earlier periods on municipal wastewater treatment plants for the years 1998 and 2001 are in the statistics for public wastewater treatment at the FDZ. Furthermore BEHRENDT ET AL. (1999) calculated nutrient loads from municipal wastewater treatment plants for the years 1985 and 1995. The emissions from both data sets were aggregated on a federal state level and the changing factors regarding the current data sets (2004/2005) were determined. Figure 21 depicts the temporal development of nutrient emission from municipal wastewater treatment plants (1985-2005) in Germany. The emissions for the federal states are in Tab.-Ann. 6 and 7.

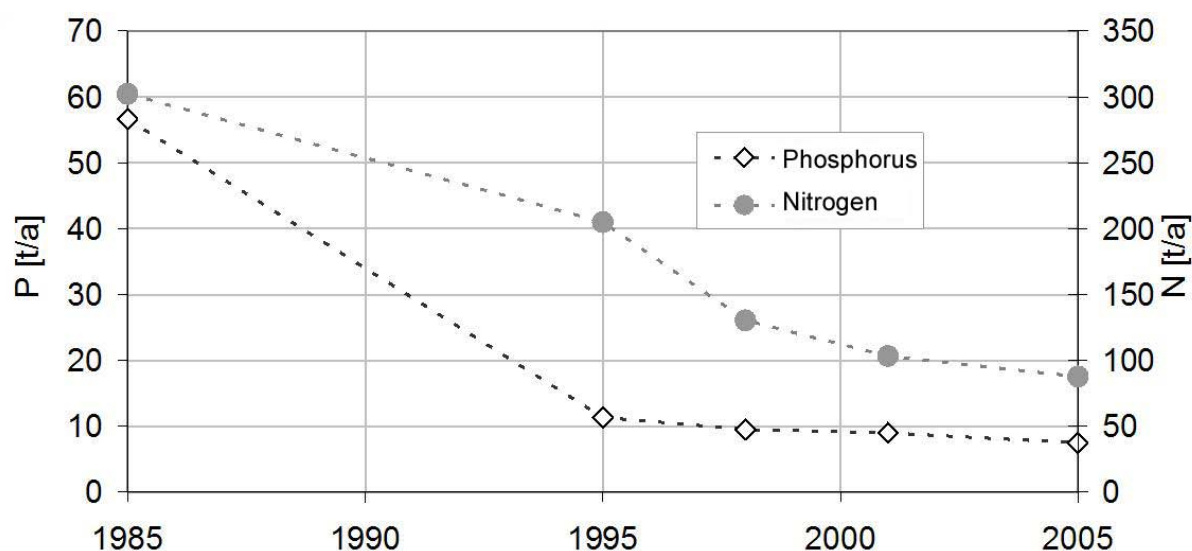


Figure 21: Temporal development of nutrient emissions from municipal wastewater treatment plants in Germany from 1985 to 2005

The main reduction of phosphorus in the emissions from wastewater treatment plants was achieved by phasing out phosphatic detergents from 1986 onwards. The increasing extension of wastewater treatment plants since 1990 especially plays a role for nitrogen.

Based on the detailed spatial data for 2004/2005 and with the help of the temporal changes on the federal state level the emissions in the analytical units were extrapolated for the complete period. This procedure is based on the assumption that the locations of the wastewater treatment plants generally stay the same. This assumption seems plausible for medium-sized to large catchment areas for which the examination of the temporal development of the emissions seems to be of importance.

Neighbouring European countries only partially have precise information on municipal wastewater treatment plants. However, the international river protection commissions (ICPRR, 2005; ICPOR, 2007; ICPER, 2005) have country-specific inhabitant loads from municipal wastewater treatment plants. Based on the amount of inhabitants connected to wastewater treatment plants, these loads were transferred for the analytical units outside Germany.

#### 4.1.1.2 Direct industrial discharges

The European Pollutant Emission Register (EPER) records the emissions of direct industrial dischargers into surface waters above a certain substance-specific threshold. The EPER's

data is reported in every three years and is available for the years 2001 and 2004. The EPER also includes the coordinates of the industrial plants so that the emissions can be precisely allocated to an analytical unit.

On behalf of the German Federal Environment Agency the Fraunhofer Institute for Systems and Innovation Research (ISI) performed a comprehensive survey on the nutrient emissions from direct industrial dischargers for the year 1997 (BÖHM ET AL., 2000). This research also comprised units that were below the EPER threshold. However, there was no data collected on the coordinates of the industrial plants. On behalf of the ISI the addresses of the direct dischargers were used to retrieve the coordinates for the locations. Subsequently the industrial plants were able to be allocated to an analytical unit. A part of the data presented by the federal authorities was made anonymous. In this case only the total emissions and the river basin units were reported. In these cases the loads were aggregated by federal state and by river basin unit and allocated to the last analytical unit in the runoff model of that river basin district.

The IGB has data on nutrient emissions from direct dischargers on river basin level for the year 1985 (BEHRENDT ET AL., 1999). The loads for the federal states were disaggregated from the share of the federal states in the river basins. Subsequently the changing factors concerning the loads for the year 1997 were calculated on a federal state level. Based on the detailed information for the year 1997 and the changes on the level of the federal states the emissions for the period 1983 to 1994 were then estimated for the analytical units.

For 1995-2005 the ISI and EPER data was taken as a basis. As this data could be allocated to an analytical unit, fixed periods were defined and no values interpolated for between these years. Table 4 shows the balance periods and nutrient emissions into surface waters in Germany. The main decrease in direct industrial discharges was between 1985 and 1995, reasons being the improved wastewater treatment in the units themselves (implementation of the requirements according to § 7a Federal Water Act), the closing of several units in the new states of Germany after 1990, the connection of direct dischargers to municipal wastewater treatment plants as well as a reduction of water consumption by the industry which allows a more efficient wastewater treatment. Table 4 depicts the reduction between 1997 and 2001 that is mainly a result of the data collection of the EPER, which only considered larger direct dischargers.

Table 4: Nutrient emissions from direct industrial dischargers into surface waters in Germany

Balancing period	Data collection	Nitrogen [t/a]	Phosphorus [t/a]	Data source
1985-1994	1985	122,350	6,546	IGB
1995-1998	1997	20,615	671	ISI
1999-2002	2001	10,389	371	EPER
2003-2005	2004	8,243	324	EPER

The nutrient emissions from direct industrial dischargers of the European neighbouring countries were investigated in the river basin commissions (ICPRR, 2005; ICPOR, 2007; ICPER, 2005) and were allocated to the analytical units outside Germany depending on the urban area.

Furthermore the nutrient emissions from swamp watering of open-cast mining areas were considered. For the river basin of the Spree there are area-specific emissions (BEHRENDT ET AL., 1999) that were assigned to the analytical units depending on the open-cast mining areas according to CORINE Land Cover (CLC 2000). In 1985 3,457 t/a, 1995 2,592 t/a and 2000 1,409 t/a of nitrogen were emitted into Germany's surface waters. Phosphorus emissions from swamp watering were only considered for the year 1985 with 144 t in Germany. It is assumed that phosphorus emissions from swamp watering have been largely eliminated due to the treatment of the swamp watering water (BEHRENDT ET AL., 1999).

## **4.1.2 Diffuse pathways**

### **4.1.2.1 Atmospheric deposition onto the water surface**

Data on the atmospheric deposition of nitrogen oxides and ammonium with a resolution of 50 x 50 km<sup>2</sup> for the years 1989-2000 were taken from the Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP, 2006). Furthermore, deposition values were available by GAUGER ET AL. (2007) for Germany for the years 1995, 1997 and 1999-2004. But as these values were neither available for all areas nor for all years they were only considered for a comparison calculation for 1999 (comp. paragraph 5.2.2).

Intersecting the deposition data with the borders of the analytical units reveals the average NO<sub>x</sub>-N- and NH<sub>4</sub>-N-deposition within every analytical unit.

The deposition rate of phosphorus, which depends on the land use of the observed area, lies between 0.3 and 3.0 kg P/(ha·a). With the help of statistical data (BEHRENDT ET AL., 2002b) an average of 0.37 kg P/(ha·a) can be derived for European catchment areas. This value was defined as constant for the whole calculation period.

### **4.1.2.2 Nutrient surplus on agricultural areas**

The nutrient surplus on agricultural areas due to mineral fertiliser, manure and atmospheric deposition are an important input data for the quantification of nutrient emissions from agriculture and are therefore considered separately for the nutrients.

The average nitrogen surplus was calculated on a district level for Germany in the reference year 2003 by BACH & SKITSCHAK (2007) (Figure 22). Annual nationwide surpluses were calculated using the OECD method (Organisation for Economic Cooperation and Development, OECD, 2001) with the help of statistical data (FAO, 2007) for catchment areas outside Germany as well as to consider temporal changes. A detailed description of the method used in MONERIS is depicted in the flow chart „model structure“.

The atmospheric deposition must be considered when quantifying the surplus. BACH & SKITSCHAK (2007) used the atmospheric nitrogen deposition by EMEP as well as according to GAUGER ET AL. (2007) that show clear differences in the resulting N-surpluses. To be able to describe the impact of the different deposition values (according to EMEP and GAUGER ET AL., 2007) not just for the nitrogen surplus but also for the resulting nitrogen emissions, a comparative calculation was made for the year 1999 with both input data sets (chapter 5.2.2).

The phosphorus surpluses on the agricultural areas were cumulatively calculated on the level of the federal states for Germany from the year 1955 onward (BEHRENDT ET AL., 1999). The starting value for the mid 1950's was back-calculated on the specifications given by WERNER & WODSACK (1994). For analytical units outside Germany national values are considered according to the nitrogen surpluses.

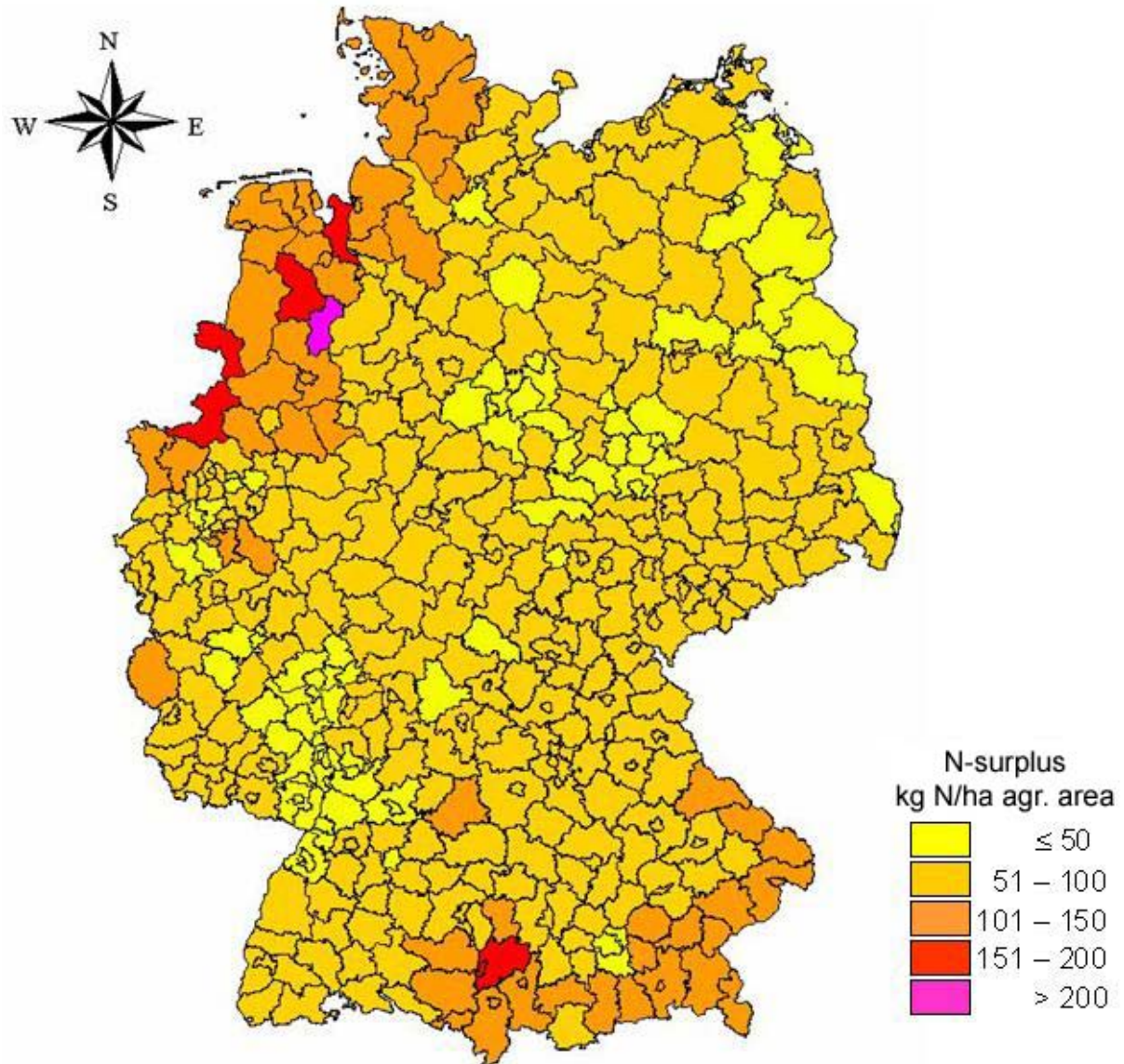


Figure 22: Nitrogen surplus of the agricultural area in the year 2003 for districts and unincorporated towns (BACH & SKITSCHAK, 2007)

#### 4.1.2.3 Erosion

The total phosphorus concentration of the topsoil changes in time and is composed of the basic P-concentration of the soils for the year 1955 and the phosphorus surplus of the agricultural soils. The basic P-concentration is spatially differentiated depending on the clay content of the general soil map (BÜK 1000) (Equation 4-1). The P-surplus is cumulatively avail-

able on the federal state level. (comp. paragraph 4.1.2.2). For the natural erosion of mountain areas only the basic P concentration according to Equation 4-1 is considered.

$$\text{Equation 4-1} \quad P_{\text{agr1955}} = 10.2 \cdot \text{BI}_{\text{SO\_Ccont}} + 150$$

$P_{\text{agr1955}}$  P content of agricultural soils in 1955 [kg/ha]

$\text{BI}_{\text{SO\_Ccont}}$  clay content of topsoil [%]

The N-concentration in the topsoil is derived from the general soil map (BÜK 1000).

The relation between the P-concentration of the suspended solids in rivers with high discharge and the calculated P-concentration of the topsoil supplies the basis to determine the enrichment ratio (EnR). The analysis of the data from the Danube basin shows that the enrichment ratio is inversely shareal to the square root of the specific sediment input (BEHRENDT ET AL., 1999).

#### 4.1.2.4 Surface runoff

When calculating the nutrient emissions from surface runoff from unsealed areas only the dissolved share of nutrient are considered. The nutrient concentration of the surface runoff is calculated as area-weighted mean of the concentrations in the surface runoff for different land use classes. (Table 5, BEHRENDT ET AL., 1999).

Table 5: Total phosphorus (TP) and total nitrogen concentrations (TN) in surface runoff for different land use classes and snow (BEHRENDT ET AL., 1999)

Landnutzungskategorie	Nährstoff	Kurzname Variable	Konzentration (mg/l)
Forest	TP	CSR1	0.035
Open areas	TP	CSR2	0.035
Snow	TP	CSR12	0.010
Farmland	TN	CSR3	0.300
Grassland	TN	CSR4	0.000
Forrest and open areas	TN	CSR5	0.000
Snow	TN	CSR13	0.100

#### 4.1.2.5 Tile drainage

The average P-concentration in drainage water for the analytical units (TD\_TPC) is calculated as the area-weighted mean of the concentrations in Table 6 (BEHRENDT ET AL., 1999) and the allocated areas in the soil map with sandy and loamy soils, fens and bogs.

Table 6: TP-concentrations in the drainage water for different soils (BEHRENDT ET AL., 1999)

Soil	Short name	Concentration (mg P/l)
Sandy soils	CDT3	0.20
Loamy soils	CDT4	0.06
Fen soils	CDT5	0.30
Bog soils	CDT6	10.00

The calculation of the N-concentrations (TD\_TNC) follows the method described by BEHRENDT ET AL. (2000) and is based on the regionally variable N surplus (paragraph 4.1.2.2). The potential nitrate concentration in seepage water according to FREDE & DABBERT (1998) is calculated via the N-surplus, the seepage water amount and an exchange factor that depends on the field capacity of the soils. This potential nitrate concentration in the topsoil is reduced by the denitrification factor that is determined to be 0.85 (BEHRENDT ET AL., 2000). A complete description of the method is shown in the flow chart „Tile drainage“.

#### 4.1.2.6 Groundwater

Figure 23 shows the procedure of calculating the nitrogen concentration in the groundwater inflow. A complete description of the method to quantify nutrient emissions from the groundwater is shown in the flow charts „Groundwater 1, 2 and 3“.

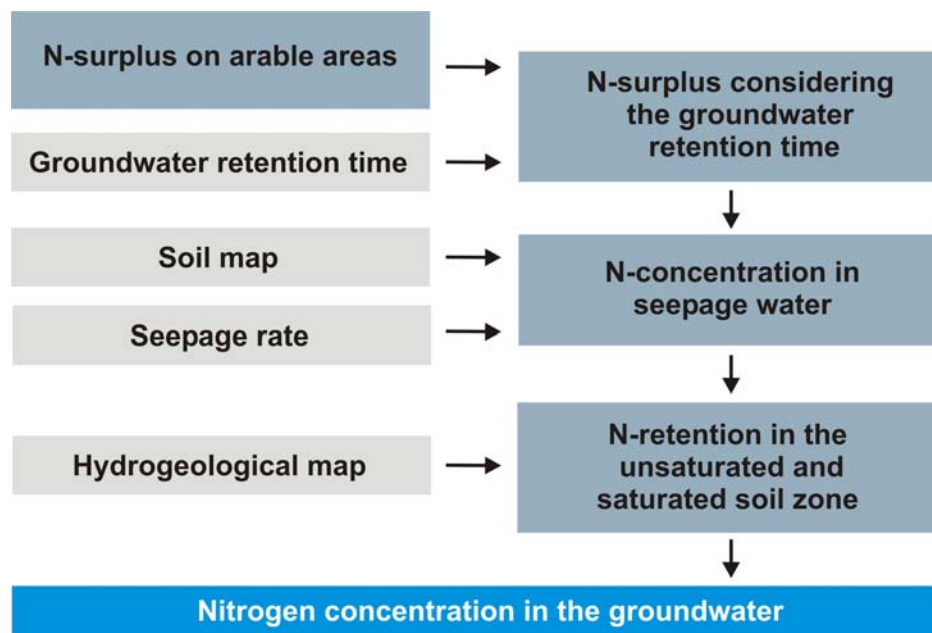


Figure 23: Calculation of nitrogen concentration in the groundwater

The N-concentrations in the groundwater (GW\_TNC) are derived from the potential nitrogen concentrations in the topsoil. As the retention time of water and substances on their way from the root zone to the groundwater and in the groundwater itself can take a long time, the retention period in the groundwater (GW\_RT) has to be considered, too. This is especially meant for considering relevant N-surpluses on arable areas at the time the soil infiltration begins. For German areas the retention time was calculated according to KUNKEL ET AL (2007) (Figure 24). In areas outside Germany the mean retention time according to MONERIS was used.



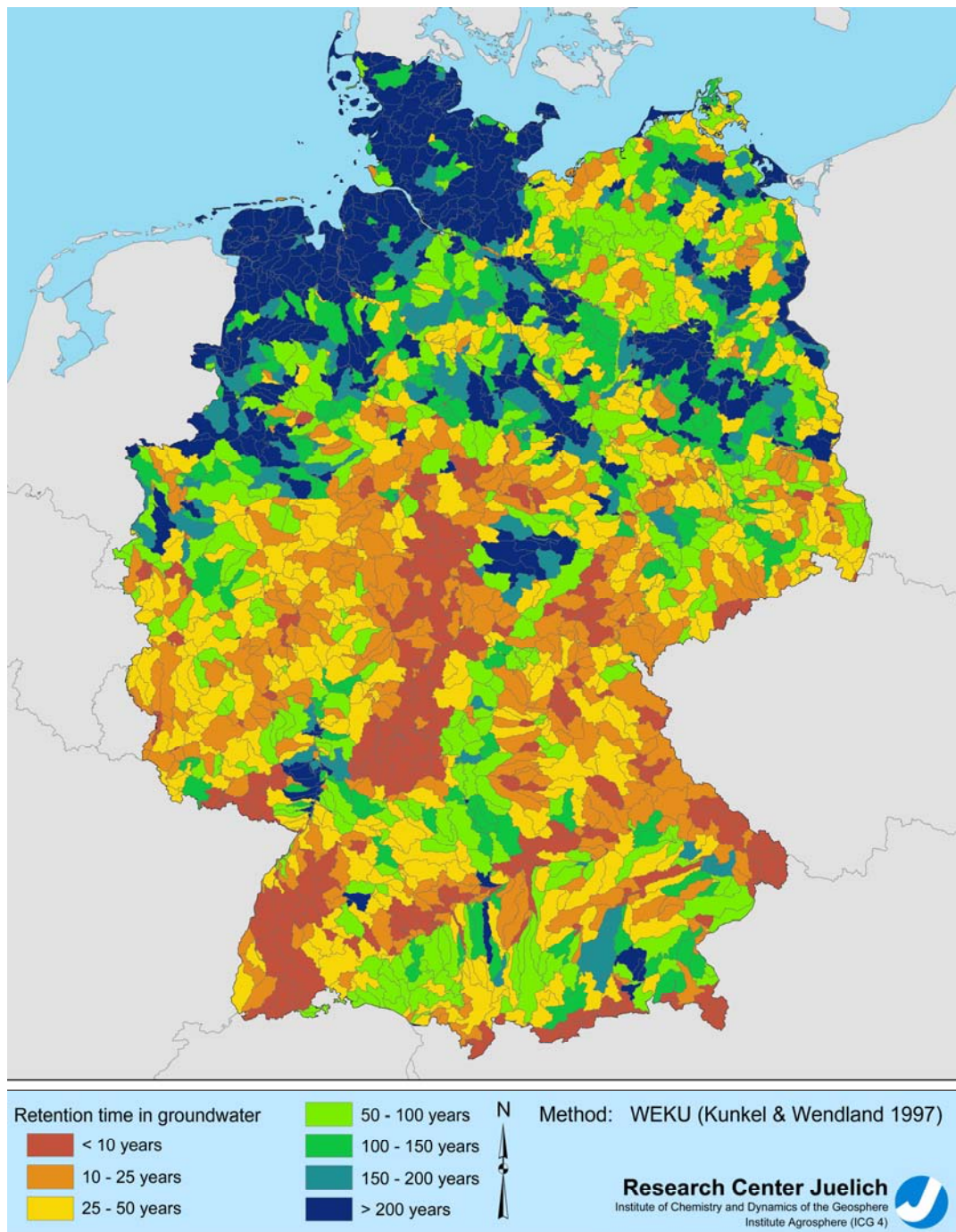


Figure 24: Retention time in upper groundwater aquifers (KUNKEL ET AL., 2007)

The surplus of the analytical units ( $BI\_Nsurpl$ ) is corrected by the ratio of the nationwide N-surpluses ( $CD\_Nsurp\_coun$ ) in the reference year (the year the survey was made) to the average surplus during the retention time in the groundwater. This way an individual average N-surplus can be calculated for each analytical unit during the retention time in the groundwater ( $CD\_Nsurp\_gwres$ ).

Nitrogen retention (especially due to denitrification) in the soil in the unsaturated zone and in the groundwater is calculated from the comparison of regional nitrate concentrations in the groundwater and the potential nitrate concentrations in seepage water. This comparison was made for Germany and shows that the nitrogen retention depends on the seepage rate and

on the hydrogeological conditions (flow chart „Groundwater 2“: G\_NC1, G\_NC2, G\_NC3, G\_NC4).

For the examination of nitrogen retention within a river the DON emissions (Dissolved Organic Nitrogen) are needed. It is assumed that especially the long-chain DON compounds only have a negligibly small retention. DON emissions via the groundwater are calculated by using the groundwater recharge for forest areas and wetlands. The DON-concentrations can be calculated separately for forest areas and wetlands and vary in general between 0 and 6 mg/l. In forest areas values are mainly lower than in wetlands and can fall to 0 mg/l in warmer climates (VENOHR, 2006).

The details on groundwater concentrations of soluble reactive phosphorus (SRP) for the different soil types are according to BEHRENDT ET AL. (2000) (Table 7). Using these values, the P-concentration in the analytical units was calculated on the basis of the concentrations and area shares of sandy and loamy soils, fens and bogs as area-weighted mean for arable areas.

Table 7: Concentrations of soluble reactive phosphorus (SRP) in the groundwater for different land use and soils

Land use	Soil	Short name	Concentration (mg P/l)
Agricultural areas	Sandy soils	CGW4	0.10
Agricultural areas	Loamy soils	CGW5	0.03
Agricultural areas	Fen soils	CGW6	0.10
Agricultural areas	Bog soils	CGW7	2.50
Forestal areas / open areas	No differentiation	CGW3	0.02

Furthermore, possible concentration differences between soluble reactive phosphorus and the total phosphorous in anaerobic groundwater must be considered (GW\_TPC) (DRIESCHER & GELBRECHT, 1993). The concentration of TP in aerated groundwater is equal to the SRP-concentration but the difference between TP and SRP should be considered for anaerobic groundwater. With the help of standardised monitoring programmes it can be deduced that the total P-concentration is twice to five times higher than the SRP concentration according to BEHRENDT (1996) und DRIESCHER & GELBRECHT (1993). As there is no information available on areas with anaerobic groundwater the areas with a higher probability of anaerobic conditions are determined by the comparison of nitrate concentrations in the groundwater and the seepage water (GW\_CR\_TN). For the calculation of the total phosphorus concentrations in the groundwater (GW\_TPC\_corr) it was defined that in case the nitrogen concentration in the groundwater passes a critical value (CGW31 = 0.1 mg/l) the TP concentrations in the groundwater are 2.5 times higher (CGW2) as the SRP concentrations.

#### 4.1.2.7 Sewer systems

According to BROMBACH & MICHELBACH (1998) the area specific load from impervious urban areas for P is 2.5 kg P/(ha·a) (CUS10). The area specific load for N results from the sum of atmospheric N deposition as well as leaf fall and animal excrement (4 kg N/(ha·a); CUS13).



The specific dissolved nutrient load of humans is 9 g N/(E·d) (CUS17) for nitrogen. For phosphorus it has to be assumed that the dissolved loads are different in every country due to differences in the use of phosphates in detergents. Therefore country-specific phosphorous levels are used for inhabitant-specific loads (CD\_Pinh\_coun) and for detergents (CD\_Pdet\_coun) in MONERIS.

For nutrient concentrations from commercial wastewater a value of 2 mg/l (nitrogen, CUS12) and 0.5 mg/l (phosphorus, CUS9) was taken as a basis (BEHRENDT ET AL., 2000).

## **4.2 Heavy metals**

### **4.2.1 Point pathways**

In addition to the point pathways of municipal wastewater treatment plants and direct industrial dischargers emissions from historic mining activities have to be considered for the heavy metals.

#### **4.2.1.1 Municipal wastewater treatment plants**

The quantification of heavy metal emissions from municipal wastewater treatment plants is based on the amount of treated sewage and the heavy metal concentrations in the wastewater treatment plant's effluent (comp. chapter 2.5.1.1). For this the current effluent concentrations (from the year 2001 onwards) were researched from the federal state authorities. Furthermore the earlier data collections from the years 1993-1997 and 1999/2000 on heavy metal effluent concentrations (BÖHM ET AL., 2001; FUCHS ET AL., 2002) were used.

Within the framework of the first data collection for the balance period 1993-1997 measurement readings for all federal states were retrieved (BÖHM ET AL., 2001). However, the data strongly varied in amount and quality. While data was reported for 14 federal states in 1999/2000 (FUCHS ET AL., 2002), for the current inquiry (2000-2005) only 9 federal states provided data. An inquiry at the federal authorities showed that heavy metals are very seldom measured in the effluent from wastewater treatment plants, as the threshold values of the Wastewater Charges Ordinance are generally not exceeded. Some of the data is not even added to a database so that it is very elaborate for the authorities to hand them out. Since the year 2008 municipal wastewater treatment plants with a design capacity above 100,000 PT are potentially liable to report heavy metals emissions within the framework of the PRTR (Pollutant Release and Transfer Register). It is therefore assumed that heavy metals are going to be measured more in the future in the effluent from wastewater treatment plants, at least for large plants.

For the quantification of heavy metal emissions from municipal wastewater treatment plants especially the large spectrum of given limits of quantification of effluent concentrations is causing significant uncertainties. As measurement readings below the limit of quantification are included in the calculation with half of the value it is necessary to exclude limits of quantification that are not analytically justified. On the basis of the actually measured value spectrum a maximum limits of quantification for every metal was defined by BÖHM ET AL. (2001) (Table 8).

Table 8: Maximum allowed limit of quantification for heavy metals in effluent from wastewater treatment plants (BÖHM ET AL., 2001)

in $\mu\text{g/l}$	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Limit of quantification	0.5	5.0	20.0	0.2	10.0	6.0	100

Figure 25 shows an exemplary spectrum for the given limits of quantification for lead. This varies between 0.1 and 100  $\mu\text{g/l}$ . The maximum allowed limit of quantification found was 6  $\mu\text{g/l}$ , Table 8). All limits of quantification above 6  $\mu\text{g/l}$  were therefore not considered. As additional quality criteria it was set that at least 10 % of a federal state's measurement readings have to lie above the limit of quantification to be included in the calculation of the emissions.

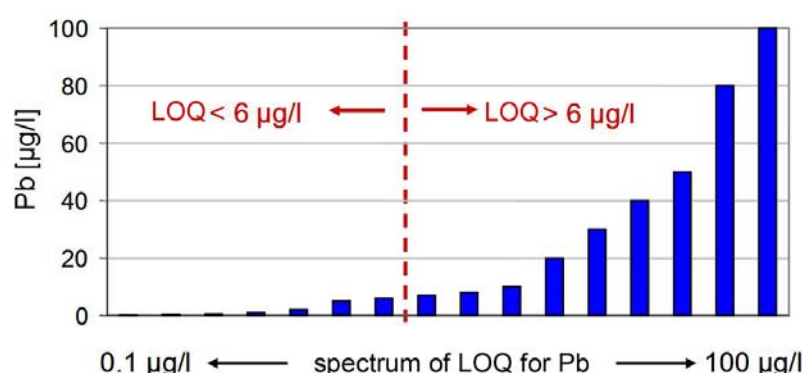


Figure 25: Spectrum of the given limits of quantification for lead

After correcting the data a mean heavy metal concentration in the effluent of wastewater treatment plant was calculated on the level of each federal state. In average it is assumed that the effluent concentrations have decreased as the treatment efficiency for heavy metals in the wastewater treatment plants was improved (e.g. with biological wastewater treatment and precipitation). On the other hand the inflow of most metals into wastewater treatment plants has been reduced due to measures taken in the catchment (e.g. pre-treated emissions from indirect dischargers, a decrease in emissions from impervious areas in combined sewer systems). Nevertheless, scattered increases of effluent concentrations appeared in the observation period 1993-2005. The following causes were detected:

- Some of the federal states conducted special monitoring programmes (especially in the coastal regions) operating with very low limits of quantification. If later monitoring periods used analysing methods with higher limits of quantification a large share of the measurement readings were below those limits of quantification and therefore those average effluent concentrations are less reliable. In these cases the concentrations from the special monitoring programmes were kept for the later balance periods.
- Some federal states had measurement readings for 1993-1997 and 2001-2005 but not for the period in between. In these cases the heavy metal concentrations for 1999/2000 were interpolated from the data available.

If there was no data available or if a dataset for a federal state did not come up to the defined quality criteria it was considered with the inhabitant-weighted average for Germany. The resulting means are shown in Table 9. For the balance period 1993-1997 it is distinguished between the old and the new states of Germany as the effluent concentrations were significantly different.

Table 9: Mean heavy metal concentrations in the effluent of wastewater treatment plants for Germany

in µg/l	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1995 (1993-1997) west <sup>1</sup>	0.25	5.08	13.21	0.19	7.86	3.29	70.53
1995 (1993-1997) east <sup>1</sup>	0.76	8.78	15.53	0.37	13.06	7.64	93.66
2000 (1999/2000) <sup>2</sup>	0.20	3.30	11.77	0.13	7.46	2.82	46.85
2005 (2001-2005) <sup>3</sup>	0.15	2.25	9.05	0.10	5.05	1.73	47.29

<sup>1</sup> BÖHM ET AL. (2001), <sup>2</sup> FUCHS ET AL. (2002) revised, <sup>3</sup> current data collection

For the period 1983-1987 there are no reliable measurement readings from the effluent of the wastewater treatment plants. The emissions from this period could therefore only be estimated by the change of the heavy metal content in the sewage sludge from 1985 to 1995. This was also split into old and new states of Germany (FUCHS ET AL., 2002).

At first the heavy metal emissions from municipal wastewater treatment plants were calculated for the reference year 2005 with the help of the treated wastewater amount (paragraph 3.4) and the current effluent concentrations from the federal states. Subsequently the emissions were calculated on a federal state level from the datasets for all reference years (1985, 1995, 2000 and 2005) (Table 10 shows the aggregated emissions for Germany). Analogue to the procedure for nutrients (paragraph 4.1.1.1) the temporal changing factors on a federal state level were calculated. Based on the spatially detailed data for 2005 and on the basis of the temporal changing factors, the effluent loads in the analytical units were extrapolated for the reference years and subsequently the data for the years in between was interpolated. This procedure is based on the assumption that the locations of the wastewater treatment plants generally stay the same. This assumption seems plausible for medium-sized to large catchment areas for which the examination of the temporal development of the emissions seems to be of importance.

Table 10: Heavy metal emissions in kg/a from municipal wastewater treatment plants in Germany

in kg/a	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1985	7,120	118,162	161,580	2,919	142,586	65,172	1,014,181
1995	2,991	54,304	133,705	2,044	84,126	36,749	726,449
2000	2,075	34,002	121,259	1,276	77,452	28,752	489,090
2005	1,390	20,914	82,936	939	46,030	16,004	436,267

For catchment areas outside Germany there is hardly any information on heavy metal emissions from municipal wastewater treatment plants. With the help of heavy metal emissions

for Germany and the number of inhabitants connected (paragraph 3.3.8) emission factors for the individual years were calculated. Subsequently, by means of the number of inhabitants connected the emissions for the analytical units outside Germany were estimated.

#### 4.2.1.2 Direct industrial discharges

Emissions from direct industrial dischargers were, deviating methodically from the surveys of earlier projects (FUCHS ET AL., 2002; BÖHM ET AL., 2001), not requested from the companies discharging directly. Instead the tables available at the Fraunhofer ISI for the federal states was extended by the results of a survey made for the European Pollutant Emission Register for the year 2004 and supplemented by sending it via the Federal Environment Ministry (BMU) to the federal states requesting them to update the data. The tables sent back from the federal states were used as a basis for the compilation of direct industrial dischargers. In addition further relevant publications were evaluated for the year 2004, e.g. the reports of river basins commissions. The current data includes the coordinates of the discharge points so that the emissions can directly be allocated to the appropriate analytical unit. The resulting loads into the river basins for the year 2004 are shown in Table 11.

Table 11: Heavy metal emissions from direct industrial dischargers in kg/a into Germany's river basins in the year 2004

in kg/a	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Danube	2	81	84	0	40	78	10,814
Rhine	319	12,047	24,900	96	11,344	11,449	48,189
Meuse	20	0	0	0	50	29	2,340
Ems	0	0	795	0	0	69	395
Weser	23	420	470	6	872	211	5,242
Elbe	167	3,163	2,543	26	3,720	2,143	33,294
Odra	38	54	330	2	190	343	17,512
North Sea (Eider)	0	3,320	12	0	9	0	109
Baltic Sea <sup>1</sup>	0	6	26	0	19	29	316

<sup>1</sup> loads into the river basin districts Schlei/Trave and Warnow/Peene

In principle the database of the projects mentioned above can be used back to the year 1983 for the allocation to the analytical units. For this the coordinates of each production location of the companies needs to be specified so they can be allocated to the appropriate analytical unit. This is done in several iteration steps for approximately 2000 datasets. In some cases the company's location was not known, for example

- as the firm does not exist any more and only the federal state and/or the river basin is known, for example in many of the GDR's people-owned enterprises (VEB),
- as for reasons of data protection only the river basin of a discharger was given by the surveillance authorities,
- as in the past only summary emissions were published, for example the emission from the chemical industries into the Rhine in 1985.

Follow-up surveys were made when possible. Anonymous emissions were allocated to the last analytical unit of the given river basin in the federal state.

For companies in the east of Germany for which the emissions were known for the reference year 1985 proportional changes were made in the analytical units up to the value of the year 1995. If available the direct industrial discharges of the upstream riparians of the Elbe, Rhine and Odra outside Germany were compiled for the base years 1985, 1997, 2000 and 2004. In addition bibliographical references and especially publications by the river basin commissions and the EPER were analysed.

Emissions of direct industrial dischargers were summarised considering the reference years for the balance periods 1983-1987, 1993-1997, 1998-2002 and 2003-2005 (Table 12).

With the exception of cadmium and chrome the direct industrial emissions were reduced again for 2003-2005 compared to the period 1998-2002.

Table 12: Heavy metal emissions in kg/a from direct industrial dischargers in Germany

in kg/a	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1983-1987 <sup>1</sup>	21,350	459,640	398,490	21,710	178,780	124,800	2,814,300
1993-1997 <sup>2</sup>	730	33,170	31,100	230	28,940	20,670	192,410
1998-2002 <sup>3</sup>	490	17,750	33,500	140	19,440	15,800	125,940
2003-2005	570	19,090	29,160	130	16,240	14,350	118,210

<sup>1</sup> FUCHS ET AL. (2002), <sup>2</sup> BÖHM ET AL. (2001), <sup>3</sup> FUCHS ET AL. (2002) updated

#### 4.2.1.3 Abandoned mining

While active mining is under the surveillance of the water authorities, historic mining locations are abandoned for several centuries but discharge punctually large loads of heavy metals due to gallery draining. These are characterized by low concentrations but large water amounts.

Within the framework of the research project the Fraunhofer ISI organised an expert workshop with a result that emphasised the local/regional importance of this pathway but also showed that the federal states only have insufficient data. Reasons being the uncertain administrative responsibilities, missing registration of the location of adit entrances and missing measurement readings. At the expert workshop it was agreed to enquire the loads from abandoned mining from the federal states, leading to 123 galleries that could be listed. For 57 of these the emissions could be calculated and were allocated to the appropriate analytical unit via the geographical coordinates. It can be assumed that the larger discharges have been completely considered whereas the extent of the smaller discharges cannot be estimated.

Table 13 shows the recorded emissions. It was assumed that the loads do not vary over the complete observation period.

A reduction of emissions in the next years is anticipated for started (Wismuth Bergbau, Saxony) and planned (Burgfeyer gallery, North Rhine-Westphalia) remediation and treatment procedures.

Table 13: Registered heavy metal emissions in kg/a from abandoned mining

Federal state	Number of mining galleries		Cd	Cr	Cu	Hg	Ni	Pb	Zn
			Registered emissions in kg/a						
Baden-Württemberg	10 <sup>1</sup>	3 <sup>2</sup>	1						409
Bavaria	3 <sup>1</sup>	0 <sup>2</sup>							
Lower Saxony	15 <sup>1</sup>	0 <sup>2</sup>							
North Rhine-Westphalia	7 <sup>1</sup>	1 <sup>2</sup>	71				13,500	146	31,200
Rhineland-Palatinate	17 <sup>1</sup>	9 <sup>2</sup>	0	81	7,585	0	2,026	208	466
Saarland	3 <sup>1</sup>	2 <sup>2</sup>	1	58	306	1	58	158	
Saxony	23 <sup>1</sup>	18 <sup>2</sup>	1,359	0	2,118	0	242	1,321	120,358
Saxony-Anhalt	44 <sup>1</sup>	23 <sup>2</sup>	582	125	3,921	9	1,221	7,185	218,402
Thuringia	1 <sup>1</sup>	1 <sup>2,3</sup>							
Germany	123 <sup>1</sup>	57 <sup>2</sup>	2,014	264	13,930	10	17,046	9,017	370,835

<sup>1</sup> number of total registered galleries, <sup>2</sup> number of galleries with data on the loads, <sup>3</sup> only arsenic

Figure 26 shows the location and relevance of the recorded abandoned mining discharges exemplary for cadmium (Cd). Considerable loads are discharged into the surface waters at the locations in Saxony (Erz Mountains), North Rhine-Westphalia (Eifel) and Saxony-Anhalt (Harz).

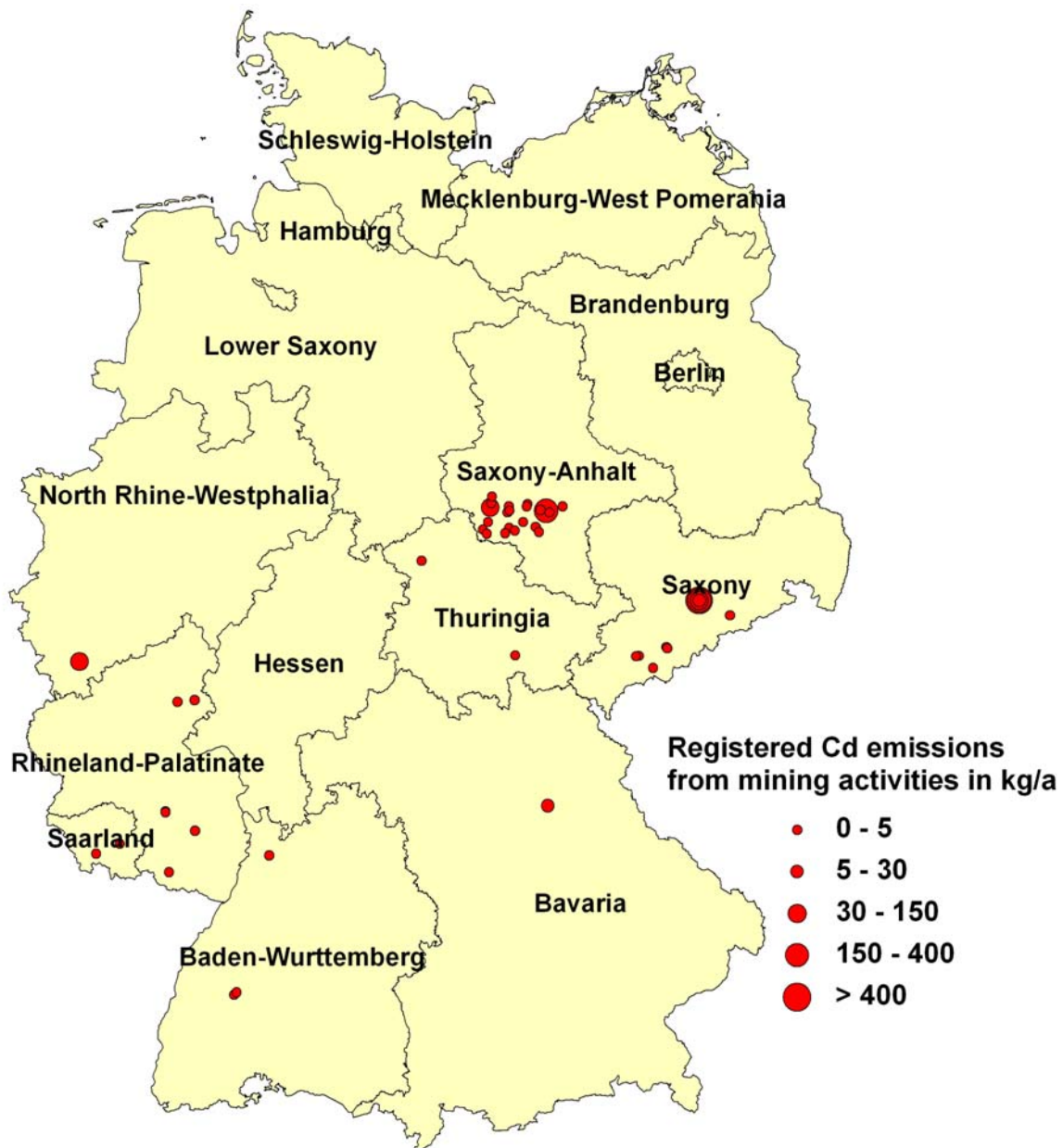


Figure 26: Recorded discharge points and loads from abandoned mining locations exemplary for cadmium (Cd)

## 4.2.2 Diffuse pathways

### 4.2.2.1 Atmospheric deposition onto the water surface

Within the framework of the "Co-operative programme for monitoring and evaluation of the long range transmission of air pollutants in Europe" the Meteorological Synthesising Center East (MSC-East, Moscow) models the atmospheric deposition for the priority heavy metals Cd, Hg and Pb in a 50 x 50 km grid for Europe. Till now the data from 1996-2004 is available. Figure 27 shows the deposition rates in Germany for the year 2004. The grid maps were intersected with the analytical units and the atmospheric deposition rate for the three metals was calculated for every year from 1996-2004. The atmospheric deposition for the year 2005 was taken from 2004.

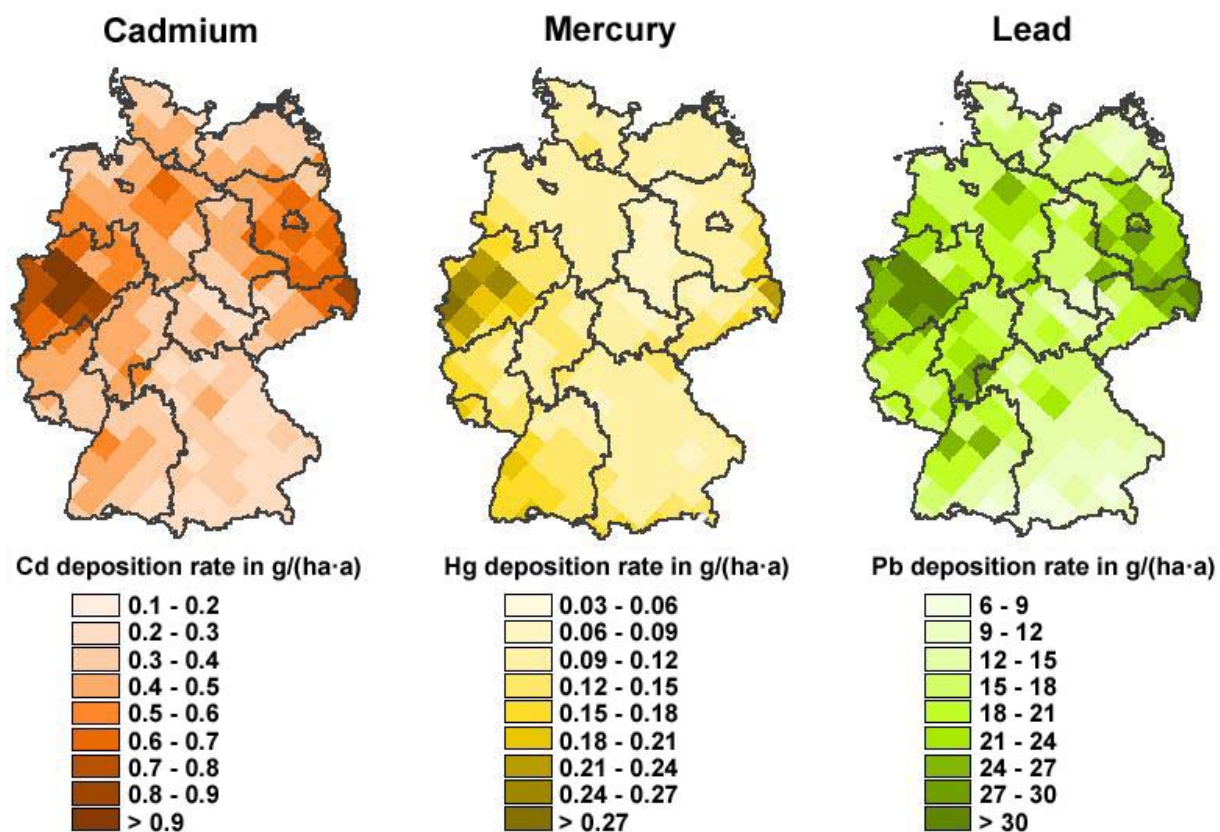


Figure 27: Deposition rates for cadmium, mercury and lead in 2004 (EMEP, 2007a)

The German Federal Environment Agency (BIEBER, 2007) observes the atmospheric deposition („bulk deposition“) for heavy metals at two monitoring stations in Germany (Waldhof, Lower Saxony and Deuselbach, Rhineland-Palatinate). Statistical series exist for Cd, Cu and Pb since 1989 and for the other metals since 1994/1995. For Hg only the wet deposition is recorded since 1993. As the total deposition is not known only the EMEP data was considered for Hg. For Cd, Cu and Pb an average was calculated for Germany from both monitoring stations.

The deposition rates for the mid 1980s was supplemented by further bibliographical references. A compilation of the data is found at FUCHS ET AL. (2002). Due to the different emission situations in the old and new states of Germany they were dealt with as two parts up until 1989. Table 14 shows the considered average deposition rates. The data for the missing years was interpolated. For the area of the former GDR it was assumed that the level of atmospheric deposition did not change in the years 1983 to 1989.



Table 14: The average deposition rate for heavy metals in Germany [g/(ha-a)]

in g/(ha-a)	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1983-1987 West <sup>1</sup>	3.40	6.00	68.00	1.20	12.30	131.0	385.0
1983-1989 East <sup>1</sup>	41.00	7.90	131.00	2.20	41.50	153.0	730.0
1989	2.64	-	22.93	-	-	41.53	-
1990	3.76	-	16.51	-	-	32.76	-
1991	1.28	-	17.31	-	-	26.09	-
1992	2.34	-	30.07	-	-	29.43	-
1993	1.42	-	27.47	-	-	22.27	-
1994	1.31	2.96	25.96	-	11.56	30.29	-
1995	1.07	2.86	21.70	-	5.74	27.54	205.71
1996	EMEP	1.52	22.49	EMEP	5.10	EMEP	299.79
1997	EMEP	2.84	30.34	EMEP	9.59	EMEP	227.00
1998	EMEP	2.14	23.17	EMEP	9.32	EMEP	143.78
1999	EMEP	2.33	11.98	EMEP	10.43	EMEP	230.04
2000	EMEP	2.53	25.02	EMEP	6.45	EMEP	160.09
2001	EMEP	1.41	21.70	EMEP	5.10	EMEP	145.45
2002	EMEP	1.61	19.41	EMEP	4.49	EMEP	93.61
2003	EMEP	1.24	13.37	EMEP	3.14	EMEP	99.68
2004	EMEP	-	-	EMEP	-	EMEP	-

<sup>1</sup> FUCHS ET AL. (2002)

All metals show considerable reductions of atmospheric deposition from 1983 to 2005, the largest reductions being in the east of Germany as after 1990 many out-of-date combustion and industrial plants were closed or reconditioned to meet the current state of art.

Analytical units outside Germany were also covered by EMEP. The deposition rates of Germany were used for all other metals.

#### 4.2.2.2 Erosion

The Working Group of the Federal States on Soil (LABO, 2003) compiled background data for heavy metals in agricultural topsoils on a federal state level. Some federal states have differentiated the data by geological conditions or intensity of use of the agricultural site. In this case the average concentration was calculated for the federal states, the individual values being weighted with the number of samples taken for considering the share of arable land of a federal state for the different use intensities and geological units respectively. The measurement values were mainly taken in the 1990s (LABO, 2003) and were therefore assumed representative for the reference year 1995. Table 15 shows the resulting average heavy metal contents for the federal states.

Table 15: The average heavy metal contents for agricultural topsoils for the reference year 1995 (determined according to LABO, 2003)

Angaben in mg/kg	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Baden-Wurttemberg	0.20	36.00	19.00	0.10	27.00	27.00	60.00
Bavaria	0.23	54.87	17.27	0.10 <sup>1</sup>	28.20	31.30	68.31
Berlin	0.15	2.20	10.00	0.03 <sup>2</sup>	0.80	22.00	16.00
Brandenburg	0.10	4.00	4.20	0.03	2.00	11.81	15.24
Bremen	0.10	8.00	6.00	0.04	2.00	17.00	17.00
Hamburg	0.40	30.00	30.00	0.20	15.00	70.00	120.00
Hesse	0.42	17.26	24.30	0.05	65.04	32.85	81.26
Mecklenburg-W. Pomerania	0.10	12.94	13.03	0.05	7.56	13.03	28.85
Lower Saxony	0.21	23.84	11.38	0.06	14.60	18.64	47.89
North Rhine-Westphalia	0.42	25.43	11.60	0.07	15.26	28.45	67.59
Rhineland-Palatinate	0.25	33.69	19.29	0.11	35.20	31.74	79.62
Saarland	0.32	27.55	14.60	0.07	21.25	28.82	74.53
Saxony	0.44	42.16	16.84	0.10	17.16	49.34	71.83
Saxony-Anhalt	0.15	17.86	9.16	0.08	12.61	19.97	40.62
Schleswig-Holstein	0.10	13.83	8.35	0.04	8.53	13.65	36.66
Thuringia	0.21	49.50	19.94	0.08	26.58	29.70	67.72

<sup>1</sup> value taken over from Baden-Wurttemberg, <sup>2</sup> value taken over from Berlin

Long-term accumulation of heavy metals in agricultural topsoils is a result of atmospheric deposition and agricultural cultivation (fertilisation). A balance was made for the input and output for the agricultural topsoils for considering this accumulation over the complete observation period 1983 to 2005 (FUCHS ET AL., 2002). On the input side the fertilising (mineral fertiliser and manure as well as sewage sludge) and atmospheric deposition was balanced. The most important output for heavy metals from arable land is the withdrawal on account of harvesting, eluviation by seepage water and surface runoff. Table 16 shows the considered average heavy metal accumulation on arable land. The balance was comprised for before and after the reference year 1995. It is apparent from Table 16 that the accumulation for the balance period after 1995 is lower than before 1995 which is attributed to the reduction of input from atmospheric deposition and fertiliser.

Table 16: Average annual heavy metal accumulation in agricultural topsoils for the periods 1983-1995 and 1995-2005 [ $\mu\text{g}/(\text{kg}\cdot\text{a})$ ]

Heavy metal accumulation		Cd	Cr	Cu	Hg	Ni	Pb	Zn
1983-1995	[ $\mu\text{g}/(\text{kg}\cdot\text{a})$ ]	0.88	22.20	51.60	0.26	0.98	19.30	206.00
1995-2005	[ $\mu\text{g}/(\text{kg}\cdot\text{a})$ ]	0.50	7.74	36.50	0.13	0.00	9.18	163.00

During the erosion process heavy metals are accumulated in the eroded sediments due to the preferential transport of fine particles. The enrichment ratio (EnR) is described by the ratio of a substance in the eroded matter and in the agricultural topsoil. FUCHS ET AL. (2002) defined the enrichment ratio for 16 medium-sized catchment areas of different regions in

Germany. The heavy metal contents in the agricultural topsoils in the catchment areas were calculated under consideration of the geological conditions (Digital Geological Map of Germany, BGR, 2001 and LABO, 1998). Data on the heavy metal contents in suspended solids is available at the UBA (1999). However, an evaluation of the data on the basis of the runoff conditions for considering effective erosion events was not possible as the number of measurements was too low (approx. 12-24 p.a.). For metals where erosion is not the most important pathway other sources (e.g. urban areas) cannot be excluded for particulate transported heavy metals into water bodies. The one exception is Cr: here erosion is the most important pathway, while at the same time emissions from urban sewer systems, which also act as important source for particulate emissions, are negligible. Therefore the calculated enrichment ratio for Cr was used for all metals.

The lower the soil erosion, the more selective will be the enrichment process due to transport processes (AUERSWALD, 1989). The variation of the enrichment ratio can therefore be seen as dependent to the specific sediment input (SED). Figure 28 shows the relation between the sediment input and the enrichment ratios for the 16 catchment areas.

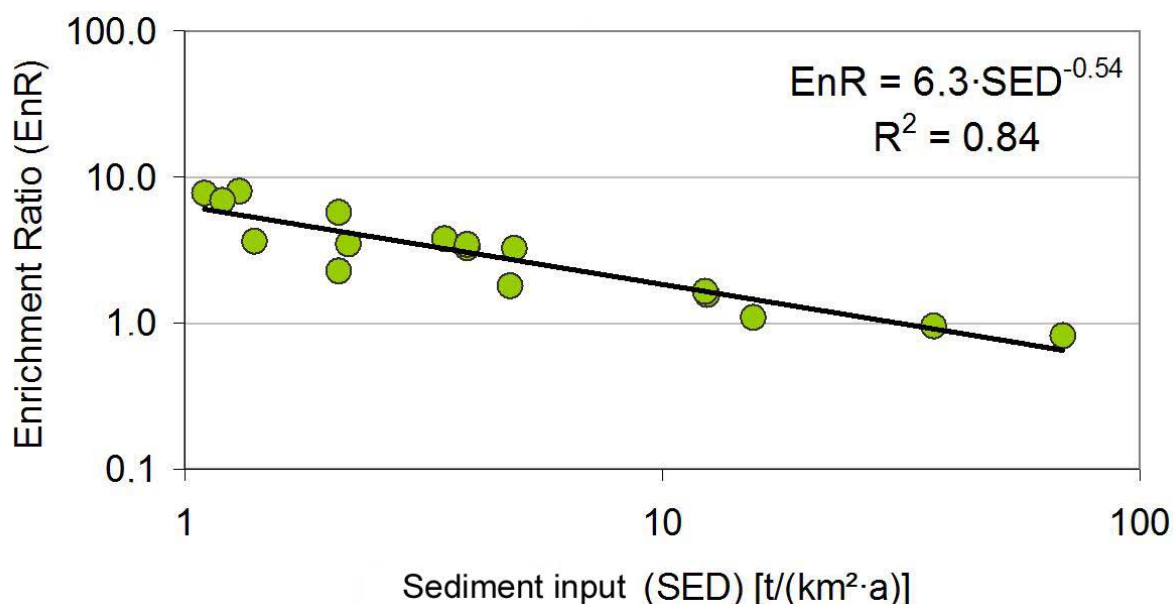


Figure 28: Correlation of the enrichment ratio EnR for chrome (Cr) to the specific sediment input (SED) for 16 medium-sized river basins in Germany (FUCHS ET AL., 2002)

The enrichment ratio for all analytical units can be calculated with the help of Equation 4-2 that is the result of the correlations of Figure 28. It is obvious that the enrichment ratio, if extrapolated over the area secured by measurement values, can give unrealistic values. The valid range of the equation is therefore set to a specific sediment input of 1-30 t/(km²·a). A fixed enrichment ratio is set for lower or higher sediment input (Equation 4-2). This procedure leads to an average enrichment ratio of 1.3 for heavy metals, which was within the dimension of the measurements (FUCHS & SCHWARZ, 2007).

Equation 4-2       $EnR = 6.3 \cdot SED^{-0.54}$       wenn  $1 < SED < 30 \text{ t/(km}^2\text{-a)}$

$EnR = 6.3$       wenn  $SED < 1 \text{ t/(km}^2\text{-a)}$

$EnR = 1$       wenn  $SED > 30 \text{ t/(km}^2\text{-a)}$

EnR    enrichment ratio for heavy metals [-]

SED    specific sediment input [ $\text{t/(km}^2\text{-a)}$ ]

Apart from the erosion from arable land, which represents the main sediment input in Germany, there is also erosion from natural open mountain areas. For estimating the heavy metal emissions from these areas an average geogenous heavy metal concentration was calculated for the whole of Germany (FUCHS ET AL., 2002). With the help of the Digital Geological Map (GK 1000, BGR, 2001) the share of the most common rock formations was calculated and with the help of heavy metal concentrations (HINDEL & FLEIGE, 1991) a weighted average was calculated (Table 17). Accumulation due to transportation was not considered for the quantification of emissions via natural erosion from open mountain areas, as the specific sediment input in such areas is high and therefore the accumulation is most probably negligible. (comp. Equation 4-2).

Table 17:    Average geogenous heavy metal concentrations in Germany

in mg/kg	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1983-2005	0.16	26.4	9.41	0.018	20.3	21.8	40.6

#### 4.2.2.3 Surface runoff

Due to missing data on heavy metal concentrations in surface runoff from unsealed areas rainfall concentrations were used for balancing emissions via surface runoff. For this the measurement readings („wet only“) of the German Federal Environment Agency were used from several monitoring points in Germany (BIEBER, 2007). Data has been available for all metals since 1995, for Cd, Hg and Pb even since 1994 and 1993 respectively. An average was calculated for every individual year from the data given by the monitoring stations. Data for the balance period 1983-1987 was extended from bibliographical references (FUCHS ET AL., 2002). Table 18 shows the database. The missing data for the years between the balance period 1983-1987 and the UBA-monitoring series was interpolated for the model calculations.

In addition to the loads from the rainfall the loads resulting from wash-off of fertilisers containing heavy metals were considered for agricultural areas. For this purpose the amount of mineral fertilisers (N-, P-, K-, Ca-, multi-element fertilisers) and organic manure (pig and cattle manure, dung and fowl excrement) as well as sewage sludge were collected on a federal state level (STATISTISCHES BUNDESAMT, 1986a, 1986b, 1987, 1991, 1995, 1996, 1998b, 1999, 2000; 2001, 2004, 2006a, 2006b; STATISTISCHES JAHRBUCH DER DDR, 1986). Subsequently the heavy metal loads applied to the agricultural areas could be calculated on a federal state level with the help of the heavy metal concentrations in the fertilisers and in the sewage sludge. The procedure as well as the underlying heavy metal concentrations are

described in detail in FUCHS ET AL. (2002). Table 19 shows the resulting heavy metal input loads onto the agricultural areas in Germany for the balance years 1985, 1995, 2000 and 2005. The share of fertilisers washed out by surface runoff is estimated according to the specifications of the ICPR (IKSR, 1999b) as being 0.3 %.

Table 18: Average heavy metal concentrations in the rainfall in Germany [µg/l]

in µg/l	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1983-1987 <sup>1</sup>	0.25	0.40	2.70	0.06	0.86	13.00	26.00
1993	-	-	-	0.02	-	-	-
1994	0.14	-	-	0.01	-	1.47	-
1995	0.11	0.39	2.80	0.02	0.82	2.24	13.63
1996	0.11	0.39	3.81	0.02	0.51	2.13	12.13
1997	0.09	0.40	3.61	0.02	0.51	1.97	9.91
1998	0.09	0.16	2.80	0.01	0.77	1.51	14.20
1999	0.05	0.14	2.75	0.01	0.85	1.15	13.58
2000	0.06	0.18	3.91	0.01	0.72	1.02	18.85
2001	0.03	0.10	2.81	0.01	0.26	1.03	7.32
2002	0.04	0.15	1.84	0.01	0.36	1.25	13.70
2003	0.06	0.17	1.38	0.01	0.33	1.25	7.40
2004	0.04	0.11	1.28	0.01	0.29	1.16	6.17

<sup>1</sup> FUCHS ET AL. (2002)

Table 19: Heavy metal loads in mineral fertilisers, organic manure and sewage sludge onto agricultural areas in Germany [kg/a]

in kg/a	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Mineral fertilisers							
1985	32,601	2,054,372	207,905	604	211,483	206,910	976,605
1995	18,495	357,502	99,985	348	77,495	133,771	659,920
2000	19,956	358,040	118,584	418	97,388	157,116	752,784
2005	14,388	262,739	95,158	322	79,197	127,156	571,541
Organic manure							
1985	11,406	412,541	3,317,305	1,484	234,180	244,838	12,271,769
1995	8,421	305,853	2,301,275	1,104	171,927	182,110	8,763,427
2000	8,133	293,630	2,342,332	1,057	167,320	175,058	8,720,964
2005	7,342	262,798	2,263,611	944	152,654	157,010	8,193,899
Sewage sludge							
1985	3,657	84,998	318,649	2,440	42,395	146,594	1,313,611
1995	1,711	48,679	253,960	1,550	27,808	78,914	876,349
2000	1,057	34,723	206,825	755	17,361	47,555	610,663
2005	879	28,887	172,069	628	14,444	39,563	508,043

#### 4.2.2.4 Tile drainage

To quantify the emissions via tile drainage the concentrations in seepage water from agricultural soils were used. Within the framework of an UBA research project 340 seepage water samples were taken from 16 agricultural soils from different locations in Germany and analysed in lysimeter experiments (BIELERT ET AL., 1999). A comparison of the median values of the seepage water concentrations of the different soils didn't show a consistent trend. Neither the classification of the type of soil nor the soil texture showed a specific behaviour that exceeded the natural range. The median values for all 340 seepage water samples were used to balance the drainage emissions (Table 20).

Table 20: Average heavy metal concentrations in seepage water (BIELERT ET AL., 1999) [µg/l]

in µg/l	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1983-2005	0.14	4.60	4.00	<0.14 <sup>1</sup>	8.90	0.28	19.00

<sup>1</sup> For Hg half of the value of the limit of quantification was considered.

#### 4.2.2.5 Groundwater

The quantification of the emissions via groundwater is based on the measurement readings of the New Geochemical Atlas of Germany (BIRKE ET AL., status of 2007). These were predominantly taken from springs of first and second order rivers at low water level (representative for the base flow) and therefore describe the loads that actually reach the surface waters through groundwater inflow. 954 monitoring points were considered altogether. Till now the median of all measurement readings was delivered by the Federal Institute for Geosciences and Natural Resources (BGR) (Table 21). It can be assumed that the large river basins are adequately represented through the median, as the sampling points are evenly spread over a grid each sized 350-400 km<sup>2</sup> across the whole of Germany. The data can only be regionalised after the New Geochemical Atlas has been finished. This should be followed up in the future as the geogenously caused heavy metal loads can play a considerable role regionally (FUCHS ET AL., 2007).

Table 21: Average heavy metal concentrations in spring water (BIRKE ET AL., status of 2007) [µg/l]

in µg/l	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1983-2005	0.02	0.26	1.03	<0.01 <sup>1</sup>	3.39	0.11	3.00

<sup>1</sup> For Hg half of the value of the limit of quantification was considered.

#### 4.2.2.6 Sewer systems

The main sources for heavy metal pollution of stormwater runoff from impervious urban areas are atmospheric deposition, road traffic (abrasion of tyres and brake pads), corrosion of metallic surfaces (roof and facade materials, rain gutters and down pipes, zinc-plated products) as well as contamination of impervious areas (HILLENBRAND ET AL., 2005).

Experiences from urban hydrology have shown that the annual pollutant loads washed off from impervious areas are mainly influenced by the pollutant deposition onto surfaces rather



than by the amount of stormwater runoff (FUCHS ET AL., 2002). MONERIS therefore uses a area-specific surface load for the emission quantification from impervious areas (comp. paragraph 2.5.2.6). In general only data on the concentration in stormwater runoff can be found in the bibliographical references. Pollutant potentials for surfaces can be back-calculated from the concentration data by considering the average long-term (1983-2005) stormwater runoff amount in urban areas and the impervious areas connected to the sewer system in Germany as well as the runoff coefficient.

BROMBACH & FUCHS (2002) searched bibliographical references for concentration data in stormwater runoff. This data collection was supplemented and updated within this project. As a basis for the calculation of the area-specific surface loads the average concentrations were calculated from the individual values for every year. Figure 29 and Figure 30 show the mean concentrations for the individual years from 1980 to 2005 for the metals Cd, Pb, Cr, Ni, Cu and Zn.

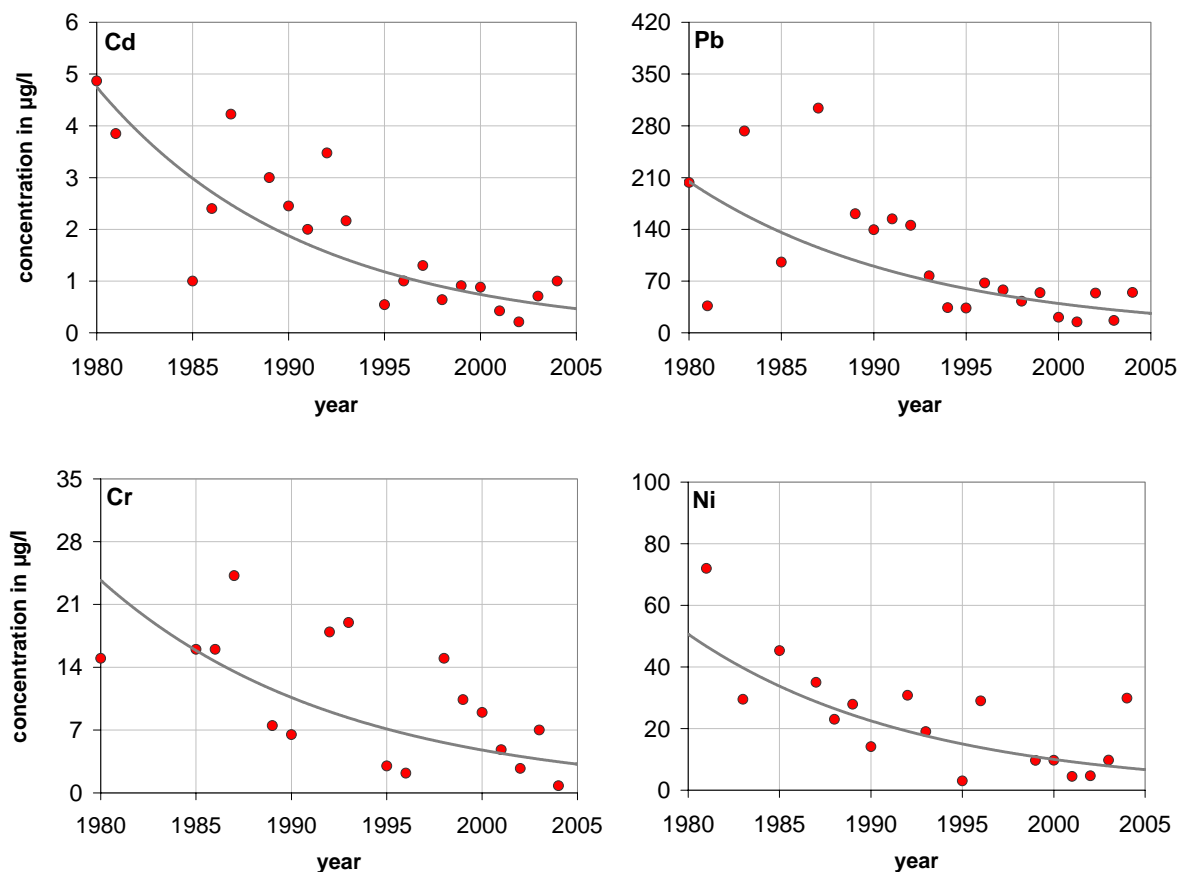


Figure 29: Average concentrations in stormwater runoff for cadmium (Cd), lead (Pb), chrome (Cr) and nickel (Ni) from 1980 to 2005

The heterogeneity of the sampling points (storm sewers, roof runoff, road runoff, etc.) and the complexity of the influencing variables such as the length of dry and rainfall periods lead to a larger range of concentrations, which is why the average values vary significantly, too. Even though, metals such as Cd, Pb, Cr and Ni have shown a clear downward trend since the 1980s (Figure 29).

For Cd combustion processes, airborne industrial emissions and traffic are considered the most important pollution sources for impervious areas. The reduction of emissions to the atmosphere is therefore the chief cause for reduced concentrations in stormwater runoff (FUCHS ET AL., 2002). A reduction of Pb concentrations in stormwater runoff is mainly due to the use of unleaded fuel. Furthermore, car parts containing Pb were to a large extent substituted in the past years (e.g. balancing weights made of Pb or Pb additives in solid lubricants in brake pads, HILLENBRAND ET AL., 2005). Reductions in the concentrations of Cr and Ni in stormwater runoff are also mainly due to the reductions in atmospheric deposition.

Cu and Zn on the other hand hardly show any reduction in the concentrations in stormwater runoff from 1980 to 2005 (Figure 30). The main sources for the release of Cu and Zn in urban areas are the corrosion of metal surfaces (roofs and rain gutters as well as zinc-plated products) and road traffic (abrasion of tyres and brake pads) (HILLENBRAND ET AL., 2005). Both emission sources did not show any reduction for the observation period. Congestion has increased noticeably since 1980. Furthermore, the reduction of corrosion rates of metal surfaces due to reduced  $\text{SO}_2$ - and  $\text{NO}_x$ -concentrations in rainfall since the 1980s was in large compensated by the increase of exposed Cu and Zn surfaces (HILLENBRAND ET AL., 2005). Even though, the minimal reduction can still be attributed to the reduction of atmospheric deposition, as for the other metals (paragraph 4.2.2.1).

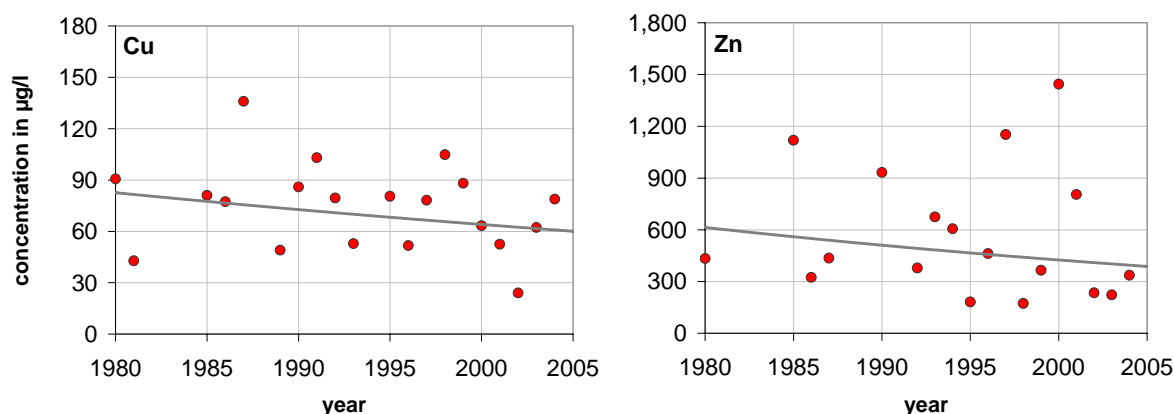


Figure 30: Average concentrations in stormwater runoff for copper (Cu) and zinc (Zn) from 1980 to 2005

Based on the trends in Figure 29 and Figure 30 a concentration was calculated for every year that was subsequently converted into an area-specific surface load according to the described procedure. Table 22 shows the area-specific surfaces loads for the four balance periods.

The calculated area-specific surface loads for the reference year 1985 can only be seen as representative for the old federal states of Germany due to the monitoring points. There are no measurement readings for the former GDR for stormwater runoff from impervious areas. Due to the different emission situations into the atmosphere the area-specific surface loads for the former GDR were increased by the difference of atmospheric deposition between the old federal states of Germany and the GDR in the reference year 1985 (Table 14, chapter 4.2.2.1). For the former GDR it must also be considered that in the reference year 1985 no copper was used as building material in roofs and rain gutters for residential as well public

buildings, which is why emissions from corrosion are only expected from historic buildings. A further important source for copper in urban areas is abrasion of brake pads. For this source it is also assumed that vehicles in the former GDR were equally not fitted with cupreous brake pads. HILLENBRAND ET AL. (2005) quantified the pollution sources for Cu in urban areas. Following the research 40 g/(ha·a) of the area-specific surface load can be lead back to the corrosion of Cu-areas and about 110 g/(ha·a) to brake pad abrasions. It was assumed that in 1985 in the former GDR about 20 g/(ha·a) were emitted from Cu-areas. The share of vehicles with cupreous brake pads was estimated to be 10 %. According to that the area-specific surface load for Cu in the former GDR was reduced by 139 g/(ha·a). Another assumption made for the area of the former GDR was that the area-specific surface load till 1989 stayed within the level of the reference year 1985 (Table 22).

Table 22: Area-specific surface load for heavy metals on impervious areas

in g/(ha·a)	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1983-1987 West	9.3	49.4	252.7	3.2	105.1	420.1	1,933.4
1983-1989 Ost <sup>1</sup>	46.8	51.0	176.7	4.2	133.6	439.3	2,278.4
1993-1997	3.7	22.4	227.0	1.3	46.7	184.8	1,774.2
1998-2002	2.3	15.1	214.1	0.8	31.1	122.6	1,694.6
2003-2005	1.6	11.0	203.8	0.6	22.4	87.9	1,630.9

<sup>1</sup> Area-specific surface load for the former GDR was adjusted.

There are no valid measurement readings for Hg on concentrations in stormwater runoff. The chief source for the pollution of impervious areas with Hg is atmospheric deposition (ECKLEY & BRANFIREUN, 2008). Maps from EMEP (2007a) show the atmospheric Hg-deposition for urban areas. In areas with an average degree of urbanisation the Hg-deposition is 0.5 g/(ha·a). This value was taken as area-specific surface load for the year 2005 and, according to the temporal changes for Cd, the deposition was back-calculated (Table 22).

The emission from storm sewers into surface waters is derived from the multiplication of the area-specific surface load with the impervious area connected to the separate sewer system.

For the urban pathways „combined sewer overflows“, „sewer system not connected to a wastewater treatment plant“ and „unconnected households“ additional emissions from households and industrial units must be considered. FUCHS ET AL. (2002) and WANDER (2004) conducted research in bibliographical references on inhabitant-specific heavy metal emissions. Heavy metal loads were retrieved for the years 1985 and 1998 from the compiled data. For the period before 1985 and after 1998 the values for the reference years were kept and were interpolated for the period in between. In the former GDR Cu-pipes were rarely used for the supply of drinking water which is why the emissions from drinking water pipes made of Cu have to be subtracted from the inhabitant-specific heavy metal loads. This was calculated according to HILLENBRAND ET AL. (2005) with the help of the concentrations in the drinking water (KRAUSE ET AL., 2001) and the amount of used drinking water. Samplings of domestic drinking water across Germany show that the Cu-concentration in the eastern federal states has risen since the 1990s but was still below the level in western Germany in 1998. The Cu-concentrations in the western federal states had risen, too (KRAUSE ET AL., 2001, Table 23).

Table 23: Inhabitant-specific heavy metal loads [mg/(inh-a)]

in mg/(inh-a)	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1983-1987	76.65	1,438	5,439 <sup>1</sup> /1,927 <sup>2,3</sup>	58.4	1,357.8	2,223	27,054
1993-1997	47.45	492	5,836 <sup>1</sup> /2,836 <sup>2</sup>	36.5	704.5	1,044	18,516
1998-2005	36.50	193	5,960 <sup>1</sup> /3,292 <sup>2</sup>	29.2	496.4	668	15,794

<sup>1</sup> old federal states of Germany, <sup>2</sup> new federal states of Germany, <sup>3</sup> 1983-1989

For the urban pathways „sewer system not connected to a wastewater treatment plant“ and „unconnected households“ only the dissolved share of the inhabitant-specific heavy metal loads is considered according to FUCHS ET AL. (2002) (Table 24).

Table 24: Dissolved share of the inhabitant-specific heavy metal loads [%]

in %	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1983-2005	75.0	93.3	73.8	83.3	78.6	95.7	67.0

The data on substance concentrations in wastewater from small industries according to NOLTE (1986) and SCHÄFER (1999) was used to balance the heavy metal emissions from wastewater of small industries (Table 25). Between the years 1986 and 1999 the concentrations were interpolated according to the procedure for inhabitant-specific emissions.

Table 25: Heavy metal concentrations in wastewater from small industries [µg/l]

in µg/l	Cd	Cr	Cu	Hg	Ni	Pb	Zn
NOLTE (1986)	43.0	382.0	688.0	5.8	268.0	514.0	2,046.0
SCHÄFER (1999)	4.6	44.0	149.0	1.5	50.0	152.0	523.0

### 4.3 Polycyclic aromatic hydrocarbons (PAH)

Polycyclic aromatic hydrocarbons are compounds made up out of several fused aromatic rings, some of which have been identified as carcinogenic for humans. More than 100 compounds are known, for example naphthalene with 2 aromatic rings, anthracene and fluoranthene with 3 rings and one often used as indicator substance, especially toxic benzo(a)pyrene with 5 rings. It is hard to detect all of the PAH due to their high number of bonds which is why the most important are compiled as „PAH sum“ ( $\Sigma$  PAH).

The individual substances anthracene, fluoranthene and naphthalene as well as the group of PAH are included in list of priority substances of the Water Framework Directive. Of all polynucleic aromatic compounds belonging to the PAH only anthracene, naphthalene and in small amounts fluoranthene are produced in Germany. The compounds are used to make dye and as intermedium, products. The use of creosotes containing a high percentage of PAH in wood preservation has been strongly restricted and is only allowed for commercial and industrial use. The most important regulations on emission limits are the wastewater

ordinance in the water sector (annex 46, coal coking) as well as the directive 2004/107/EG in air pollution control in which the Cd, Hg and Ni as well as the PAH are regulated. The restriction directives for creosotes (2001/90/EG) and for PAH in plasticiser oils and tyres (2005/69/EG) were transferred to the German law system via the ‘chemical prohibition order’ (Chemikalienverbotsverordnung) (HILLENBRAND ET AL., 2008).

In the following the data basis for the quantification of the most important point and diffuse pathways for PAH into waterbodies is described. To be able to directly compare them the EPA-PAH<sub>16</sub>-sum parameter ( $\Sigma$  EPA-PAH<sub>16</sub>) will be used. If there is only data for a few PAH then an extrapolation is carried out. The values used for that extrapolation are listed according to the kind of data available for the individual pathways. Of course, extrapolations are no alternative for the analysis of individual matters.

### 4.3.1 Point pathways

#### 4.3.1.1 Municipal wastewater treatment plants

The quantification of emissions from municipal wastewater treatment plants is carried out through multiplication of specific concentrations factors in their wastewater with the amount of treated wastewater (paragraph 3.4). After interpreting the bibliographical references (IVASHECHKIN, 2005 and GETTA & KORBER, 2005) there are measured concentrations between < 0.1 and 0.8 µg/L (Table 26, recalculated for  $\Sigma$  EPA-PAH<sub>16</sub>), an average concentration however cannot be estimated due to the small amount of measurements. The main reason for this is that the PAH concentrations in municipal wastewater are hard to detect, even the inflow concentrations are below the limit of quantification.

Table 26: Average concentrations in wastewater treatment plant effluents  $\Sigma$  EPA-PAH<sub>16</sub>

Federal state, number of WWTPs, number of PAH, limit of quantification (LOQ))	Concentration [µg/l]	Reference
Hesse, 10 WWTPs (PAH < LOQ (0.01 µg/l), extrapolated)	0.080	IVASHECHKIN (2005)
North Rhine-Westphalia, 2 WWTPs (PAH < LOQ (0.02 µg/l), extrapolated)	0.160	IVASHECHKIN (2005)
North Rhine-Westphalia (KA-Emscher) (PAH <sub>6</sub> , extrapolated)	0.820	GETTA & KORBER (2005)
Saxony, 14 WWTPs (PAH <sub>8</sub> , extrapolated)	0.105	IVASHECHKIN (2005)
Saxony, 9 WWTPs (PAH <sub>8</sub> , extrapolated)	0.109	UBA (data request)
Comparison: average value Germany (back-calculated from sewage sludge data)	0.137	

For a first assessment of the data on PAH concentrations in wastewater treatment plant loads with the description „below limit of quantification“ a value according to half of the limit of quantification is assumed. The given limits of quantification for an analysis apply to the different PAH compounds. For a comparison with measured concentrations it is used a  $\Sigma$  EPA-PAH<sub>16</sub> parameter (calculated with half of the limit of quantification multiplied with the factor

16) (comp. Table 26). Measurement values that comprise less than 16 PAH representatives are extrapolated so they can be compared easier. In the case of sewage sludges and PAH<sub>6</sub> (TVO) sum parameters it is possible to derive ratios on the basis of the published measurements. Using the survey by KOLLOTZEK ET AL. (1996), an average ratio for sewage sludges of  $2.3 \times \Sigma \text{PAH}_6 \text{ (TVO)} = \Sigma \text{EPA-PAH}_{16}$  can be calculated. In contrast there is no sufficient data basis available to extrapolate PAH<sub>8</sub> parameter. For a first estimation an average ratio of  $1.85 \times \Sigma \text{PAH}_8 = \Sigma \text{EPA-PAH}_{16}$  is assumed.

### Calculation of PAH effluent concentrations from sewage sludge data

A back-calculation from PAH effluent concentrations from sewage sludge data can be used as alternative to the data basis. The elimination for municipal wastewater treatment is estimated to be about 90 %, whereas the PAH compounds separated from the sewage end up in the sewage sludge (IVASHECHKIN, 2005).

For the calculation of PAH concentrations in wastewater treatment plant discharges an average concentration of  $\Sigma \text{EPA-PAH}_{16}$  of 5.5 mg/kg TS in sewage sludges in Germany is assumed (according to UBA 2007a) (comp. Table 27). Based on an amount of 2,106,756 t TS of sewage sludge in Germany in 2004 (BMU, 2007) about 11.6 t  $\Sigma \text{EPA-PAH}_{16}$  are deposited every year. The share of sewage sludge used in farming in Germany according to the UBA is 29.8 % in 2006 (UBA 2007a) whereby 613,476 t of sewage sludge carry about 3 t  $\Sigma \text{EPA-PAH}_{16}$  back into the open environment. Assuming that the efficiency of the existing clarification process for PAH is at least 90 % (IVASHECHKIN, 2005) then 12.9 t of  $\Sigma \text{EPA-PAH}_{16}$  annually are in the inflow of wastewater treatment plants in Germany. According to this assumption a maximum 10 % and with that about 1.3 t (loads from wastewater treatment plants)  $\Sigma \text{EPA-PAH}_{16}$  reach the waterways annually. Using the amount of treated wastewater in Germany in 2004 of 9.410 million cubic metres (DESTATIS, 2004) for further calculations shows an average effluent concentration of 0.137 µg/L  $\Sigma \text{EPA-PAH}_{16}$  (comp. Table 26).

Table 27:  $\Sigma \text{EPA-PAH}_{16}$  content in sewage sludge

Ø (mg/kg dry matter)	Reference
5.5	Average Germany (UBA, 2007a)
6.7	Average North Rhine-Westfalia (MUNLV, 2004)

### The determination of an inhabitant specific effluent load in neighbouring countries

There is no data of annual waste water amounts or of PAH concentrations in wastewater treatment plant effluent for Germany's European neighbouring countries. An average annual load per inhabitant is calculated for the population connected to a wastewater treatment plant based on the calculated PAH loads for Germany. Based on 77,374,058 connected inhabitants in Germany this amounts to an average load into waterbodies of 15.95 mg  $\Sigma \text{EPA-PAH}_{16}$  per inhabitant and year. In the European neighbouring countries the loads can be estimated on a catchment area level using the number of inhabitants who are connected to the sewer system and wastewater treatment plants.



#### 4.3.1.2 Direct industrial discharges

Data from the European Pollutant Emission Register EPER can be used for the pathway „direct industrial discharges“. The EPER has threshold limits for atmospheric emissions of 50 kg annually as well as of 5 kg per year for emissions into water according to Borneff PAH<sub>6</sub> (EPER, 2007).

For taking into account 16 PAH the same conversion factor as in sewage sludges is assumed:  
 $\Sigma \text{EPA-PAH}_{16} = \text{Borneff PAH}_6 \times 2.3$ .

#### Direct industrial dischargers

The loads reported from direct industrial dischargers into surface water bodies were 192 kg (Borneff-PAH<sub>6</sub>) for the year 2004. 125 kg are allotted to the sector inorganic basic chemicals or fertilisers (QK: 4.2/4.3), 51 kg to combustion plants > 50 MW (QK: 1.1), 10 kg on organic chemical primary matter (QK: 4.1) as well as 5.7 kg on coking plants (QK: 1.3) (comp. Table 28).

Table 28: Borneff-PAH<sub>6</sub> loads from direct industrial dischargers into waterbodies

EPER reporting year: 2004	Location	Economic main activity	[kg/a]
Zentralkokerei Saar	66763 Dillingen/Saar	Manufacture of coke oven products	5.65
Degussa AG - Werk Witten	58453 Witten	Manufacture of plastics in primary forms	10.10
InfraServ GmbH, IPH	65926 FFM-Höchst	Manufacture of chemicals and chemical products	51.00
BASF AG	67056 Ludwigshafen	Manufacture of other inorganic basic chemicals	125.00
(Source: EPER 2007)		<b><math>\Sigma \text{PAH}_6</math> (corrected)<sup>1</sup></b>	<b>192.00</b>
		<b><math>\Sigma \text{PAH}_{16}</math> (extrapolated)</b>	<b>442.00</b>

<sup>1</sup> Advice: According to the information given by the firm Papierf. Schöller & Hösch, Gernsbach (04/2008) the value reported to the EPER, 157 kg/a (about 1000 times higher) is due to a mistake during the transfer of the data and will therefore not be considered in the sum.

#### Indirect industrial dischargers

Indirect water emissions reported in the EPER are 1,480 kg (Borneff PAH<sub>6</sub>) for the report year 2004 (Table 29), the main part being allotted to the mineral oil and gas refineries. However, these loads are reduced significantly by the municipal wastewater treatment plants and are contained in the emission pathway „municipal wastewater treatment plants“ (comp. chapter 4.3.3.1).

Table 29: Borneff-PAH<sub>6</sub> loads from indirect industrial dischargers into waterbodies

<b>EPER reporting year: 2004</b>	<b>Location</b>	<b>Economic main activity</b>	<b>[kg/a]</b>
Sasol Germany GmbH	58453 Witten	Manufacture of other organic basic chemicals	5.8
Latoschik & Fischer	32120 Hiddenh.	Waste disposal	7.6
DSM Kunstharze GmbH	49716 Meppen	Manufacture of plastics in primary forms	10.8
KBS Kokereibetriebsges.	47166 Duisb,Schw.	Manufacture of coke oven products	15.0
DSK AG Kokerei Prosper	46236 Bott.-Wel.	Hard coal mining industry	123.0
RUHR OEL GmbH Horst	45899 Gelsenk.	Manufacture of refined petroleum products	539.0
RUHR OEL GMBH Scholven	45899 Gelsenk.	Manufacture of other organic basic chemicals	779.0
(Source: EPER, 2007)		<b>Σ PAH<sub>6</sub></b>	<b>1480.0</b>
		<b>Σ PAH<sub>16</sub> (extrapolated)</b>	<b>3404.0</b>

### 4.3.2 Diffuse pathways

Polycyclic aromatic hydrocarbons are generated during incomplete combustions from practically all organic matter, e.g. forest fires, domestic fires, combustion engines, grilling and smoking and are therefore predominantly spread over the atmospheric pathway. They can be transported over long distances as gas or adsorbed on airborne particles. The following will therefore outline the data situation on PAH emissions in the atmosphere.

#### 4.3.2.1 Atmospheric PAH emissions

According to an estimation of the UBA for 1994 the largest atmospheric PAH emissions are from the sectors house heating with 932 t/a, impregnation of wood with 529 t/a, production plants for non-ferrous metals and aluminium (258 t/a), emissions from wood treated with creosote (213 t/a), plants producing iron and steel (140 t/a) as well as traffic (146 t/a). Industrial heating plants and power stations (3.2 t/a) as well as waste disposal plants (0.03 t/a) seemed to be of less importance.

Currently, according to the „Pollutant Release and Transfer Register“ (PRTR, 2007) the situation is changing. There is new data available for Germany due to international report commitments (e.g. Convention on Long-range Transboundary Air Pollution, UN ECE-CLRTAP) as well as research projects (modelling and monitoring) (PRTR, 2007). It has to be mentioned, that the data on PAH emissions are up-to-date but neither complete nor conclusive. According to KUNZE (2008) the PRTR database (as of May 2008) is missing the source groups „thermal waste treatment“, „open combustion“, „animal carcass plants“, „anode production“ and „wood preservation units“ either in parts or completely. No data sources could be found for the activity rates on emissions, that would allow a report from 1990 to today (UBA, 2008a). Different specifications on PAH emissions in older UN ECE-CLRTAP reports compared with the currently available reports for UN ECE-CLRTAP 2007 and 2008 can be

lead back to qualitatively improved activity rates of the database which were able to be considered for the most recent reports

## Diffuse PAH sources according to the EU PRTR regulation

$\Sigma$  PAH (PAH<sub>4</sub>)

According to the  $\Sigma$  PAH from the four PAH compounds benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthenes and indeno[1,2,3-cd]pyrene in PRTR (2007) the diffuse emissions for Germany in the year 2005 amount to 97.96 t, where about 88 % are allotted to domestic fires, 5.8 % to industrial processes and about 1.8 % to traffic (exhaust and abrasions) (Figure 31).

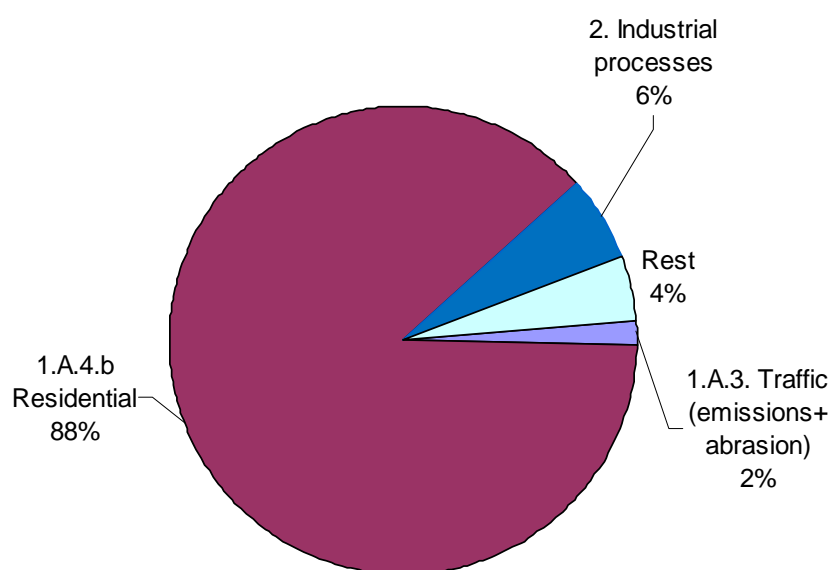


Figure 31: Share of emission sources of  $\Sigma$  PAH (PAH<sub>4</sub>) in total emissions, reference year 2005 (PRTR, 2007)

Table 30: Diffuse emissions  $\Sigma$  PAH (PAH<sub>4</sub>) [t/a] (PRTR, 2007)

Visualisation of the time courses: [www.ki-werkstatt.de](http://www.ki-werkstatt.de)

	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Total	177.70	136.85	98.97	92.69	85.46	87.94	87.59	95.95	89.56	89.28	88.33	97.01	92.77	97.08	96.40	97.96
1. Energy	171.87	131.08	93.35	87.27	79.85	82.03	81.86	90.01	83.46	83.17	82.18	91.21	87.12	91.25	90.37	92.18
1. A. Combustion of Fossil Fuels	166.43	126.77	89.45	84.08	76.97	79.10	79.04	87.17	80.74	80.89	79.76	89.27	85.19	89.17	88.13	89.95
1.A.3. Traffic	1.19	1.30	1.33	1.34	1.38	1.42	1.45	1.49	1.52	1.56	1.57	1.59	1.61	1.61	1.63	1.64
1.A.3. b Road Transportation	1.19	1.30	1.33	1.34	1.38	1.42	1.45	1.49	1.52	1.56	1.57	1.59	1.61	1.61	1.63	1.64
1.A.4. Other Combustion Plants	165.20	125.50	88.10	82.70	75.60	77.70	77.60	85.70	79.20	79.30	78.20	87.70	83.60	87.60	86.50	88.30
1.A.4.a Commercial / Institutional	17.16	17.49	9.50	8.65	3.88	7.43	6.21	6.47	3.29	3.04	3.39	3.37	3.12	2.39	2.16	2.13
1.A.4.b Residential	148.07	107.98	78.62	74.09	71.71	70.24	71.38	79.22	75.93	76.29	74.80	84.31	80.46	85.17	84.34	86.19
1.B. Diffuse Emissions of Fuels	5.77	4.31	3.90	3.19	2.88	2.94	2.82	2.84	2.72	2.27	2.42	1.93	1.93	2.08	2.25	2.23
2. Industrial Processes	5.77	5.71	5.55	5.36	5.54	5.84	5.65	5.86	6.03	6.03	6.07	5.72	5.56	5.75	5.94	5.70
2.A. Mineral Products	2.86	2.57	2.70	2.71	2.87	2.91	2.77	2.85	2.90	2.95	2.85	2.52	2.40	2.52	2.63	2.44
2.C. Metal Production	2.91	3.14	2.85	2.64	2.67	2.93	2.89	3.01	3.13	3.09	3.22	3.20	3.17	3.23	3.32	3.26
7. Others	0.06	0.07	0.07	0.07	0.07	0.07	0.07	0.07	0.08	0.08	0.08	0.08	0.08	0.08	0.08	0.08

The colours serve as visualisation in the table. Completely brown rows mean maximum values; completely yellow rows mean minimum values. Values in between are shown via the green level in the RGB composition. ([http://www.diffuse-sources.prtr.de/visualisierung\\_von\\_zeitreihen.pdf](http://www.diffuse-sources.prtr.de/visualisierung_von_zeitreihen.pdf))

## Benzo(a)pyrene

For the sources compiled within the framework of the PRTR (2007), the diffuse emissions of the PAH indicator substance benzo(a)pyrene (B(a)P) are noted with 36.12 t for the year 2005. 91.1 % are allotted to domestic heating, 3.2 % to industrial processes and about 0.78 % to traffic (sum of combustion processes and substance abrasions) (comp. Figure 32).

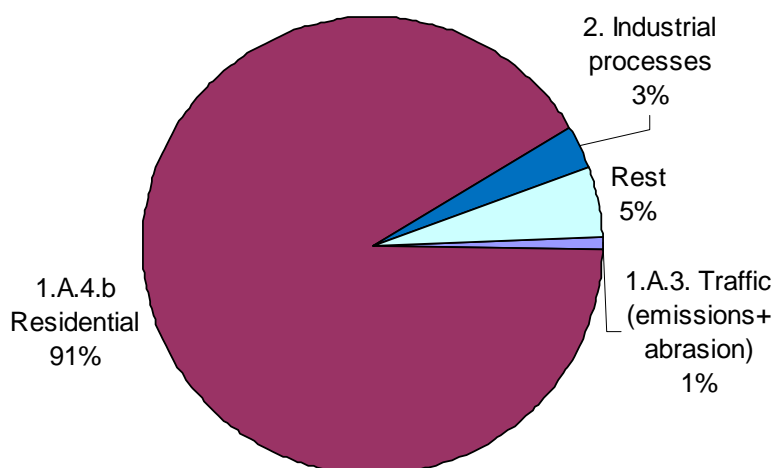


Figure 32: Share of emission sources of benzo(a)pyrene (B(a)P) in total emissions, reference year 2005 (PRTR, 2007)

The relative importance of the emission sources for B(a)P largely corresponds with the importance of the PAH sum parameter of the four individual compounds (comp. Figure 31). Like for  $\Sigma$  PAH (PAH<sub>4</sub>), an increase for B(a)P has been observed especially in the essential field of domestic fire places, after a decline till the end of the 1990s (comp. Table 30 and Table 31). This result backs the assumption, that B(a)P can be used as indicator substance for atmospherically transporting PAH (especially from the source domestic heating).

Table 31: Diffuse benzo(a)pyrene B(a)P emissions [t/a] (PRTR, 2007)

Visualisation of the time courses: [www.ki-werkstatt.de](http://www.ki-werkstatt.de)

	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Total	64,26	49,83	36,02	33,9	31,13	32,49	32,27	35,38	32,94	33,06	32,6	35,96	34,47	35,96	35,62	36,12
1. Energy	62,14	47,81	34,23	32,25	29,59	30,75	30,6	33,76	31,3	31,46	31,04	34,47	33,05	34,61	34,33	34,95
1. A. Combustion of Fossil Fuels	61,87	47,81	34,23	32,25	29,59	30,75	30,6	33,76	31,3	31,46	31,04	34,47	33,05	34,61	34,33	34,95
1.A.1. Power Industry	0,01	0,01	0,01	0,01	0,01	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02
1.A.3. Traffic	0,19	0,2	0,21	0,21	0,22	0,22	0,22	0,23	0,23	0,24	0,24	0,25	0,25	0,25	0,25	0,25
1.A.4. Other Combustion Plants	61,7	47,6	34	32	29,4	30,5	30,4	33,5	31,1	31,2	30,8	34,2	32,8	34,3	34,1	34,7
1.A.4.a Commercial / Institutional	7,1	7,1	4,4	4,1	2,2	3,6	3,1	3,3	2,2	2,1	2,2	2,1	2,1	1,8	1,8	1,8
1.A.4.b Residential	54,6	40,5	29,6	28	27,1	26,9	27,2	30,2	28,8	29,1	28,6	32,1	30,7	32,5	32,3	32,9
1.B. Diffuse Emissions of Fuels	0,27	0,22	0,2	0,16	0,14	0,15	0,14	0,14	0,14	0,11	0,12	0,1	0,1	0,1	0,11	0,11
2. Industrial Processes	2,09	2	1,76	1,62	1,52	1,72	1,64	1,59	1,61	1,57	1,54	1,46	1,39	1,32	1,26	1,14
2.A. Mineral Products	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02	0,02
2.C. Metal Production	2,07	1,98	1,74	1,6	1,5	1,69	1,62	1,57	1,59	1,55	1,51	1,44	1,37	1,3	1,24	1,12
7. iii Abrasion (Tires, Brakes, Roads)	0,02	0,02	0,02	0,02	0,02	0,03	0,03	0,03	0,03	0,03	0,03	0,03	0,03	0,03	0,03	0,03

## Current emission developments according to the UN ECE-CLRTAP (2008)

In addition to the data for 2005 published in the PRTR (2007), the inventory tables of the UBA (as most up-to-date data of 06/2008) on PAH<sub>4</sub> and B(a)P has been utilized. These ta-

bles has been evaluated by the UBA for the „Reports 2008“ (report year 2006) under the Convention on Long-range Transboundary Air Pollution (UN ECE-CLRTAP).

The source groups „thermal waste treatment“, „open combustion“, „animal carcass plants“, „anode production“ and „wood preservation units“ are still missing in the current version of the inventory tables (Table 32). Until now no sources for data on activity rates could be found for these emission sources which would allow a coverage of the time from 1990 till today (KUNZE, 2008). In contrast up-to-date and qualitatively improved activity rates are considered for the existing emission sources.

With regard to the absolute scale as well as the relative relevance of the individual sources for diffuse B(a)P and  $\Sigma$  PAH<sub>4</sub>-emissions a continuing trend with slightly increasing absolute values can be detected in comparison to the data from PRTR (2007) (comp. Table 30 and Table 31) (UBA, 2008b).

Table 32: Benzo(a)pyrene B(a)P und  $\Sigma$  PAH<sub>4</sub> [t/a] from the UN ECE report (as of 06/2008), report year 2006

	Sectors of the UN ECE Report	B(a)P	PAH <sub>4</sub>
1 A 1 a	1 A 1 a Public powerplants and district heating plants	0.07	0.32
1 A 1 b	1 A 1 b Petroleum refineries	0.01	0.00
1 A 1 c	1 A 1 c Transformation facilities for solid fuels and other energy industries	0.00	0.14
1 A 2	1 A 2 Manufacturing industry and building industry	0.05	0.97
1 A 3 b	1 A 3 b Road traffic	0.28	1.74
1 A 4 a	1 A 4 a Commercial and institutional incineration plants	1.77	2.19
1 A 4 b	1 A 4 b Domestic heating	33.38	87.94
1B1	1B1 Diffuse Emissions of solid fuels	0.11	2.21
2 A	2 A Mineral products (b)	0.02	2.49
2 C	2 C Metal production	0.97	3.10
<b>TOTAL</b>	<b>Total amount for Germany</b>	<b>36.69</b>	<b>101.10</b>

(UBA, 2008b): inventory tables in the 2008 report (as of 06/2008)

#### *PAH share in the PM<sub>10</sub>-particulate matter fraction*

According to the PRTR (2007) in the year 2005 approx. 13 % of the particulate matter emissions of the PM<sub>10</sub>-fraction are allotted to the sector domestic heating, 22.5 % to industrial processes and about 21.4 % to traffic (combustion processes and substance abrasions) (comp. Figure 33 and Table 33). A rough estimate of the PAH contents in particulate matter can be made under the presumption that the PAH is completely bound to particles.

On the basis of the PRTR data (2007) the calculated percentage of  $\Sigma$  PAH in the PM<sub>10</sub> fraction is 0.051 %. This leads to calculated  $\Sigma$  PAH percentages in particulate matter of 0.35 % for the source households, 0.013 % for industrial processes and 0.0043 % for traffic (combustion processes and substance abrasions). The percentage of B(a)P in total PM<sub>10</sub> is 0.019 %. Based on the available values calculated for B(a)P percentages in particulate matter 0.13 % are allotted to the source households, 0.0026 % to industrial processes and 0.00069 % to traffic (combustion processes and substance abrasions) and therefore likewise the  $\Sigma$  PAH relevance: households > industry > traffic.

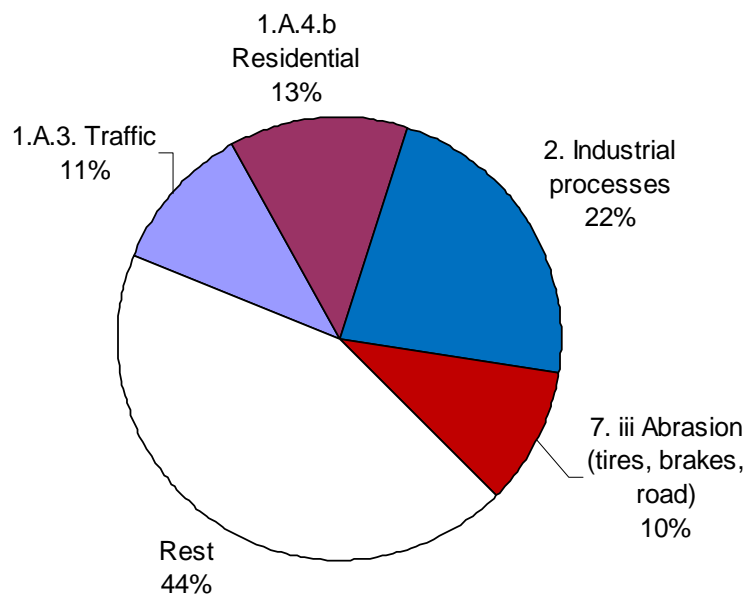


Figure 33: Share of emission sources of PM<sub>10</sub>-fraction in total emissions, reference year 2005 (PRTR, 2007)

Table 33: PM<sub>10</sub> emissions [1,000 t/a] (PRTR, 2007)

Visualisation of the time courses: [www.ki-werkstatt.de](http://www.ki-werkstatt.de)

	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Total	1613.2	252.7	220	195.7	187.5	234	219.6	227.4	211.3	222.5	210.4	206.5	200.7	196.7	196.4	193.5
1. Energy	1476.5	113.8	87.2	68.7	59.1	101.3	92.7	91.1	80.8	76.9	71.3	71.7	68	66.9	65.6	63.9
1. A. Combustion of Fossil Fuels	1474.5	112.35	86.06	67.81	58.43	100.6	91.99	90.44	80.21	76.45	70.77	71.31	67.61	66.46	65.18	63.42
1.A.1. Power Industry	1178.45	29.72	24.69	14.43	12.29	21.67	19.36	16.31	14.77	12.8	11.78	11.99	11.48	11.85	11.56	11.28
1.A.3. b Road Transportation	35.91	36.07	38.3	39.21	39.96	38.7	36.72	34.97	32.1	31.49	29.54	27.58	25.57	22.94	22.71	20.79
1.A.4. Other Combustion Plants	NE	NE	NE	NE	NE	31.58	28.96	32.52	28.06	27.54	25.74	28.49	27.31	28.36	27.52	27.99
1.A.4.a Commercial / Institutional	NE	NE	NE	NE	NE	6.48	5.14	5.69	3.15	2.75	2.49	2.48	2.41	2.2	2.05	1.98
1.A.4.b Residential	NE	NE	NE	NE	NE	23.77	22.59	25.72	24.09	23.92	22.53	25.28	24.18	25.45	24.77	25.31
1.B. Diffuse Emissions of Fuels	2.25	1.49	1.13	0.93	0.71	0.71	0.68	0.64	0.58	0.48	0.5	0.41	0.4	0.43	0.46	0.46
2. Industrial Processes	74.5	79.41	73.6	67.69	68.91	73.45	67.2	64.96	59.98	53.68	47.56	45.18	44.29	44.34	44.94	43.58
2.A. Mineral Products	22.3	25.3	24.98	23.05	22.72	22.12	19.57	17.76	16.08	15.81	15.04	14.02	13.42	13.25	13.07	12.64
2.B. Chemical Industry	0.1	0.1	0.09	0.08	0.07	0.52	0.53	0.5	0.48	0.46	0.53	0.5	0.47	0.47	0.48	0.47
2.C. Metal Production	44.45	45.45	40.95	37.26	38.82	43.3	39.46	40.74	38.62	34.2	30.15	28.93	28.7	28.89	29.59	28.72
7. iii Abrasion (Tires, Brakes, Roads)	14.94	15.62	16.21	16.47	16.85	17.25	17.53	17.83	18.2	18.68	18.79	19.12	19.37	19.3	19.63	19.6

NE: not estimated

#### PAH share of the PM<sub>2.5</sub>- particulate matter fraction

The calculated share of PM<sub>2.5</sub> in the PM<sub>10</sub> fraction is 57.3 %; the share of Σ PAH in the PM<sub>2.5</sub> fraction is 0.088 %.

The available data also points towards differences in the PAH share of particulate PM<sub>2.5</sub> from different sources. This leads to calculated Σ PAH share in PM<sub>2.5</sub> of 0.37 % for the source households, 0.034 % for industrial processes and 0.0055 % for traffic (combustion processes and substance abrasions). The calculated share of B(a)P in PM<sub>2.5</sub> is 0.033 %. This leads to a calculated B(a)P share in particulate PM<sub>2.5</sub> of 0.14 % for the source households, 0.0069 % for industrial processes and 0.00089 % for traffic (combustion processes and substance abrasions). This results in following relevances as for Σ PAH: households > industry > traffic (Figure 34 and Table 34).



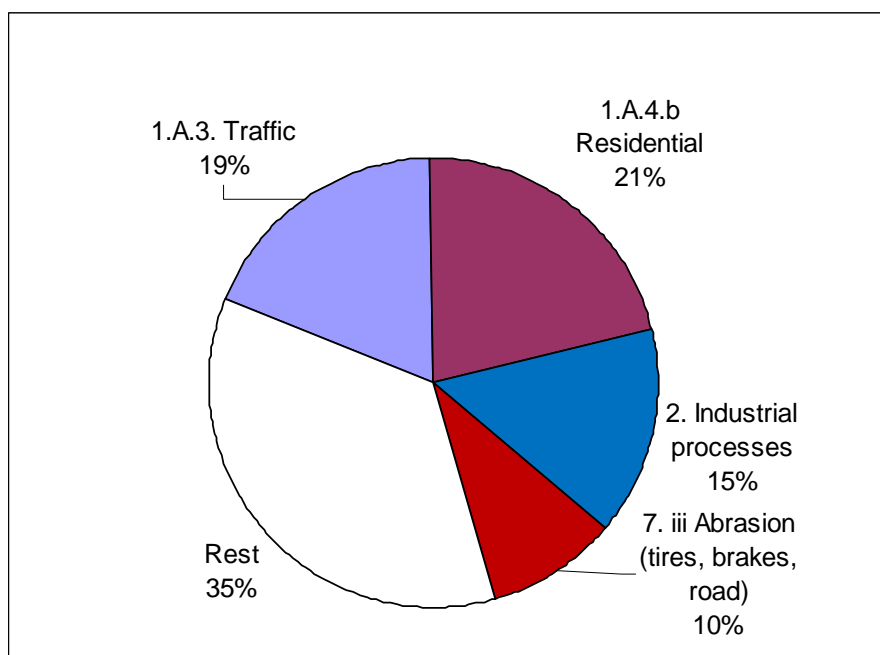


Figure 34: Share of emission sources of PM<sub>2.5</sub>-fraction in total emissions, reference year 2005 (PRTR, 2007)

Table 34: PM<sub>2.5</sub> emissions [1,000 t/a] (PRTR, 2007)

Visualisation of the time courses: [www.ki-werkstatt.de](http://www.ki-werkstatt.de)

	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Total	1378.4	169.1	143.1	123.8	115.3	157.5	146.2	144.9	132.2	130.1	121.3	120.2	115.9	114	113.1	110.9
1. Energy	1316.1	104.8	81.5	65.3	56.8	95.7	87.6	86.3	76.6	72.9	67.5	67.9	64.3	63	61.8	60.1
1. A. Combustion of Fossil Fuels	1315.03	104.18	81.08	64.96	56.58	95.42	87.34	86.06	76.43	72.74	67.36	67.74	64.17	62.9	61.66	59.91
1.A.1. Power Industry	1047.53	26.42	21.95	12.82	10.93	19.1	17.08	14.38	13.02	11.31	10.42	10.61	10.16	10.49	10.23	9.98
1.A.3. b Road Transportation	35.91	36.07	38.3	39.21	38.96	38.7	36.72	34.97	32.1	31.49	29.54	27.58	25.57	22.94	22.71	20.79
1.A.4. Other Combustion Plants	NE	NE	NE	NE	NE	29.65	27.14	30.64	26.43	25.69	23.97	26.54	25.44	26.41	25.61	26.03
1.A.4. a Commercial / Institutional	NE	NE	NE	NE	NE	6.05	4.8	5.54	3.13	2.53	2.28	2.28	2.21	2.01	1.87	1.8
1.A.4. b Residential	NE	NE	NE	NE	NE	22.39	21.24	24.1	22.57	22.38	21.05	23.61	22.59	23.76	23.12	23.6
1.B. Diffuse Emissions of Fuels	1.04	0.63	0.44	0.36	0.25	0.25	0.24	0.22	0.19	0.16	0.16	0.13	0.13	0.14	0.14	0.14
2. Industrial Processes	33.71	36.06	33.31	30.21	30.17	33.46	30.03	27.64	24.73	21.92	18.59	17.39	16.81	16.83	17.03	16.53
2.A. Mineral Products	9.55	11.34	11.1	9.9	9.62	11.46	9.41	7.9	6.53	6.41	5.89	5.15	4.73	4.62	4.51	4.22
2.C. Metal Production	19.48	20.19	17.74	15.88	16.21	17.3	15.86	15.94	15.06	13.27	11.18	10.81	10.69	10.8	11.07	10.89
7. iii Abrasion (Tires, Brakes, Roads)	8.08	8.55	8.76	8.9	9.1	9.32	9.47	9.64	9.83	10.09	10.15	10.33	10.46	10.42	10.6	10.59

NE: not estimated

On the basis of the available PRTR data the diffuse atmospheric emissions of the PAH indicator substance B(a)P in Germany for the years 2001 to 2005 was in average about 35.6 t/a. About 90 % are allotted to domestic heating (solid fuels: wood, coal) while only 5.4 % were attributed to combustion plants in industry, trade and service as well as 3.6 % for industrial processes producing metals. The influence of the traffic sector (roads) is estimated to be about 0.8 % (PRTR, 2007). Assuming the B(a)P percentage to be about 5 % (SCHEFFER & SCHACHTSCHABEL, 2002; GÖTZ, 2008) of  $\Sigma$  EPA-PAH<sub>16</sub> total emissions in Germany can be estimated to be within the range of about 712 t/a.

This insight reveals the special relevance of domestic heating (wood and coal fuel) for the particulate emissions and the associated PAH emissions via the atmospheric pathway as well as the total PAH emissions in Germany. Within domestic heating the percentage of

wood fuel has risen sharply in the past years (comp. Figure 35) and therefore compensates the decline of the use of coal (UBA, 2007c). The associated influence of the heating period leads to significant differences for the average atmospheric deposition about by the factor 2 between measurements in summer and in winter (UMWELTBEOBACHTUNG, 2007). Large differences can appear depending on the predominant heating technology.

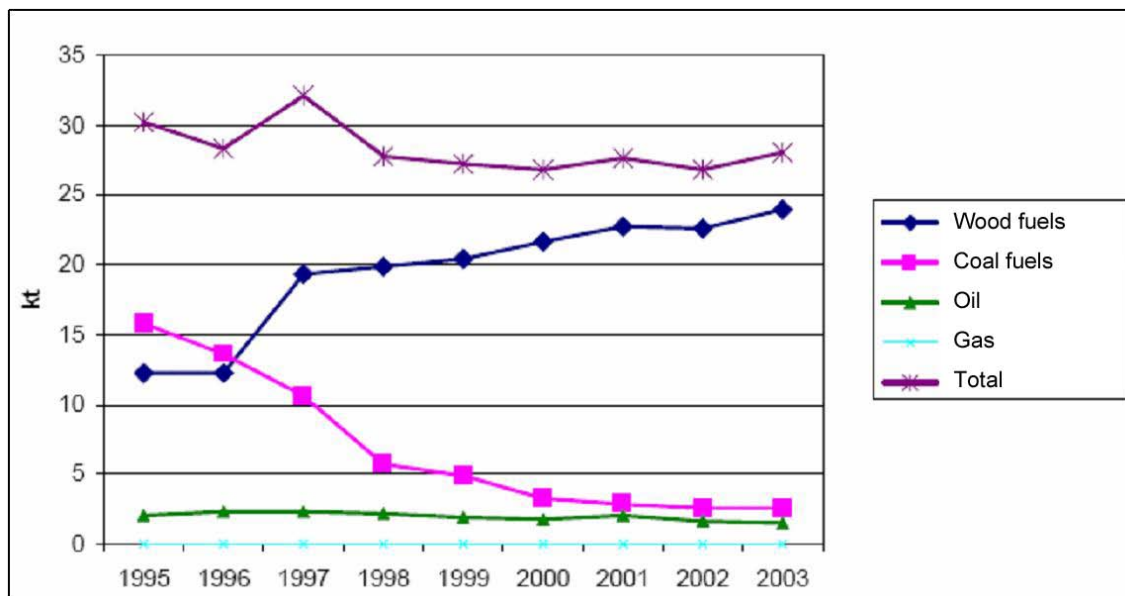


Figure 35: Development of the particulate matter emissions from units according to the First Ordinance for the Implementation of the Federal Immission Control Act (BMU, 2007)

#### 4.3.2.2 Atmospheric deposition onto water surface

Bibliographical references on resulting PAH deposition are characterised by strong variabilities (factors 10 to 1,000) (GÖTZ, 2008). Main reasons for this variability are the varying methods of determination (collecting technique, amount of researched PAH congeners), period under observation as well as geographically determined differences.

A survey of the German Federal Environment Agency as well as the federal state monitoring network about atmospheric deposition of PAH in Germany (from 07/2007 to 01/2008) has been done (GÖTZ, 2008). Not all 16 federal states were able to provide data (Table 35).

According to the survey, the dominant measurement systems collect data on PAH in connection with dust deposition as well as particle matter concentrations in the ambient air (percentages of the  $PM_{10}$ -fraction). For dust deposition only individual phases (dry or wet deposition flows) are considered meaning that a complete PAH coverage is not given. Measurements of PAH in the  $PM_{10}$  fraction are influenced by the choice of the monitoring point (often points with high pollution) as well as the necessary conversions into deposition rates. An average deposition speed of 0.2 cm/s can be assumed for  $PM_{10}$  particles (GÖTZ, 2008). But these are approximated values concerning the total airborne particulate matter, which is why considerable deviations are assumed for individual PAH components due to the particular physiochemical properties. Since 2002 there has been a standardized measurement according to

DIN 19739, which is optimised for PAH to measure the complete deposition by using funnel-adsorber-collectors. They are currently only used in a few measurement networks.

Table 35:  $\Sigma$  EPA-PAH<sub>16</sub> deposition rates in the federal states (federal states, monitoring network survey 2007, GÖTZ, 2008)

Federal state	Remarks	Deposition in g/(ha a) ( $\Sigma$ EPA-PAH <sub>16</sub> )
Baden-Wuerttemberg	(DIN, PM10)	4.2
Berlin	(PM10)	12.8
Brandenburg	(DD)	3.0
Hamburg	(DD)	5.1
Hesse	(DIN, PM10)	4.7
Mecklenburg-Western Pomerania	(PM10)	8.2
North Rhine-Westphalia	(PM10)	13.8
Saxony	(PM10)	11.7
Saxony-Anhalt	(DD, DIN, PM10)	4.2
Schleswig-Holstein (UBA, coast)	(DD)	0.3
<b>Average value Germany *</b>		<b>6.8</b>

References: DD = direct deposition measurement (particulate matter),

DIN = DIN 19739

PM<sub>10</sub> = measurement PM<sub>10</sub> fraction (ambient air)

\* Average for Germany: 6.8 g/(ha·a)

Bavaria, Bremen, Lower Saxony, Rhineland-Palatinate, Saarland, Thuringia

For Germany an average deposition for PAH<sub>16</sub> of about 6.8 g/(ha·a) is calculated with the help of the available averages for each federal state (Table 35). Additionally, the transnational matter transfer plays an important role. Regionally increased depositions in Saxony (Eastern European neighbouring countries with a heavy use of coal fuel) and in North Rhine-Westphalia (industrial emissions in the Benelux) can be linked to emission situations in the neighbouring countries due to typical atmospheric flow conditions (comp. Figure 36, Table 35 and Table 36).

The atmospheric deposition of PAH in the neighbouring European countries is derived with the help of up to date information on emissions according to EMEP (2007b). A conversion was carried out with the help of the modelled total deposition of the PAH indicator substance B(a)P (EMEP reference year 2005) as well as details on the respective country's area (FEDERAL FOREIGN OFFICE, 2008) to obtain results that could be compared to Germany.

By evaluating the measured data for atmospheric deposition in Germany (comp. GÖTZ, 2008) a percentage of approx. 5 % of the PAH indicator compound B(a)P in a  $\Sigma$  EPA-PAH<sub>16</sub> parameter can be presumed. Assuming that B(a)P percentages are 5 % leads to calculated average  $\Sigma$  EPA-PAH<sub>16</sub> deposition values for countries bordering Germany (comp. Table 36).

Table 36: Average  $\Sigma$  EPA-PAH<sub>16</sub> deposition rates for bordering countries

Country	Deposition in g/(ha·a) ( $\Sigma$ EPA-PAH <sub>16</sub> )
Poland	14.5
Czech Republic	10.6
Switzerland (Empa, 2006)	1.6
Austria	5.1
Liechtenstein (no data, same as CH)	1.6
Italy	6.8
France	2.0
Luxembourg	8.0
Belgium	11.0
Netherlands	7.9
Denmark	4.2

Sources: EMPA (2006), EMEP (2007b; data for the reference year 2005)

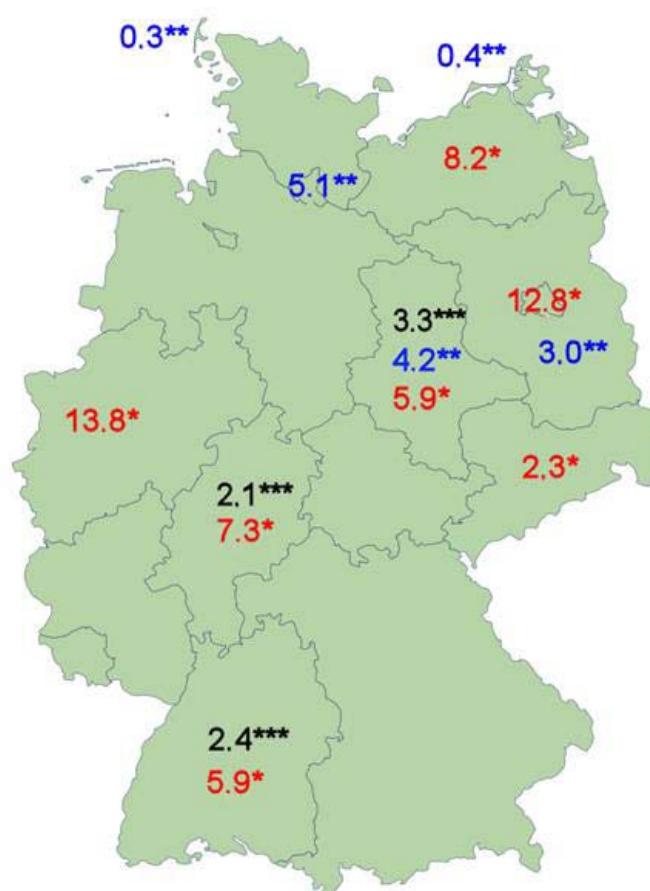


Figure 36: Average PAH deposition rates in the federal states in g/(ha a), Götz (2008)  
 \*measurement PM<sub>10</sub> ambient air, \*\* (dust) deposition measurement,  
 \*\*\*according to DIN 19739

#### 4.3.2.3 Erosion

Emissions into waterbodies through erosion are defined by the substance content in the topsoil, the sediment input into surface waters and an enrichment ratio due to transport (ER). Owing to the preferred transport of fine soil particles, fine grain sizes accumulate in the eroded sediment during the erosion process (FUCHS ET AL., 2002). As PAH are attached to finer particles due to their high specific surface (SCHEFFER & SCHACHTSCHABEL, 2002) they also accumulate through the erosion process. The enrichment of the substance in the eroded matter is described as the enrichment ratio EnR in comparison to the initial concentration of the matter in the field topsoil. For a first large-scale observation it is assumed for PAH that the EnR is in the same order as for heavy metals (comp. paragraph 4.2.2.2).

The available data of LABO (2003) on organic contaminants and their background values for soils show significant differences for the concentrations of PAH in topsoils for the individual federal states (comp. Table 37). The main reasons for the deviations apart from regional differences are especially the different reference periods, the amount of examined PAH compounds as well as the distinction according to land use. If different types of soils for arable areas are given a weighted average is calculated depending on the amount of samples taken.

If Borneff-PAH<sub>6</sub> is measured it is extrapolated by the factor 2.3 to get  $\Sigma$  EPA-PAH<sub>16</sub>. If there is no data available for a federal state, an average for Germany ( $\Sigma$  EPA-PAH<sub>16</sub>) is used which is composed of the values of other federal states. For Germany bibliographical references referring to SCHEFFER & SCHACHTSCHABEL (2002) assume an average  $\Sigma$  EPA-PAH<sub>16</sub> concentration of 0.2 mg/kg on arable areas, 0.4 mg/kg in forest soils as well as 1.1 mg/kg in urban areas. The natural background values are stated as being between 1 and 10 µg/kg.

Table 37:  $\Sigma$  EPA-PAH<sub>16</sub> background values in topsoil of arable areas in Germany (50P-values) (LABO, 2003)

Federal state	Topsoil content [mg/kg] ( $\Sigma$ EPA-PAH <sub>16</sub> )
Baden-Wurttemberg	0.19
Brandenburg	0.16
Hamburg	1.30
Hesse	0.10
Mecklenburg-Western Pomerania	0.19
Lower saxony	0.21
Rhineland-Palatinate	0.26
Saarland	0.74
Saxony	0.28
<b>Germany (average)</b>	<b>0.38</b>

#### Data on bordering countries

The PAH concentrations in soils of neighbouring countries are derived with the help of data from the bordering German federal states. For countries with no common border with Germany (e.g. Italy) the German average is adopted. For the Eastern European countries the

highest available value of the neighbouring federal state is assumed due to the more intense use of coal fuels (Table 38).

Table 38:  $\Sigma$  EPA-PAH<sub>16</sub>- background values in topsoil of arable areas for neighbouring countries

Country	Topsoil content [mg/kg]
Austria / Switzerland / Liechtenstein	0.19
France / Luxembourg	0.26
Netherlands / Denmark	0.21
Poland, Czech Republic, Italy, Belgium	0.38 (average Germany)

#### 4.3.2.4 Surface runoff

Rainfall runoff has to be considered the main component when quantifying the dissolved substances through surface runoff from unsealed areas. Precipitation lead to the discharge of dissolved and unsolved substances from the atmosphere and therefore to wet atmospheric deposition. A direct conclusion from general PAH depositions to substance concentrations in precipitation is not possible as the „dry“ share in the form of dust deposition often exceeds the „wet“ share often by more than one dimension (HELLMANN, 2004).

The  $\Sigma$  EPA-PAH<sub>16</sub> concentration in precipitation in Germany is 0.3 to 9.4 µg/L according to a bibliographical analysis by WELKER (2004) and is therefore characterised by a wide fluctuation margin. The average of the dominating concentration area according to WELKER (2004) is approx. 1.3 µg/L. Within the framework of the sewage disposal plan of Berlin (BERLIN, 2001) a wide value range of 0.25 to 1 µg/L was detected.

Apart from potential emission sources the different regionally dominating precipitation distribution and with that the choice of monitoring points has an essential influence. As it is assumed that in the case of surface runoff the PAH interacts with the soil substrate (sorption processes) for the observation of the total area of Germany the lowest measured value of 0.25 µg/L (comp. Table 39) is set.

Table 39:  $\Sigma$  EPA-PAH<sub>16</sub> concentration in precipitation

Concentration in precipitation [µg/L]	Reference
0.25 to 1	BERLIN (2001)
0.3 to 9.4	WELKER (2004)
1.3	Ø concentration Germany, derived from WELKER (2004)

According to an analysis of several publications by HELLMANN (2004) the occurrence of PAH in soils can especially be seen as the consequence of atmospheric input. Additional PAH input is caused by the use of sewage sludges and other fertilisers in farming (UBA, 2007a). But it cannot be assumed that there are any significant matter shifts into lower soil layers beneath the machining depth according to HELLMANN (2004).

#### 4.3.2.5 Groundwater

Even though PAH is ubiquitous in the environment no significant groundwater pollutions are expected beneath less extremely contaminated soils due to the low water solubility as well as the extensive sorption in humus and clay minerals close to the surface. On the basis of this assumption PAH is often found round point source loads which generally cannot be mapped by the „surveying monitoring networks“ of the federal states. Findings are therefore accordingly associated to special monitoring networks (abandoned hazardous sites, landfill site surveillance, control monitoring points in redevelopment areas and so on) (UBA, 2008c). Research in the groundwater database of the German Federal Environment Agency with 800 monitoring points for Germany in June 2008 showed that merely two federal states had reported data on individual PAH. The recorded measurement readings only rely on seven monitoring points. The reported values are all smaller than the respective limits of quantification which lie between 0.004 and 0.01 µg/l for the different PAH congeners (UBA, 2008b).

To comprise all relevant monitoring networks a survey of the responsible monitoring network centres was made for all 16 federal states in the period of 03/2008 to 07/2008 whereas not all federal states could provide data. As a basis of comparison for the data collected it can be assumed that the „normal concentration“ of  $\Sigma \text{PAH}_6$  is 50 ng/l in the groundwater according to HELLMANN (2004) whereas an anthropogenic influence can be assumed for an increase of more than the factor two. The German Working Group on water issues of the Federal States and the Federal Government (LAWA) mentions in the report on „deriving insignificance thresholds“ (12/2004) an ecotoxicologically justified insignificance threshold (Geringfügigkeitsschwellenwert, GFS) of 0.2 µg/l for the sum of  $\text{PAH}_{15}$  (according to  $\Sigma \text{EPA-PAH}_{16}$  without naphthalene).

#### Baden-Württemberg

According to the LFU (2002) the limit of quantification for  $\text{PAH}_6$  (TVO) is exceeded at 5.4% of the monitoring points in Baden-Württemberg whereas for individual compounds of  $\Sigma \text{EPA-PAH}_{16}$  the limit of quantification in average are only exceeded at 2.5 % of the monitoring points. The cause was identified as being abandoned hazardous sites. The limit of quantification of most individual PAH compounds was 0.005 µg/l (LFU, 2002). For first rough estimations of a  $\Sigma \text{EPA-PAH}_{16}$  sum parameter in the scope of a wide-spread observation a background value is assumed according to half of the limit of quantification. On the basis of the most common limits of quantification for individual compounds (0.005 µg/l) as well as the assumption of a percentage of indicator substance B(a)P of approx. 5 % of  $\Sigma \text{EPA-PAH}_{16}$  (SCHEFFER & SCHACHTSCHABEL, 2002; GÖTZ, 2008) an assumed background value for the sum parameter of 0.05 µg/L is derived which corresponds to the dimensions given by HELLMANN (2004). Attention should be paid to the positive results, especially if naphthalene, phenanthrene and fluoranthene were detected while the other compounds of TVO  $\text{PAH}_6$  or  $\Sigma \text{EPA-PAH}_{16}$  sum parameters are often not found or only to a small extent.

According to the LFU (2005) the results for Baden-Württemberg are, sorted by the detection frequency: naphthalene, phenanthrene, fluoranthene, pyrene, acenaphthene, fluorene, anthracene, chrysene, benzo(a)anthracene, benzo(a)pyrene, benzo(ghi)perylene, benzo(b)fluoranthene, indeno(1,2,3-cd)pyrene, benzo(k)fluoranthene, acenaphthylene and



dibenzo(ah)anthracenes. The actual ratio is subject to strong local variations as well as to physico-chemical properties (water solubility, etc.).

### **Mecklenburg-Western Pomerania**

In Mecklenburg-Western Pomerania the determination of PAH has not been part of the standard monitoring programme for groundwater for many years now. The sampling is always taken on a case-by-case basis and mainly in connection with the processing of abandoned hazardous site projects (MLUV, 2008). In the years 1993 and 1994 individual parameters were analysed at selected monitoring stations in the federal state monitoring network. There are results for 48 monitoring stations. For fluoranthene the limit of quantification of 0.005 µg/l exceeded eight times with an average concentration of approx. 0.019 µg/l (LOQ). For benzo(b)fluoranthene two individual measuring values of 0.013 and 0.006 µg/l were detected at a limit of quantification of 0.005 µg/l were found. The values for benzo(k)fluoranthene (LOQ = 0.005 µg/l), benzo(a)pyrene (LOQ < 0.005 µg/l) and indeno-(1,2,3-cd)pyrene (LOQ < 0.01 µg/l) fell below the limit of quantification (MLUV, 2008).

### **Schleswig-Holstein**

In Schleswig-Holstein no systematic PAH surveys of the groundwater are made by the federal state authorities (LANU, 2008). The background values typically fall below the limit of quantification. It is known that some of the water works in Schleswig-Holstein sporadically conduct PAH surveys for clear water and raw water, but there are no compilations or evaluation on them (LANU, 2008).

### **Rhineland-Palatinate**

In the scope of the groundwater surveillances by the State Agency for Environment, Water Management and Trade Control (LUWG) of Rhineland-Palatinate small amounts of surveys are made for PAH (1863 individual analyses) (LUWG-RP, 2008). In addition, data is available from a voluntary cooperation agreement from the raw water surveillance of the public water supply operators with 11473 individual analyses. It must be pointed out that the available measurement readings are raw unchecked data. A summary view of the total  $\Sigma$  EPA-PAH<sub>16</sub> parameter is not possible with this data. In the framework of the raw water as well as in the groundwater surveillance the PAH concentrations are analysed according to the drinking water ordinance (Trinkwasserverordnung TrinkwV), Annex 2, as far as possible. In the data on groundwater only 1.4 % proof of matter (mainly naphthalene) and in the collected raw water surveillance only 3.2 % proof of matter (mainly fluoranthene) were found for different limits of quantification. The highest detected values were 0.004 resp. 0.05 µg/l (LUWG-RP, 2008).

### **Bavaria**

Measurement readings for PAH concentrations in the groundwater are available for chosen monitoring points near populated areas from the Bavarian federal state monitoring network

for groundwater quality from the year 2007 (LFU-B, 2008). The analysis of the available data shows that the majority of PAH findings have concentrations below the limit of quantification. The limits of quantification for individual PAH were in the range of 0.003 to 0.02 µg/l. Moreover, only very few analyses had concentrations slightly above the respective limits of quantification (LFU-B, 2008).

## Berlin

In Berlin an average  $\Sigma$  EPA-PAH<sub>16</sub> concentration in the groundwater of 0.02 µg/l is detected on the basis of measurements from 1 299 monitoring points (BERLIN, 2008). Based on the used analysis methods a minimum of 0.0125 µg/l and a maximum concentration of 0.09 µg/l could be proven.

## Background values

SCHRAMM (2008) measured PAH in high mountain areas not or hardly anthropogenically affected. The background concentrations found in the groundwater are again a dimension below the values of the general groundwater surveillance (Table 40). As there are only very few measurement readings available, due to the very sensitive analysis methods, until now it is not possible to derive a reliable value concerning the wide-spread ubiquitous PAH concentration in groundwater. Besides, for the evaluation of PAH loads into waterbodies not the natural PAH concentration should be observed but also the share of PAH concentrations which are anthropogenic.

Table 40: PAH background concentrations (pg/l) in groundwater (SCHRAMM, 2008)

EPA-PAH	Concentration [pg/l]	Limit of detection [pg/l]
Naphthalene	15.3	0.06
Acenaphthylene	8.1	0.02
Acenaphthene	152.0	0.02
Fluorene	161.0	0.01
Phenanthrene	323.0	0.02
Anthracene	84.9	0.03
Fluoranthene	334.0	0.02
Pyrene	383.0	0.02
Benzo(a)anthracene	27.5	0.02
Chrysene	104.0	0.02
Benzo(b)fluoranthene, Benzo(j)fluoranthene	26.6	0.02
Benzo(k)fluoranthene	14.5	0.02
Benzo(a)pyrene	18.6	0.02
Indeno(1,2,3-c,d)pyrene	8.6	0.02
Benzo(g,h,i)perylene	18.2	0.02
Dibenzo(a,h)anthracene	1.4	0.01
<b><math>\Sigma</math> PAH<sub>16</sub></b>	<b>1,681</b>	

Based on the data it can be determined that most of the EPA-PAH<sub>16</sub> are lower than the limit of quantification for groundwater. Derived from half of the most common limits of quantification for individual compounds of 0.005 µg/l as well as the assumed percentage of the indicator substance B(a)P of approx. 5 % in  $\Sigma$  EPA-PAH<sub>16</sub>, a background concentration average of approx. 0.05 µg/l is found which corresponds with the specifications of HELLMANN (2004). For this value it needs to be mentioned that in practice it is assumed that the groundwater shows a quantitative dominance of the PAH congeners naphthalene, phenanthrene und fluoranthene.

Current measurements with low limits of quantification only amount to an average of 0.02 µg/l for the  $\Sigma$  EPA-PAH<sub>16</sub> sum parameter (BERLIN, 2008). For areas of pure air resp. background areas with low anthropogenic influence (comp. SCHRAMM, 2008) the values are assumed even lower. This can be seen as indication that if analysis methods are used with adequate limits of quantification then a lower background concentration can be proven, lower than often assumed. For further calculations for a wide-spread observation an average  $\Sigma$  EPA-PAH<sub>16</sub> concentration of 0.02 µg/l is therefore assumed for groundwater.

#### **4.3.2.6 Drainage**

There is only a small amount of data for PAH concentrations in drainage water. The topsoil is assumed to be the sink and sorbent for emissions due to its linkage to organic substances and clay minerals as well as its low water solubility (SCHEFFER & SCHACHTSCHABEL, 2002). Therefore it cannot be assumed that there is any significant shift into the subsoil after the soil infiltration (HELLMANN, 2004). As in the case of the groundwater an average  $\Sigma$  EPA-PAH<sub>16</sub> background concentration of 0.02 µg/l is assumed (comp. paragraph 4.3.2.4).

#### **4.3.2.7 Sewer systems**

The main sources of diffuse emissions through precipitation runoff from urban areas are atmospheric deposition, traffic (tyre abrasions, brakes, exhaust) as well as the pollution of impervious areas. Apart from the generally wide-spread atmospheric deposition the local contributions within the urban background charge such as dominating fuels in heating systems act as further influencing parameters.

The results of a substantial evaluation of bibliographical references by WELKER (2004) accordingly show a wide fluctuation range concerning the PAH concentrations in the runoff from urban areas (comp. Figure 37). The evaluated studies show that apart from the analysis (amount of considered PAH compounds) further essential influence factors such as location (e.g. country of origin, town, traffic routes) and point in time (e.g. year, season) could be identified. As the bibliographical references often only provide the pollutant concentrations for the stormwater runoff measurements (WELKER, 2004) it is necessary to derive the area-specific surface loads for PAH as well as the temporal development with the help of concentration specifications.

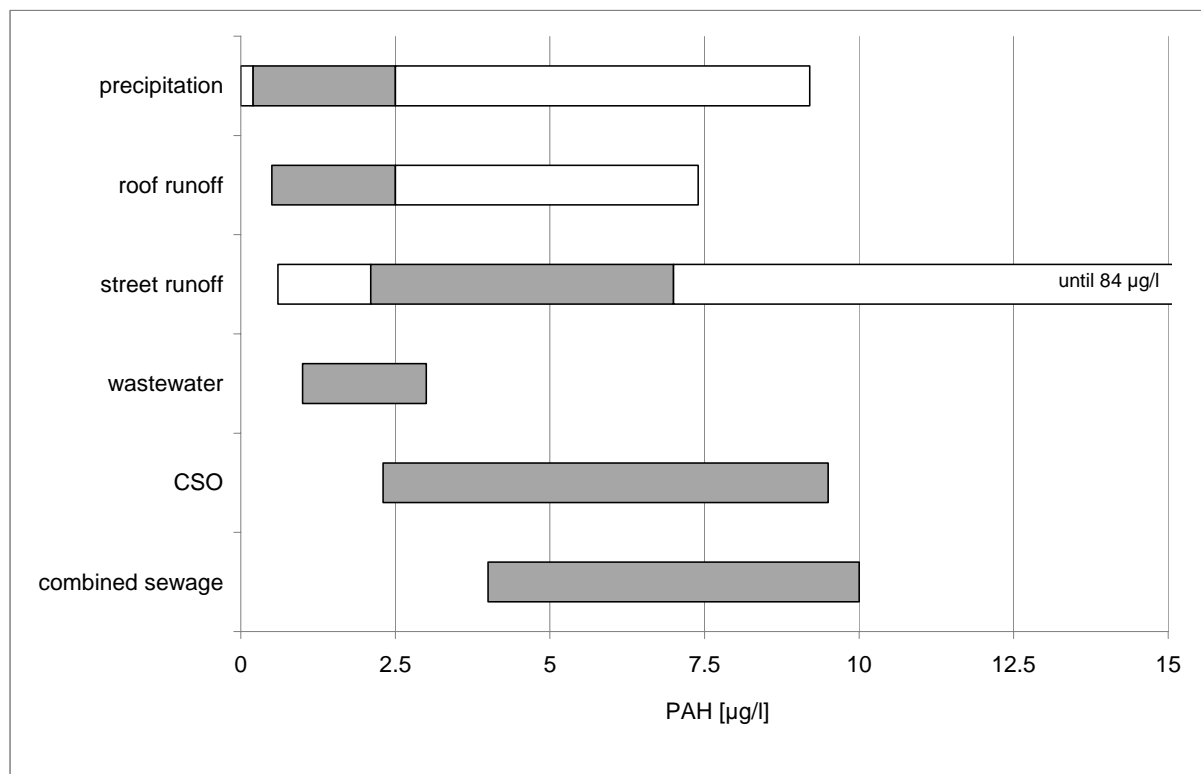


Figure 37: PAH concentrations in different urban runoff components (WELKER, 2004, edited)  
grey = most common concentration regions, white = max. fluctuation range / extreme values

### Input loads onto impervious areas through atmospheric deposition

For loads onto impervious urban areas from atmospheric deposition the data obtained from paragraph 4.3.2.2 can be used. According to this in Germany 6.8 g/(ha·a) of the specific surface load is caused by atmospheric deposition. On the basis of current data of the atmospheric monitoring network of the German Federal Environment Agency as well as the federal state monitoring networks in charge of atmospheric quality (GÖTZ, 2008) a regionally differentiated summary is available on a federal state level.

### Input loads onto impervious areas through traffic

Apart from the atmospheric deposition the emissions from the traffic sector are the second essentially potential PAH source for impervious urban areas. Emissions from traffic are generally split into two distribution types. One is the PAH from traffic that is gaseous or adsorbed to particulate  $PM_{10/2.5}$  which is transported over the atmospheric pathway that leads to atmospheric input loads. These can be seen as ubiquitous basic loads of the area-specific surface load. The other is a higher area-specific surface load in traffic areas due to PAH emissions in the form of particles from tyres and brake abrasions deposited directly on or next to the traffic area.

### *Concentrations in stormwater runoff water from traffic areas*

There are several publications on PAH concentrations in road runoff. In Germany these are especially made for motorways. Only recently road runoff from roads with less traffic are considered (WELKER, 2004).

The available measured data are generally characterised by a high variability whereby parameters such as the amount of vehicles per day or the population density only allow limited conclusions on actual input loads. In addition the often pronounced „first flush“ effect can be observed at the beginning of rainfall events. According to WELKER (2004) concentrations in road runoff in Germany can have  $\Sigma$  EPA-PAH<sub>16</sub> concentrations between 0.6 and 84 µg/l whereby the average concentration of the dominating concentration range of road runoff is approx. 4 µg/l ( $\Sigma$  EPA-PAH<sub>16</sub>). Within the framework of a data collection by the IWG (2008) an average concentration of 3.5 µg/l ( $\Sigma$  EPA-PAH<sub>16</sub>) could be determined in road runoff.

Table 41:  $\Sigma$  EPA-PAH<sub>16</sub> concentrations in road runoff

Road runoff	Highway runoff	Reference
0.24 to 3.1 µg/l	0.54 bis 21.8 µg/l	BERLIN (2001)
0.6 to 84 µg/l		WELKER (2004)
4.0 µg/l (average for Germany)		WELKER (2004)
3.5 µg/l		IWG (2008)

With the MONERIS model, which apart from calculating the emissions can also estimate the effluent from storm sewers, area-specific surface loads can be back-calculated from concentrations. Based on the assumption of PAH effluent concentrations of 3.5 µg/l according to the specifications given by the IWG (2008) (comp. Table 41) an average surface load of 10.9 g/(ha·a) for urban areas can be calculated. It has to be pointed out that the data available so far does not allow further differentiations of the urban areas (e.g. into traffic areas, roof areas, etc.). As already depicted obvious differences between the different surface categories are assumed. This is why the known PAH loads and area specifications are used to check whether the area-specific surface loads found for traffic areas also apply to other surface categories.

### *Area-specific surface load from tyre and brake abrasions*

After the first relevant studies on this topic from the Swedish Chemicals Agency (KEMI, 2003) the PAH emissions from tyre abrasions were examined at the end of the 1990s in the scope of the BMU research project “Exemplary coverage of the exposure of selected rubber derivatives conventionally used in tyres to the environment and their disposal” (UBA 1998) by the German Federal Environment Agency. The study came to the result that in Germany tyre abrasions have already emitted approx. 6 to 18 tons of PAH into the environment and therefore have already exceeded the PAH emissions by vehicle diesel exhaust since 1988 (UMWELT, 2006). Current specifications of the UN ECE-CLRTAP emission report (UBA, 2008c) set emissions of the PAH indicator parameter B(a)P for the year 2006 for the traffic sector at approx. 285 kg allotting 29 kg to tyre and brake abrasions. The calculation assumes a B(a)P concentration of 3.9 mg/kg for tyre abrasions and 0.74 mg/kg for brake abrasions (UBA, 2008a).

The assumption of a B(a)P share of approx. 5 % in  $\Sigma$  EPA-PAH<sub>16</sub> leads to a calculated total emission of the sum parameter of approx. 5.7 tons annually from road traffic with a share of approx. 580 kg annually coming from tyre and brake abrasions. The dimension of the given emissions largely corresponds to the concentrations previously published by the PRTR (2007) (comp. 4.3.2.2). It must be pointed out that within the framework of the UN ECE-CLRTAP and the PRTR only the emission pathway via the atmosphere is considered.

In contrary to questions on air pollution control and the distribution via the atmospheric pathway the total amount of tyre abrasion must be considered for the potential loads in urban areas. It is assumed that particles that are not transported via the atmospheric pathway accumulate in the dust on the roads. Stormwater runoff can transport them into roadside soils, surface waters or the sewer systems depending on the available drainage. When evaluating these PAH sources the chosen emission factors for tyre abrasions in connection with the vehicle category and mileage as well as the assumed PAH concentrations in tyres are to be seen as important (NTZIACHRISTOS, 2003). As pointed out above, concentrations for the PAH indicator substance B(a)P are assumed to be 3.9 mg/kg for tyre abrasions and 0.74 mg/kg for brake abrasions (UBA, 2008a). Regarding the total abrasion amounts 111,420 t/a for tyre abrasions and 12,350 t/a for brake pad abrasions are used according to HILLENBRAND ET AL. (2005) (comp. Table 42).

Table 42: Benzo(a)pyrene (B(a)P) emissions from brake and tyre abrasion

Source	B(a)P [mg/kg]	Abrasion [t/a]	B(a)P-load [kg/a]	Reference
Brakes	0.74	12,350	9	UBA (2008a) / HILLENBRAND ET AL. (2005)
Tyres	3.90	111,420	435	UBA (2008a) / HILLENBRAND ET AL. (2005)
<b>Sum</b>			<b>444</b>	

The calculated total emissions from tyre and brake abrasions from road traffic in Germany amount to 444 kg B(a)P annually (comp. Table 42). Compared to the recorded B(a)P-emissions of 36.7 t/a for the atmospheric pathway by the UBA (2008a) this value corresponds to the total emitted amount by approx. 1.2 %. Regarding the trans-medium total emissions of B(a)P only a small relevance of the source „tyre and brake abrasions“ is given under the most up-to-date available emission values on the total emission amount.

The relevance of PAH emissions by abrasion products in traffic areas can be evaluated with the help of the contribution to the area-specific surface load. The PAH area-specific surface load due to abrasion is calculated on the basis of the total emissions in road traffic in Germany of 435 kg B(a)P annually from tyre abrasions as well as 9 kg B(a)P annually of brake abrasions (comp. Table 42). The traffic areas in Germany is approx. 1,744,600 hectares for the year 2004 (DESTATIS, 2008a). For further calculations it is assumed that the abrasions remain in the direct proximity of those traffic areas. On the basis of the observed data a calculated average of B(a)P input load onto traffic areas of 0.254 g/(ha·a) is given which, assuming a B(a)P share of approx. 5 % in  $\Sigma$  EPA-PAH<sub>16</sub>, leads to average loads of the sum parameters of approx. 5.1 g/(ha·a) (comp. Table 43).

Table 43: Benzo(a)pyrene (B(a)P) /  $\Sigma$  EPA-PAH<sub>16</sub> emissions from abrasion products from traffic areas

Source	B(a)P [kg/a]	Traffic areas 2004	Surface load [g/(ha·a)] ( $\Sigma$ EPA-PAH <sub>16</sub> )
Brakes	435	1,744,600	0.249
Tyres	9	1,744,600	0.005
Sum	444	1,744,600	0.254
Assumption: B(a)P-share of 5 % in $\Sigma$ EPA-PAH <sub>16</sub>			<b><math>\Sigma</math> EPA-PAH<sub>16</sub> = 5.1</b>

*Total surface load of traffic areas in Germany*

The total surface load for traffic areas in Germany according to current knowledge consists of the atmospheric deposition of approx. 6.8 g/(ha·a) as well as the PAH loads of approx. 5.1 g/(ha·a) due to abrasion. This leads to an average area-specific surface load of approx. 11.9 g/(ha·a). This very much coincides with the surface load of 10.9 g/(ha·a) that was calculated above for traffic area runoff with MONERIS.

Due to legal regulations changes in the absolute level are already clearly noticeable and will be in future and will also show shifts of the relevance of the input loads. With the help of previous publications the calculated share of tyre and brake abrasions amounts to 42.9 % of the total PAH surface load of traffic areas while 57.1 % are allotted to atmospheric deposition. After an initiative of Germany and Sweden restrictions for PAH in tyres used in road traffic were passed in the directive 2005/69/EG on a European level on the 16th November 2005. According to this from the year 2010 onwards only oils with softening agents with <1 mg/kg B(a)P as well as with <10 mg/kg for the sum of all PAH are allowed. In addition all these thresholds apply to imported tyres. As a consequence of the implementation of the directive 2005/69/EG several current tyre models already show a reduced PAH charge (ADAC, 2008). Till the year 2012 the reduction of PAH emissions from tyres is expected to be more than 95 % (UMWELT, 2006). Based on this prognosis it is further expected that it will only come up to approx. 0.4 g/(ha·a)  $\Sigma$  EPA-PAH<sub>16</sub> of the surface load for traffic areas (comp. Table 44). Should the atmospheric deposition of approx. 6.8 g/(ha·a) and the future surface load of 7.2 g/(ha·a) stay the same, then from 2012 onwards PAH emissions from abrasions would only have a share of approx. 5.6 %.

Table 44: Prognosis benzo(a)pyrene (B(a)P) /  $\Sigma$  EPA-PAH<sub>16</sub> emissions 2012 (tyre abrasions PAH-95 %)

Source	B(a)P [kg/a]	Traffic areas 2004	Surface load [g/(ha a)] ( $\Sigma$ EPA-PAH <sub>16</sub> )
Brakes	21.75	1,744,600	0.012
Tyres	9.00	1,744,600	0.005
Sum	30.75	1,744,600	0.018
Assumption: B(a)P-share of 5 % of $\Sigma$ EPA-PAH <sub>16</sub>			<b><math>\Sigma</math> EPA-PAH<sub>16</sub> = 0.4</b>

Bibliographical references describe the annual  $\Sigma$  EPA-PAH<sub>16</sub> abrasion rates to be 1.5 g/(ha·a) for traffic areas and 5 to 18 g/(ha·a) for motorways (BERLIN, 2001) resp. 4 to



5 g/(ha·a) for roof areas, 6 to 21 g/(ha·a) for general traffic areas and 5 to 10 g/(ha·a) for „other: bicycle paths and pavements, yards“ (SCHÄFER, 1999).

The calculated surface load of traffic areas is 10.9 g/(ha a). But traffic areas take up only approx. 38 % of the area category „residential and traffic areas“ (DESTATIS, 2008b). 72 % of the area is allotted to the use as „building space and open spaces“, „production area without mining land“, „recreational area“ as well as „cemetery“ (DESTATIS, 2008b). It can be assumed that for these areas the dominating influence is the atmospheric deposition with approx. 6.8 g/(ha·a). For further calculations of the pathway „sewer systems“ in MONERIS the mean of 9 g/(ha·a) is used which was calculated as area weighted value.

#### **4.3.2.8 Inland navigation / emissions via products**

Even though the use of PAH has largely been limited for different products such as clay pigeons, adhesives containing tar, tar oil in anti-corrosion paints, softening agents in rubber products, an agent in moth balls and in other special products, emissions from end-of-life products still emit into the environment and waterbodies.

The emissions from tar-containing paints on ships, which had some importance in earlier years (GANDRASS & SALOMONS, 2001), don't play a role any more due to the regulated maintenance periods. The PAH emissions from ships' exhaust generally reach the air via exhaust pipes or chimneys (MAN DIESEL, 2008; ZKR, 2008) and therefore indirectly contribute to the water pollution via atmospheric deposition. In contrast outboard motors cause significant direct emissions into waterbodies as the exhaust is lead directly into the water (HORN ET AL., 2005). HORN ET AL. (2005) calculated the water emissions to be between 48 and 216 mg PAH<sub>16</sub> when running a motor for half an hour. Using the value 48 mg as best case and estimating approx. 200,000 boats with outboard motors in Germany as well as an average usage of approx. 60 hours annually leads to a total load of approx. 1.2 t  $\Sigma$  EPA-PAH<sub>16</sub> annually. Due to the small amount of available measurements this value, however, is uncertain.

For calculating the emissions from outboard motors only the water surfaces of navigable sections of first order rivers were considered. In follow-up projects maps of navigable waterways should be blended with model river area maps to obtain the navigable areas.

Additionally, the emissions from old anti-corrosion paints containing PAH from steel construction for hydraulic engineering were estimated in cooperation with the Federal Waterways Engineering and Research Institute (BAW) in Karlsruhe. For this an internal BAW database was used which contains information on the state of the old paintwork, the treated area and the location of the structure. In the past paints were used containing coaltar pitch (Steinkohlenteerpechhaltige Farben, STKP) that were followed by tar epoxides (Teerepoxide, TE) and then by epoxide-tar substitute (Epoxid-Teerersatzstoffen, EP-TE). Due to this the concentrations of the especially hazardous contents were reduced, measured as benzo[a]pyrene equivalents from approx. 30,000 (STKP) via 5,500 (TE) to approx. 600 (EP-TE). The corresponding concentrations in the different waste from sandblasting fabricated parts are depicted in Figure 38 (BAW, 2007).

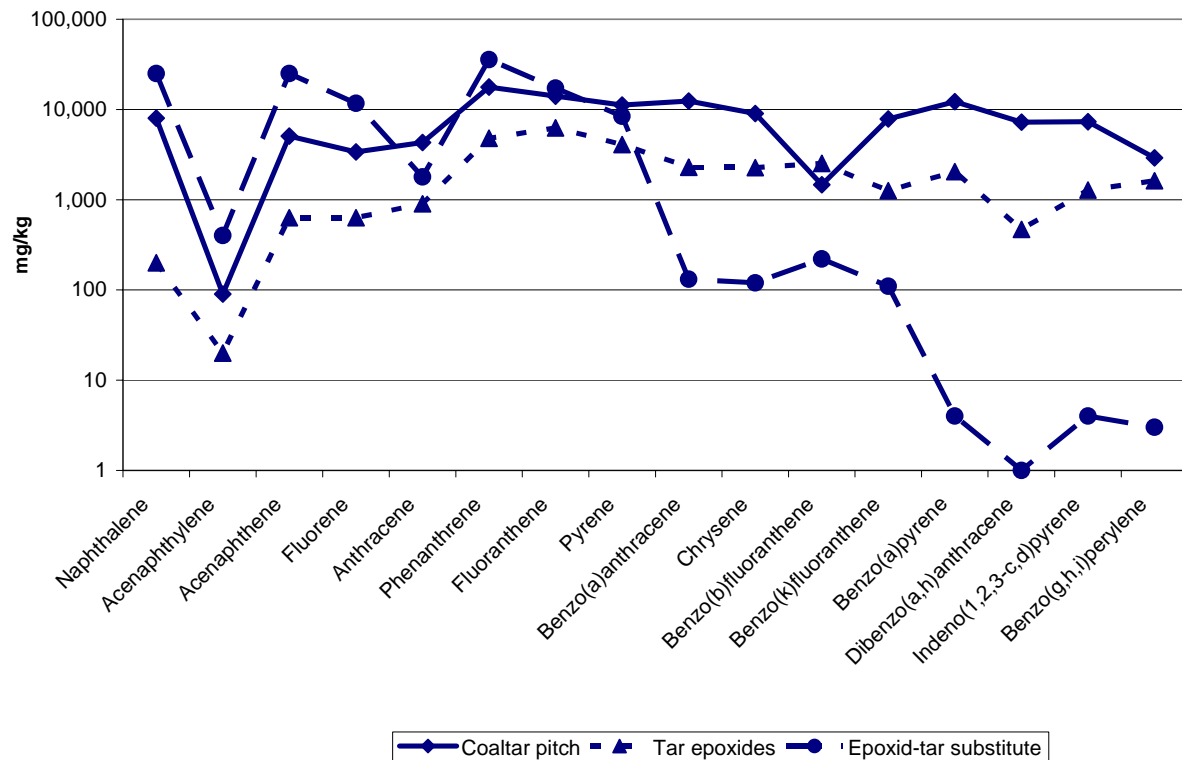


Figure 38: PAH-concentrations in blasting waste from different tar-based coatings

From the BAW database information: the average perforation corrosion, the average surface of fabricated parts as well as the assumption of a specific weight of the paints of  $1.2 \text{ g/cm}^3$ , a layer thickness of 0.5 mm and a lifetime of 20 years, a loss of paint on the rusty parts of about 45 kg annually. In addition emissions from PAH diffusing from the paint layer into the water can occur. But this is not easy to estimate as there are no according analyses. It is known from other bibliographical references that especially in old paints mainly water-soluble PAH such as naphthalene (is with approx. 30 mg/l by far the most water soluble PAH), acenaphthylene (3.9 mg/l), acenaphthene (3.4 mg/l), fluorene (1.7 mg/l), phenanthrene (1.1 mg/l) and anthracene (0.07 mg/l) (HENZLER, 2004) elute. Taking the worst-case estimation that all fabricated parts in the database are treated with coal tar pitch and that phenanthrene and anthracene, which are in STKP with approx. 5 % (MAYER & LEWIS, 2004), are washed out by a third as in train tracks (KOHLER & KÜNNINGER, 2003), then calculations lead to a loss of 0.35 tons annually from coatings. In total the estimated emissions from loads from corrosion treatment for steel constructions in hydraulic engineering for the year 2004 amount to approx. 400 kg. This amount is decreasing due to the declining use of paints containing PAH. The distribution of the 400 kg over the MONERIS analytical units was carried out as for recreational crafts.

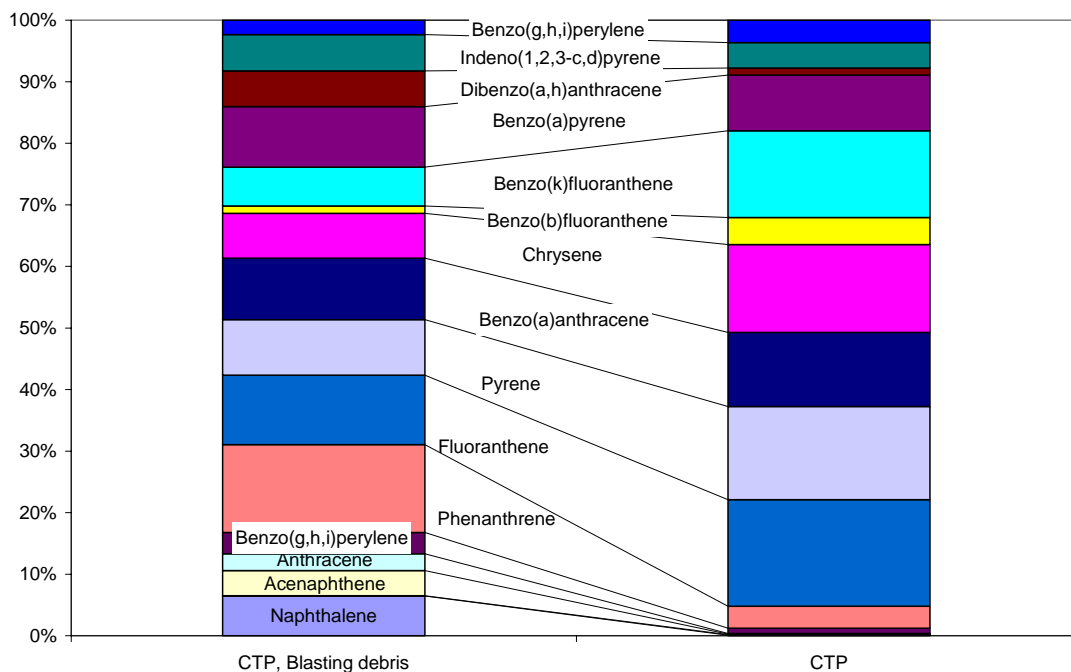


Figure 39: Comparison of the composition of blasting waste of paints containing coaltar pitch and coaltar pitch

In addition emissions from special applications hard to substitute still have to be considered, such as using creosote containing PAH to impregnate railway sleepers and wooden masts or emissions from processing bitumen in road construction.. In these fields of application the PAH emissions have been reduced by technical solutions such as appropriate fractioning for creosote or processing bitumen at lower temperatures.

Despite fundamental efforts to avoid products with considerable amounts of PAH to reach consumers, PAH is often found in rubber handles or other rubber parts of torches, window wipers or other tools, in certain cases even up to percentages (TEST, 2006). Corresponding PAH emissions from creosote and consumer products generally do not directly reach the waterbodies but primary the soil resp. the atmosphere (KÖHLER & KÜNNINGER, 2003).

## 5 Results

The following chapter will show the model results. A detailed description of the results for the analytical units can be taken from a web-based graphical user interface.

### 5.1 Calibration and validation of area runoff

In Figure 40 the position for the discharge monitoring stations used for calibrating and validating is depicted. The runoff calibration is made on the basis of the average discharge at 155 monitoring stations for the period of 1983 to 2005 (comp. paragraph 2.3).

The modelled discharges are added up based on the runoff equation (FNE, comp. paragraph 2.2) for the calibration and validation, whereas splitting of the discharge due to natural and artificial river bifurcations has to be considered (comp. paragraph 2.2). Altogether five splittings were integrated into the runoff equation within Germany (Table 45).

Table 45: Average transition of discharge from the main tributaries into canals in percent of the runoff in the headwaters

Canal	Average transition (% of discharge)
Ijssel-Canal	13 %
Nordumfluter (Spreewald)	80 %
Dahme-Umflut-Canal	26 %
Oder-Spree-Canal	40 %
Teltow-Canal	70 %

Table 46 depicts the average deviation, the coefficient of determination and the efficiency of the model between the modelled and measured discharges for the calibration and validation of the individual years for the long-term average (1983-2005). Figure 41 graphically shows the comparison of modelled and measured long-term average discharges. Figure 42 shows the spatial distribution of the average area runoff between 1983 and 2005.

The calibration for long-term averages as well as for individual years shows average deviations between the calculated and measured discharges of less than 10 %. Even though it was tried to reduce the error, errors are inevitable for some stations under the premise of correct balances and considering realistic specific runoff. In addition, water abstractions and discharges that weren't recorded could also be the reason for the deviations. A comparison of the modelled discharge with the hydrological atlas (BMU, 2003) proves that the characteristic distribution of area runoff in Germany (e.g. due to orographically caused rain shadow or orographic rainfall) is mapped well.

The average deviation from modelled to measured long-term average discharge for the validation lies around 28 % (Table 46). Especially in the 80s and at the beginning of the 90s there were a lot less stations available in comparison to the years following. For the validation discharge monitoring stations from considerably smaller catchment areas were used than for the calibration. This causes considerably higher deviations between the calculated and measured discharges because the measured discharge from smaller rivers is liable to be

more uncertain due to the higher dynamics (Figure 41). But the average deviation between the calculated and measured discharge for 16 of the 23 years is clearly below 25 %. Nevertheless, the validation for the individual years revealed an excellent statistical match ( $r^2$  and EF value) between the calculated and measured discharge (Table 46).

In total the model efficiency achieves high values according to Nash-Sutcliffe, which can be attributed to the good concordance of the simulated and measured discharge in large river basins. The model efficiency is strongly dominated by the quality of the predictions in the higher value ranges, while the deviations in the lower value range are of hardly any consequence. In contrast all deviations between simulated and observed values enter the average deviation. Due to the wide distribution in the lower value range the average deviation can be quite high even though the model efficiency reached a good value.

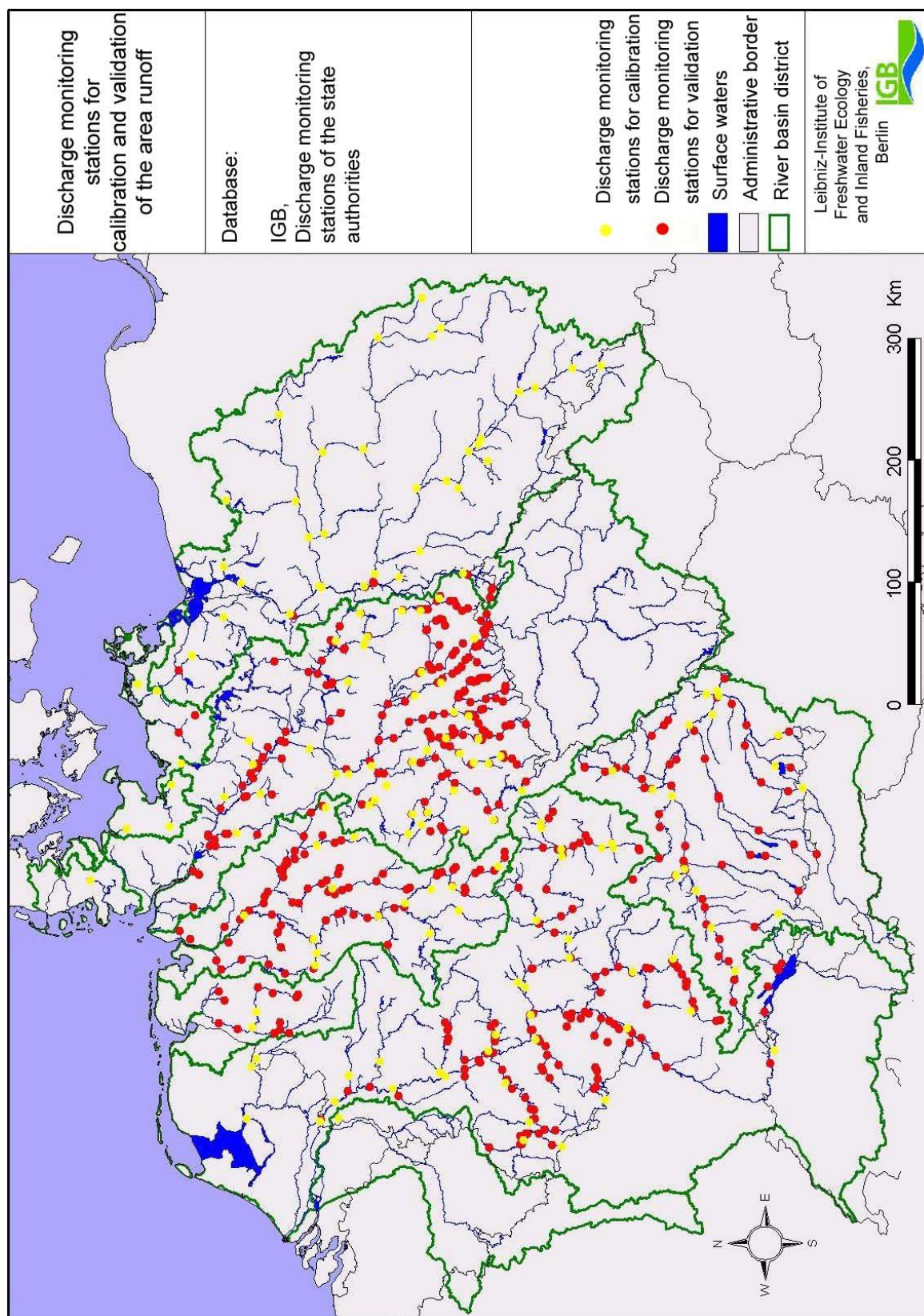


Figure 40: Discharge monitoring stations used for calibration and validation

Table 46: Overview over average deviation, coefficient of determination ( $r^2$ ), model efficiency (EF value) between the calculated and measured discharges for the stations (amount) considered for the calibration and validation in the individual years between 1983 and 2005 and in the long-term average (long-term)

Year	Calibration				Validation			
	Average deviation [%]	Amount	$r^2$	EF-value	Average deviation [%]	Amount	$r^2$	EF-value
1983	9.2	155	1.00	1.00	16.0	65	1.00	1.00
1984	9.4	155	1.00	1.00	15.5	86	1.00	1.00
1985	9.0	155	1.00	1.00	24.0	88	1.00	0.99
1986	8.9	155	1.00	1.00	17.8	88	1.00	1.00
1987	8.8	155	1.00	1.00	15.2	92	0.97	0.96
1988	8.8	155	1.00	1.00	20.5	97	0.99	0.99
1989	9.8	155	1.00	1.00	20.8	99	1.00	0.99
1990	8.6	155	1.00	1.00	19.0	118	0.99	0.99
1991	9.4	155	1.00	0.99	19.2	125	1.00	0.99
1992	8.7	155	1.00	1.00	22.3	133	1.00	1.00
1993	8.9	155	1.00	1.00	21.1	144	1.00	0.99
1994	8.6	155	1.00	1.00	24.5	143	0.99	0.99
1995	8.8	155	1.00	1.00	23.7	156	0.99	0.99
1996	8.8	155	1.00	1.00	24.9	156	1.00	1.00
1997	9.1	155	1.00	1.00	23.9	157	1.00	1.00
1998	8.7	155	1.00	1.00	31.5	279	1.00	1.00
1999	8.9	155	1.00	1.00	31.0	323	1.00	1.00
2000	9.0	155	1.00	1.00	33.8	323	1.00	1.00
2001	8.3	155	1.00	1.00	29.7	311	1.00	1.00
2002	8.1	155	1.00	1.00	29.3	296	0.99	0.99
2003	9.0	155	1.00	1.00	26.0	292	1.00	1.00
2004	9.7	155	1.00	1.00	31.3	286	1.00	1.00
2005	9.0	154	1.00	1.00	11.0	156	0.99	0.99
long-term	8.9	155	1.00	1.00	27.9	513	0.98	0.96



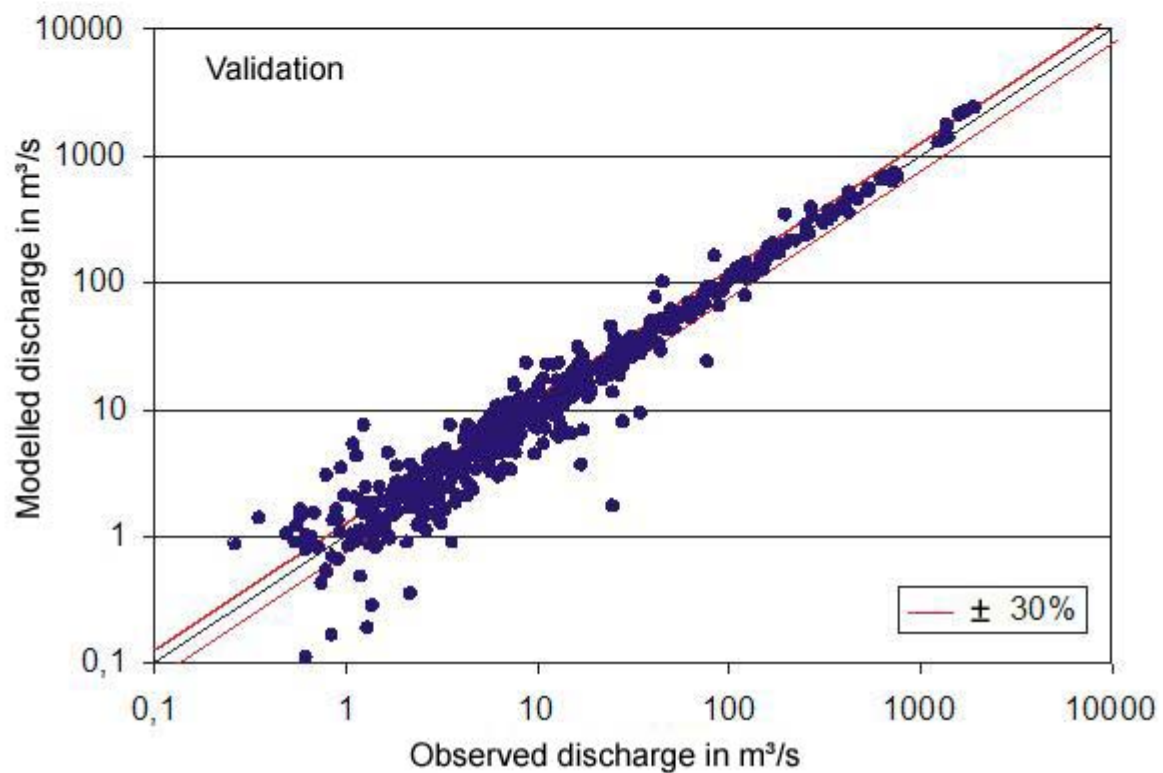
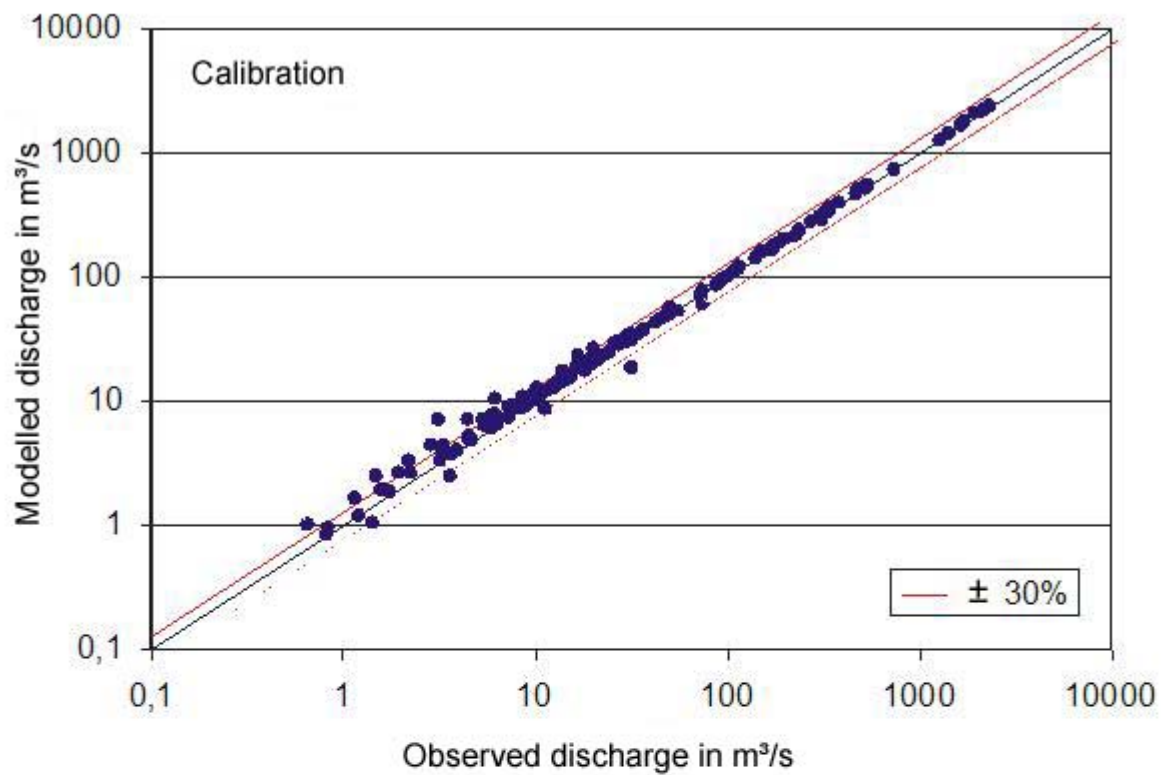


Figure 41: Comparison of the calculated and measured average discharge in the years 1983 to 2005 (long-term) for the stations considered for calibration and validation

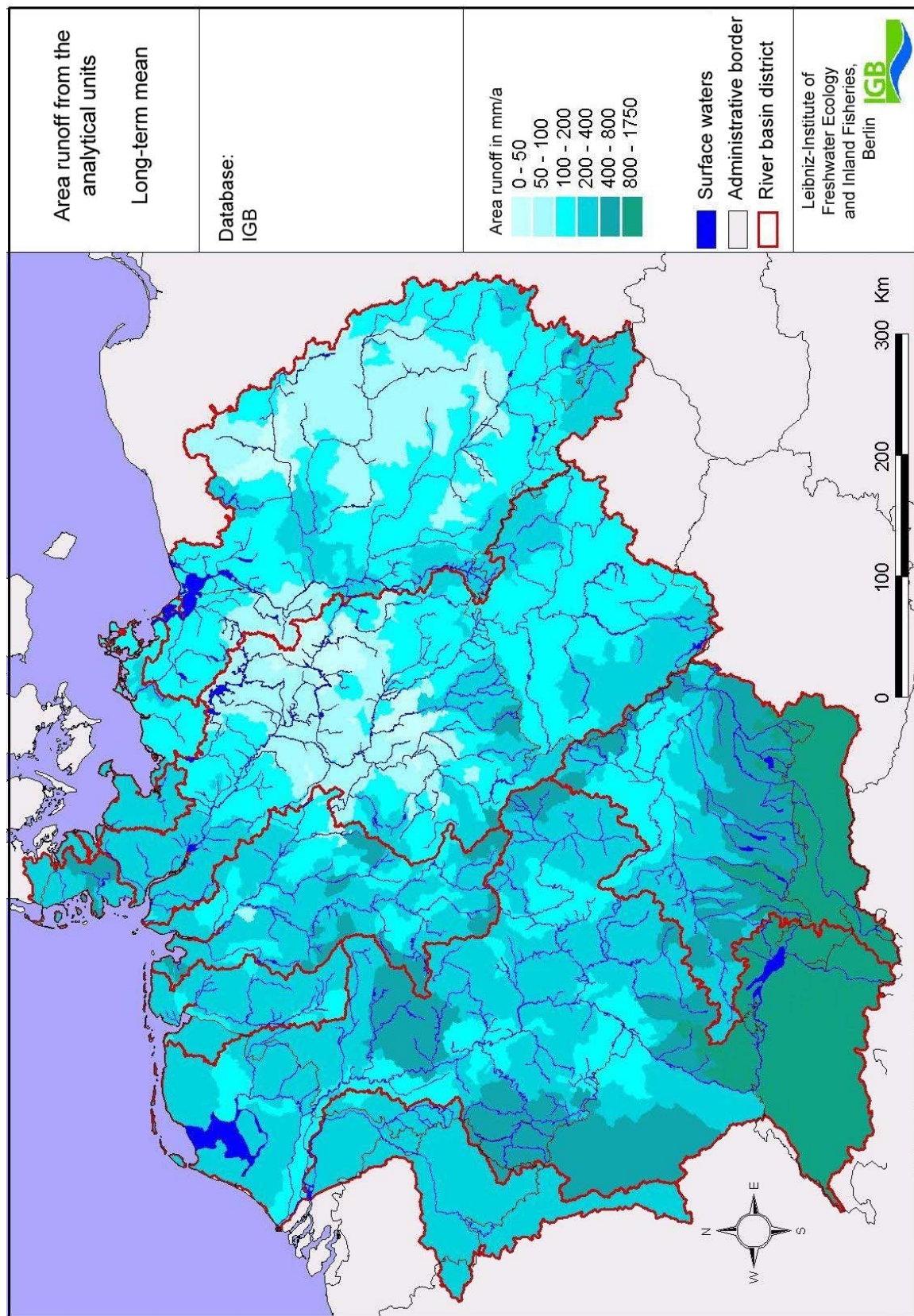


Figure 42: Spatial distribution of the long-term average area runoff between 1983 and 2005

## 5.2 Nutrients

### 5.2.1 Emissions into surface water

The emissions of total nitrogen (TN) and total phosphorus (TP) were calculated for the individual years between 1983 and 2005 and subsequently aggregated to the periods 1983-1987, 1993-1997, 1998-2002 and 2003-2005. The period 1988-1992 was left out of the valuation of the results as in these years there were considerable changes due to the German reunification for example in the land use or for the point source dischargers. Accordingly the input data is quite uncertain. Altogether the emissions from 332 (TN), 413 (DIN) and 339 (TP) stations could be calculated.

The total emissions described in the following always refer to the German analytical units, unless otherwise mentioned. A further spatial difference is based on the river systems (RB). The results for the North Sea and the Baltic Sea only refer to direct catchment areas of the coastal regions that aren't already covered by other river basins.

A clear reduction of emissions for TN and TP was calculated for the research period. The TN emissions were reduced from 1,031 kt/a (1983-1987) to 565 kt/a (2003-2005) by about 45 % (Figure 44, Table 47). The reduction for TP was even higher. Here the emissions were reduced from 79.5 kt/a to 22.2 kt/a by about 72 % (Figure 45, Table 48). The emissions reduction does strongly vary between the river basins whereas the highest reductions were achieved in the areas directly influenced by the reunification (Elbe and Odra) (Table 47, Table 49). It can be stated for all river basins that the largest reduction of emissions can be calculated for the period mid 80s to mid 90s. In the following years further reductions followed but these are significantly lower than in the first half of the calculation period. Especially in the second half the calculated emissions varied strongly. The differences in the emissions for the individual years of a period are mainly due to rainfall and the resulting changes of the discharge and runoff situation. Accordingly, the influence of runoff changes are in the same dimension as the changes in the anthropogenic factors for the second half of the research period (Figure 43). This is confirmed by comparing the calculated emissions for long-term average area runoff (long-term, LT), the absolute minimum of annual area runoff (dry year, DY) and the absolute maximum of the annual area runoff (wet year, WY).

In comparison with the LT-emissions the DY-conditions had emissions calculated to be 26 % (TN) and 16 % (TP) lower. On the other hand WY conditions showed emissions of up to 36 % (TN) and 23 % (TP) higher (Table 49, Table 50). It is clear, therefore, that the climatic/hydrologic regimes have a higher influence on the TN-loads than on the TP-loads. The reason for this can be seen in the fact that phosphorus has an essential share in the total emissions from point sources. These are not influenced as strongly by precipitation as emissions from diffuse sources.

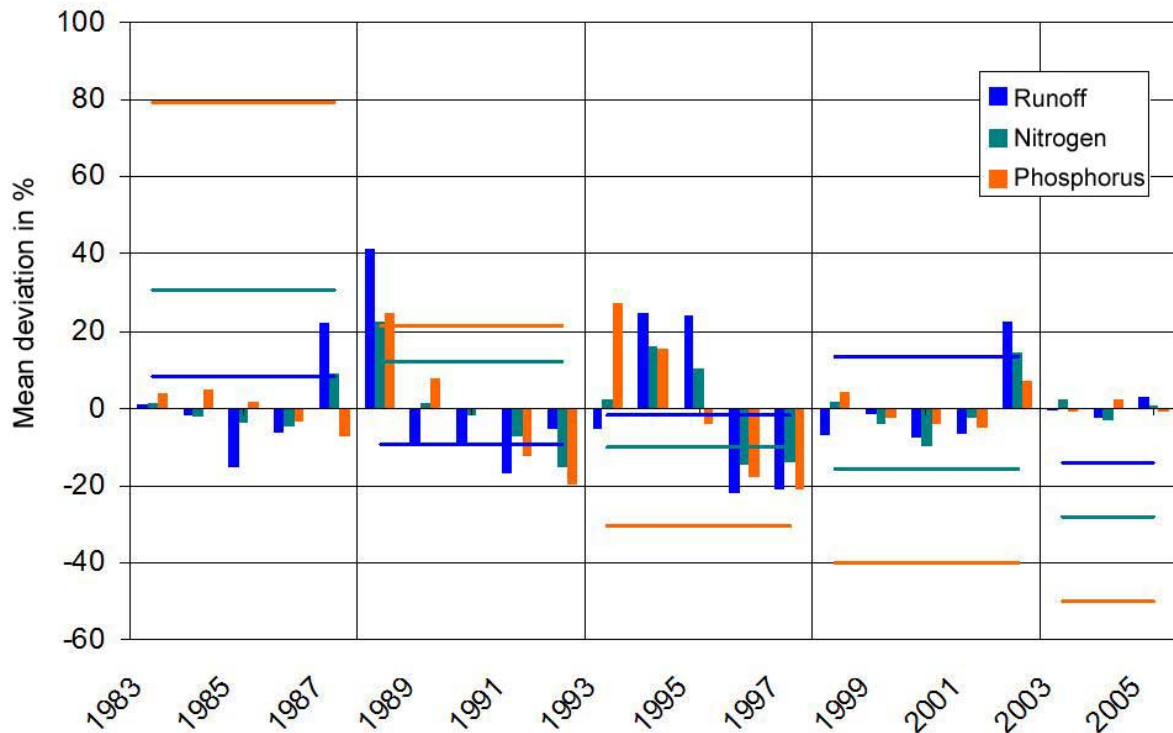


Figure 43: Change of runoff, total nitrogen (TN) emissions and total phosphorus (TP) emissions in the observation period  
 Lines: changes in the average runoff and emissions of one period in relation to the average conditions in the complete period (1983-2005). Bars: changes in the runoff and emissions of the individual years in relation to the average of that period

The emissions in the years 1983-1987 show a distinct distribution pattern after which specific emissions of < 15 kg TN/(ha·a) and < 50 kg TP/(km<sup>2</sup>·a) were calculated in the catchment areas of Elbe, Odra and Weser – with the exception of the emissions from larger cities. South of the Weser the emissions are far above 20 kg TN/(ha·a) and 100 kg TP/(km<sup>2</sup>·a) (Figure 46, Figure 47).

If this is compared to the emissions in the period of 2003 to 2005, it can be stated that the largest absolute reduction took place in the area of the Middle Rhine Highlands (Rhenish Slate Mountains, Westerwald, Harz, Thuringerwald and Black Forest) (Figure 48, Figure 49). On the contrary the reductions in the South of the Swabian Alb and the Bavarian Forest (southern Danube catchment area) as well as in many areas of North Rhine-Westphalia (especially in the Ruhr area) are significantly lower.

Apart from the reduced emissions another change was detected for the share of individual pathways in total emissions. On the one hand the share of pathways differ strongly between the river basins (Figure 50, Figure 51). On the other hand it can be seen that the emissions from sewer systems have been reduced considerably. In the period 1983-1987 the sewer systems in Germany accounted for 40 % (TN) and 85 % (TP) of the total emissions. In the period of 2003-2005 the share dropped to 20 % (TN) and 50 % (TP). Accordingly the share for emissions from arable areas increased.



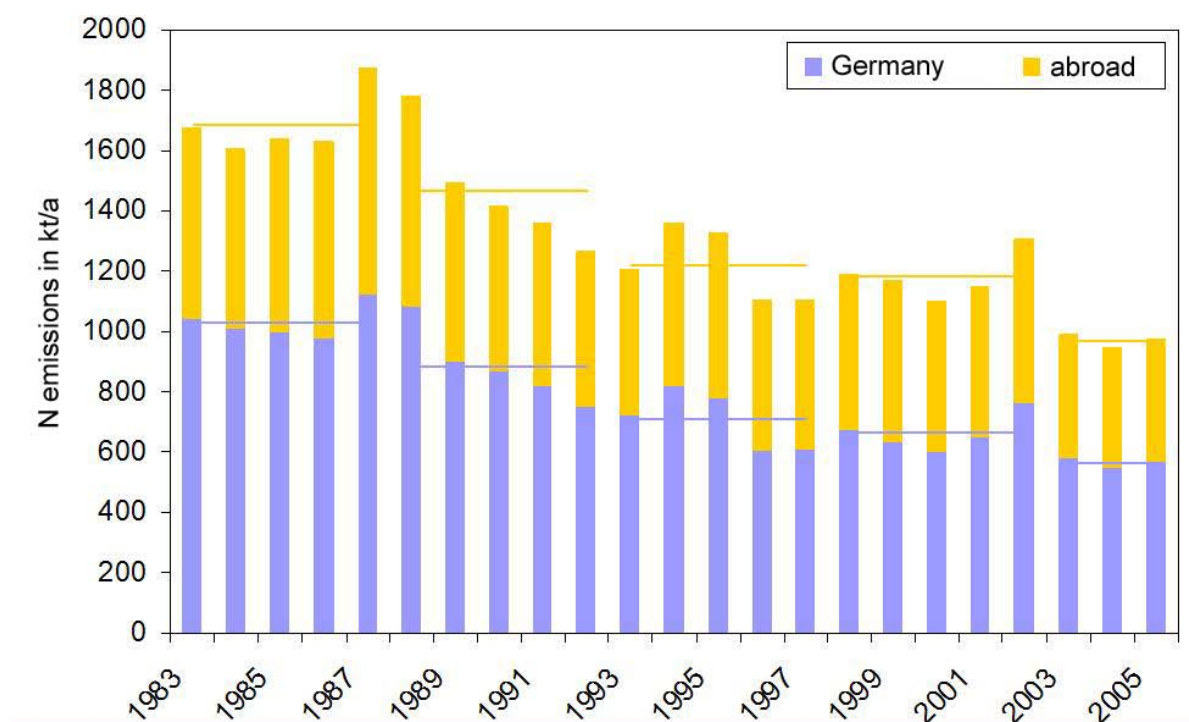


Figure 44: Change of the total nitrogen emissions (TN) in Germany and the parts of the river basins outside Germany in the years 1983-2005

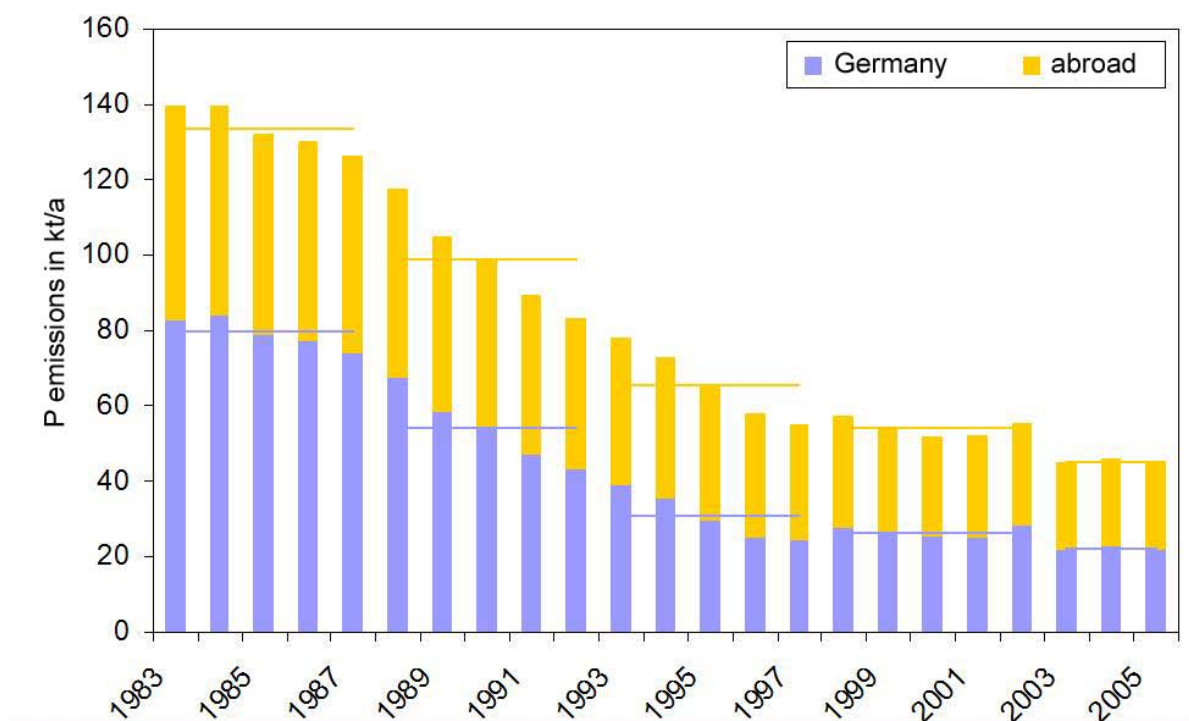


Figure 45: Change of the total phosphorus emissions (TP) in Germany and the parts of the river basins outside Germany in the years 1983-2005

Table 47: Nitrogen emissions into the German river basins for the periods 83-87, 93-97, 98-02 and 03-05 and their changes

River basin		Nitrogen			
		1983-1987	1993-1997	1998-2002	2003-2005
Danube	Emissions in kt/a	162.1	128.7	136.2	115.3
	%-reduction zu 83-87	0.0	-20.6	-16.0	-28.8
Rhine	Emissions in kt/a	397.2	283.5	255.1	201.5
	%-reduction zu 83-87	0.0	-28.6	-35.8	-49.3
Ems	Emissions in kt/a	32.8	31.5	28.2	26.3
	%-reduction zu 83-87	0.0	-4.0	-14.3	-19.9
Weser	Emissions in kt/a	119.1	92.0	81.4	69.2
	%-reduction zu 83-87	0.0	-22.8	-31.7	-41.9
Elbe	Emissions in kt/a	238.4	126.1	115.4	104.2
	%-reduction zu 83-87	0.0	-47.1	-51.6	-56.3
Odra	Emissions in kt/a	27.4	11.4	13.2	12.8
	%-reduction zu 83-87	0.0	-58.3	-51.9	-53.4
North Sea	Emissions in kt/a	18.4	15.2	16.0	16.8
	%-reduction zu 83-87	0.0	-17.3	-13.2	-8.8
Baltic Sea	Emissions in kt/a	35.6	19.3	19.6	18.6
	%-reduction zu 83-87	0.0	-46.0	-44.9	-47.7
Germany	Emissions in kt/a	1031.1	707.6	664.9	564.8
	%-reduction zu 83-87	0.0	-31.4	-35.5	-45.2

Table 48: Phosphorus emissions into the German river basins for the periods 83-87, 93-97, 98-02 and 03-05 and their changes

River basin		Phosphorus			
		1983-1987	1993-1997	1998-2002	2003-2005
Danube	Emissions in kt/a	10.0	4.7	4.7	3.9
	%-reduction zu 83-87	0.0	-53.2	-52.7	-61.1
Rhine	Emissions in kt/a	35.0	12.8	11.5	9.4
	%-reduction zu 83-87	0.0	-63.4	-67.2	-73.3
Ems	Emissions in kt/a	2.4	1.3	1.1	0.9
	%-reduction zu 83-87	0.0	-47.0	-53.9	-60.6
Weser	Emissions in kt/a	10.0	4.2	3.6	3.0
	%-reduction zu 83-87	0.0	-58.0	-64.1	-69.7
Elbe	Emissions in kt/a	17.7	6.4	4.4	3.8
	%-reduction zu 83-87	0.0	-64.1	-75.2	-78.4
Odra	Emissions in kt/a	1.6	0.5	0.4	0.4
	%-reduction zu 83-87	0.0	-67.6	-76.7	-78.6
North Sea	Emissions in kt/a	0.6	0.3	0.3	0.2
	%-reduction zu 83-87	0.0	-61.1	-57.6	-62.5
Baltic Sea	Emissions in kt/a	2.0	0.7	0.6	0.5
	%-reduction zu 83-87	0.0	-67.6	-71.9	-74.4
Germany	Emissions in kt/a	79.5	30.8	26.5	22.2
	%-reduction zu 83-87	0.0	-61.2	-66.6	-72.1

Table 49: Nitrogen emissions into the German river basins for the long-term average (LT), long-term maximum (WY) and long-term minimum (DY) and their changes towards the long-term average (LT)

Nitrogen		LT	WY	DY
Danube	Emissions in kt/a	126.4	177.1	87.6
	%-reduction zu LT	0	40.1	-30.7
Rhine	Emissions in kt/a	225.5	305.7	161.5
	%-reduction zu LT	0.0	35.5	-28.4
Ems	Emissions in kt/a	28.2	38.7	21.2
	%-reduction zu LT	0.0	37.5	-24.5
Weser	Emissions in kt/a	74.7	105.2	54.5
	%-reduction zu LT	0.0	40.9	-27.1
Elbe	Emissions in kt/a	108.7	145.2	88.8
	%-reduction zu LT	0.0	33.6	-18.3
Odra	Emissions in kt/a	13.0	20.6	10.6
	%-reduction zu LT	0.0	58.9	-18.0
North Sea	Emissions in kt/a	16.9	18.7	13.6
	%-reduction zu LT	0.0	10.8	-19.2
Baltic Sea	Emissions in kt/a	19.6	23.8	14.4
	%-reduction zu LT	0.0	21.3	-26.8
Germany	Emissions in kt/a	612.9	835.0	452.3
	%-reduction zu LT	0.0	36.2	-26.2

Table 50: Phosphorus emissions into the German river basins for the long-term average (LT), long-term maximum (WY) and long-term minimum (DY) and their changes towards the long-term average (LT)

Phosphorus		LT	WY	DY
Danube	Emissions in kt/a	4.1	5.1	3.5
	%-reduction zu LT	0.0	24.0	-15.9
Rhine	Emissions in kt/a	9.8	11.3	8.4
	%-reduction zu LT	0.0	16.3	-13.8
Ems	Emissions in kt/a	1.0	1.4	0.7
	%-reduction zu LT	0.0	39.1	-27.2
Weser	Emissions in kt/a	3.1	4.1	2.6
	%-reduction zu LT	0.0	30.3	-18.5
Elbe	Emissions in kt/a	3.9	4.9	3.4
	%-reduction zu LT	0.0	23.7	-12.9
Odra	Emissions in kt/a	0.4	0.7	0.3
	%-reduction zu LT	0.0	94.9	-23.7
North Sea	Emissions in kt/a	0.2	0.3	0.1
	%-reduction zu LT	0.0	24.1	-41.7
Baltic Sea	Emissions in kt/a	0.6	0.7	0.4
	%-reduction zu LT	0.0	29.5	-28.9
Germany	Emissions in kt/a	23.1	28.5	19.4
	%- reduction zu LT	0.0	23.4	-16.0



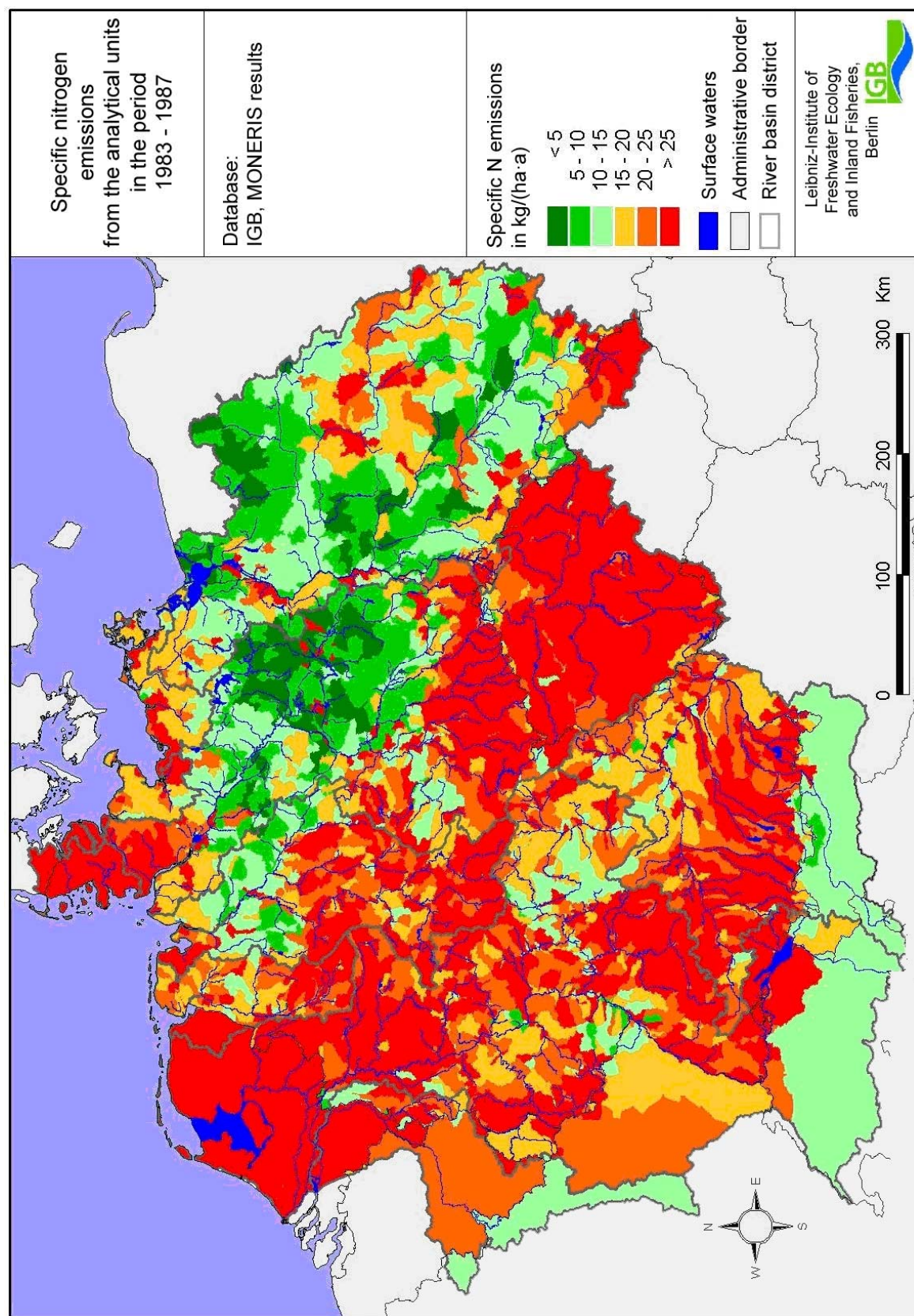


Figure 46: Specific nitrogen emissions in the period 1983-1987



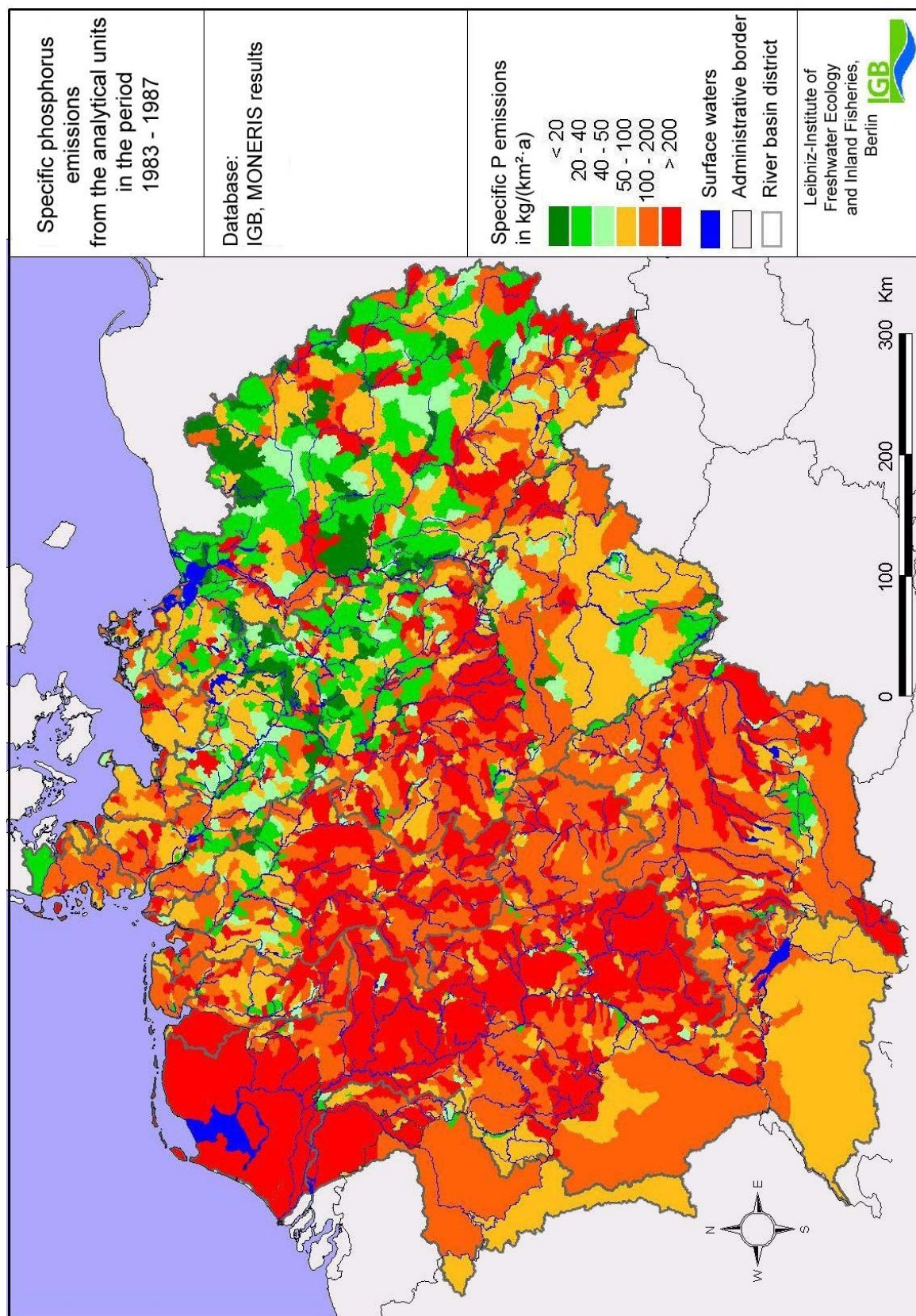


Figure 47: Specific phosphorus emissions in the period 1983-1987



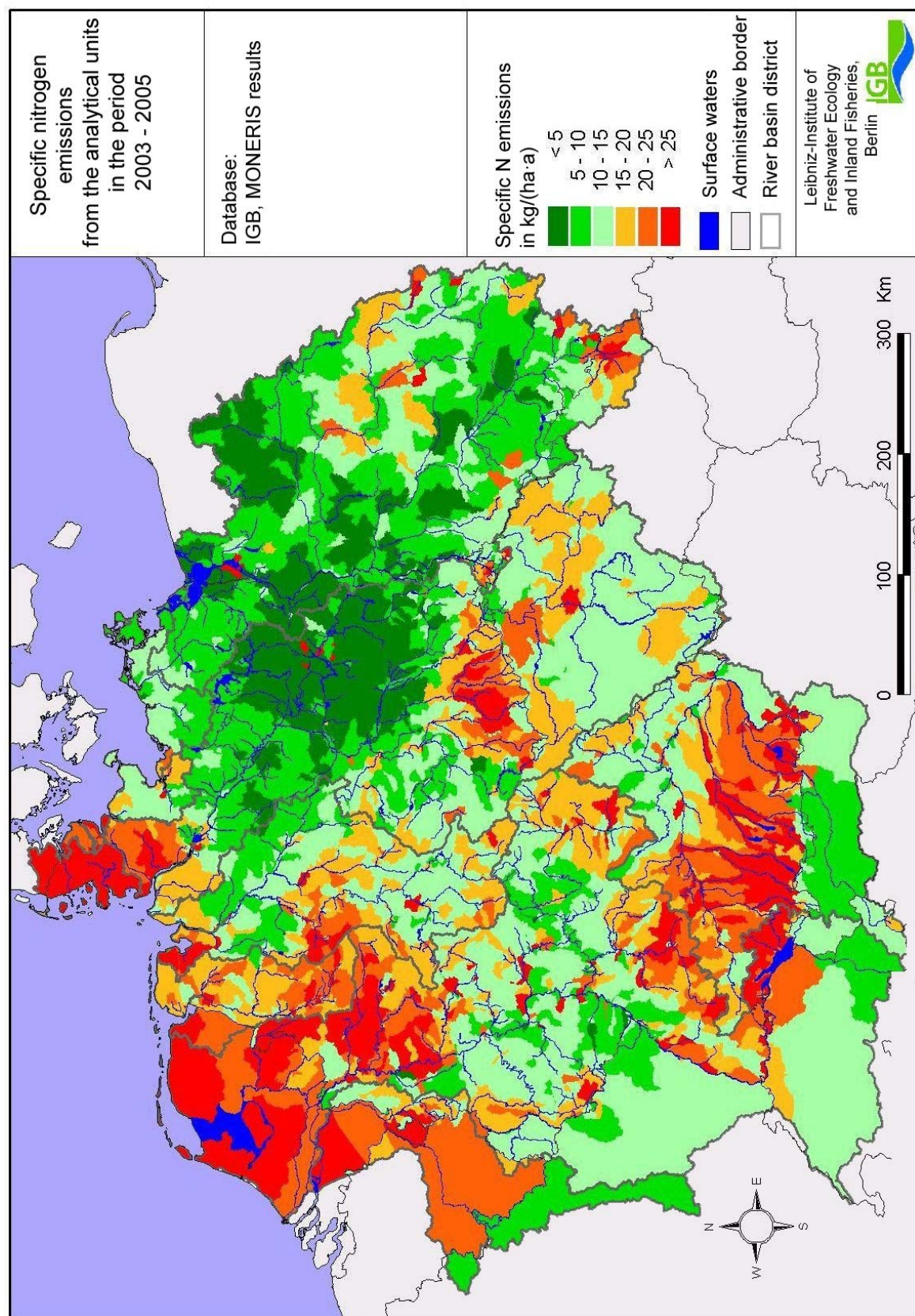


Figure 48: Specific nitrogen emissions in the period 2003-2005



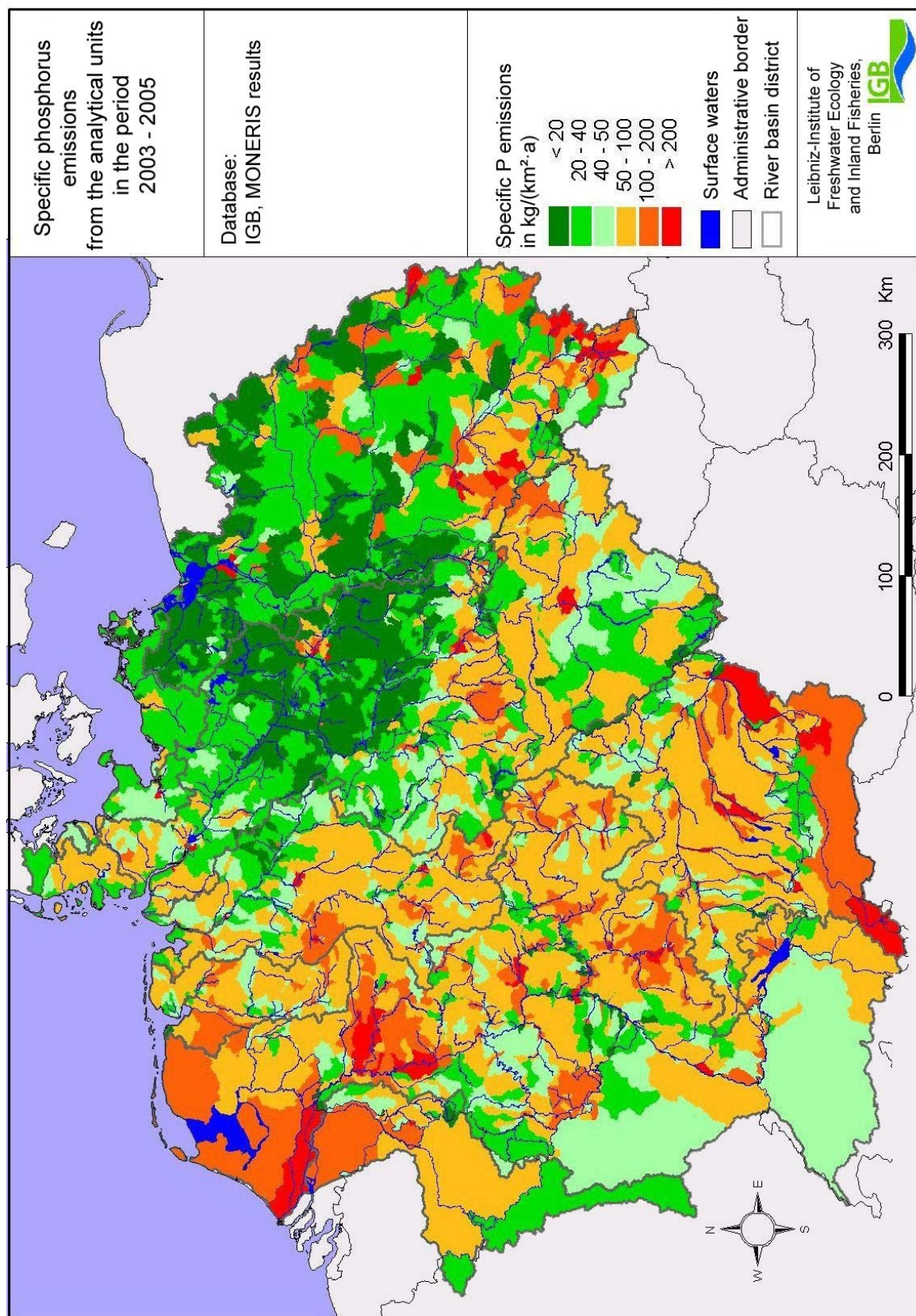


Figure 49: Specific phosphorus emissions in the period 2003-2005

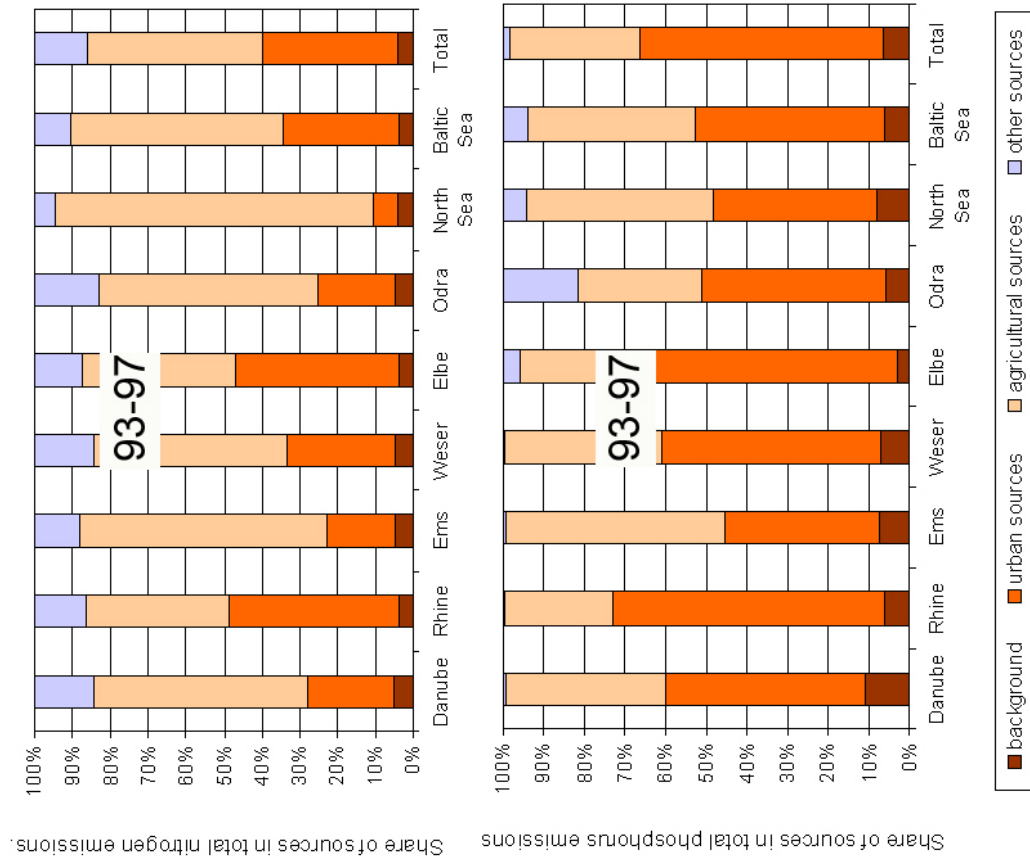


Figure 50: Share of sources in total nutrient emissions in the German river basins in the periods 1983 -1987 and 1993-1997

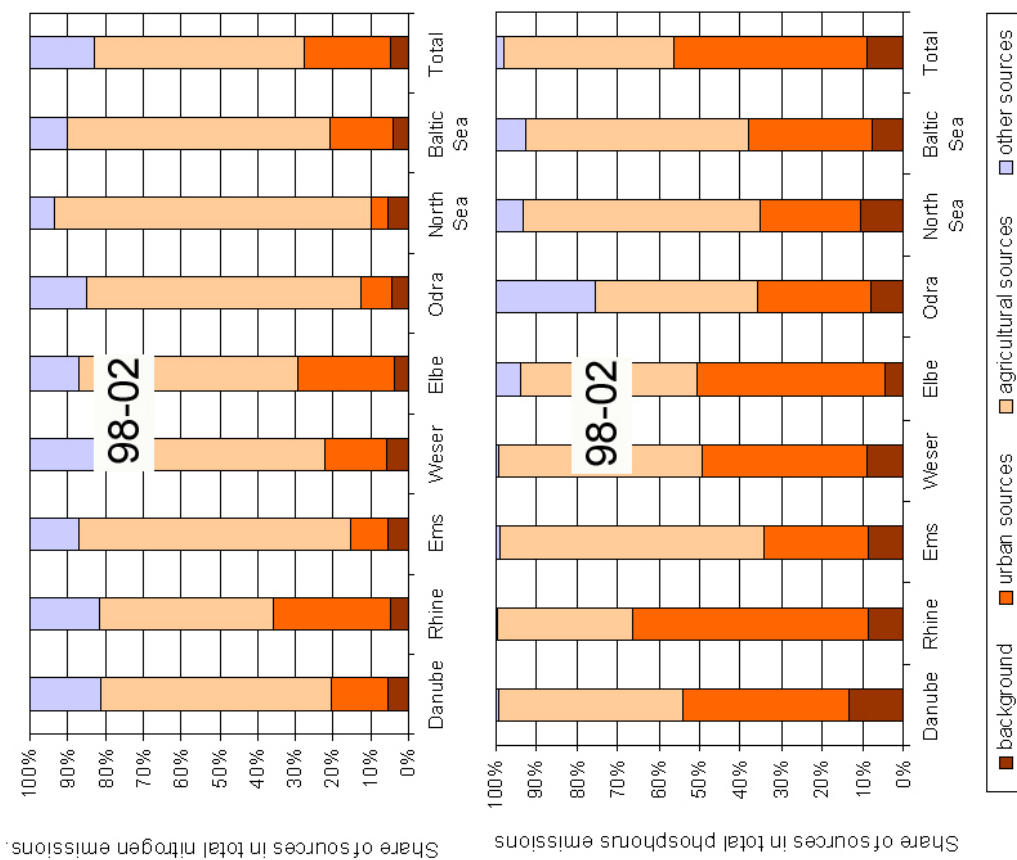
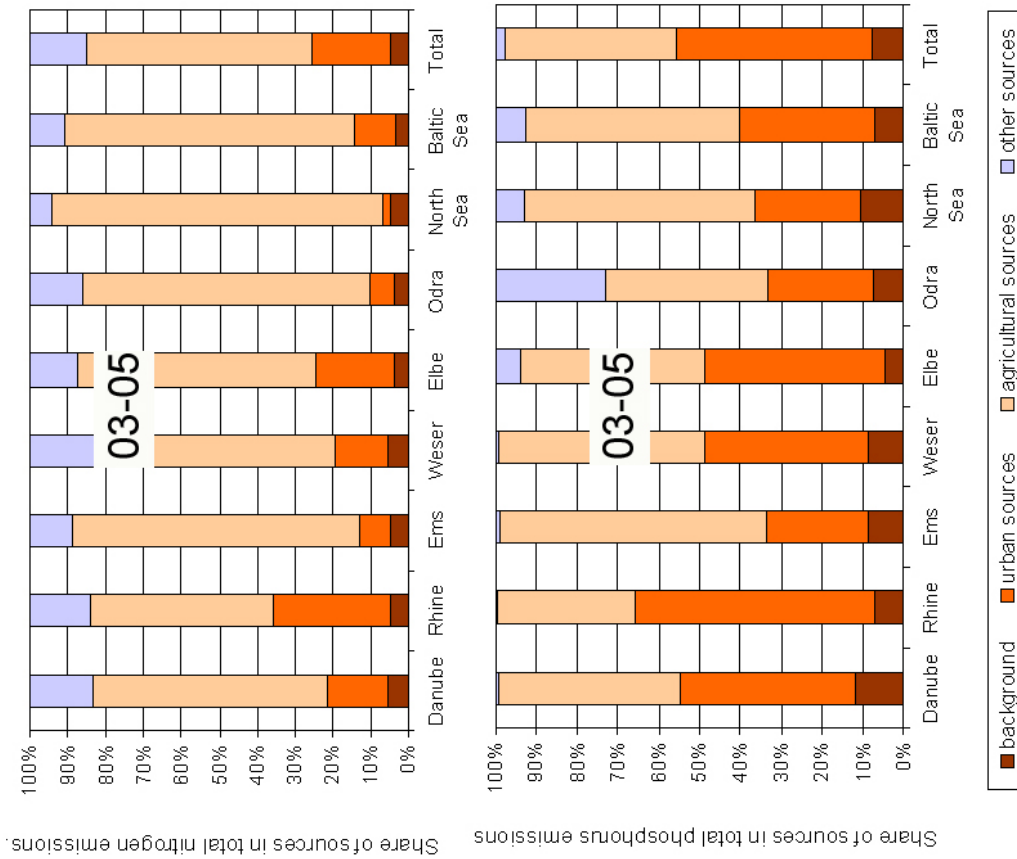


Figure 51: Share of sources in total nutrient emissions in the German river basins in the periods 1998-2002 und 2003-2005

### **5.2.2 Comparison of the modelled nitrogen emissions considering the atmospheric deposition according to EMEP and GAUGER**

Apart from the nitrogen deposition maps according to EMEP the nitrogen deposition maps according to GAUGER ET AL. (2007) have been available since 2007, too. The EMEP maps are available for the period of 1989 – 2004 as average annual values in a grid size of 50 x 50 km. GAUGER ET AL. (2007) calculated the average annual nitrogen deposition for a grid size of 1 x 1 km but only for chosen years (chapter 3.3). Except for the higher spatial resolution the other difference between both approaches is that GAUGER ET AL. (2007) defined specific deposition rates for land use while the EMEP maps consider average deposition rates.

The use of the deposition according to GAUGER ET AL. (2007) was not suitable for this project as there was no data available on the analytical units outside Germany and for most of the calculation years. Merging both data sets would have led to inconsistencies within the data. Their influence on the model calibration would have been hard to differentiate and interpret. The differences of both deposition maps and their influence on the N surpluses (BACH & SKITSCHAK, 2007) as well as on the calculated nitrogen loads with MONERIS in the German analytical units exemplary for the year 1999 are discussed in the following.

Overlaying both maps shows that the data from GAUGER from the year 1999 reveals in average 10 % higher deposition rates for Germany than the EMEP values. The regional differences are also considerably higher. While the GAUGER data shows more than 1.5 times the EMEP values for the north west of Germany, it is the opposite for the east (especially in the new states of Germany). Similarly high differences in the data sources can be found for the south of Germany, too. GAUGER ET AL. (2007) predominantly calculated significantly higher depositions rates for the Danube catchment area (Figure 52). In the comparison of nitrogen surpluses on agricultural areas

BACH & SKITSCHAK (2007) found significantly lower values when using GAUGER maps which at first seems to contradict the results of the total deposition. While the EMEP maps can hardly reproduce land use specific differences in the deposition rates due to their grid size, there are significant differences according to GAUGER ET AL. (2007). So the deposition on agricultural areas according to GAUGER ET AL (2007) is about 23 % lower than the average deposition on all areas. The comparison of the GAUGER depositions on agricultural areas with the total deposition according to EMEP shows a similar pattern as for the total deposition (Figure 53).

In the west of Germany the deposition on agricultural areas according to GAUGER ET AL. (2007) is higher (> 25 %) than the total deposition according to EMEP area-wide, while the deposition in the east of Germany is lower (Figure 53).

Due to a lower deposition on agricultural areas BACH & SKITSCHAK (2007) calculated significantly lower nitrogen surpluses. Analogue to the spatially differentiated differences between the deposition according to GAUGER ET AL. (2007) and to EMEP there are significant differences in the calculated N surpluses. The GAUGER values in the west of Germany showed surpluses that were up to 10 % higher, while in most of the other analytical units 10 – 50 % lower surpluses were calculated compared to EMEP (Figure 54).



Using the GAUGER data reveals a 10 % higher nitrogen deposition nation-wide but at the same time a reduction of the N surpluses from 85 kg/(ha·a) (EMEP) down to 79 kg/(ha·a) (GAUGER ET AL., 2007). More important for the calculated emissions is the different spatial distribution of the deposition values and the N surpluses using both data sources. According to this distribution the emissions are 25 % higher in the west while in the east and south of Germany emissions were reduced by up to 25 % (Figure 55). In total both effects weaken each other (increase of the total deposition and reduction of the N surpluses) so that in average only 1 % lower nitrogen loads are calculated (Table 51). The effect widely differs from river basin to river basin. River basins in the west and north of Germany (Ems, Weser as well as direct catchment areas to the North Sea and Baltic Sea) show a significant rise in emissions from atmospheric deposition. In the other areas emissions calculated from this pathway only show a small change or even a slight reduction. The lower N surpluses (GAUGER data) cause a reduction of the nitrogen loads via drainage (< 11 %) and via groundwater (< 7 %) (Table 51). In total the changes of nitrogen emissions into river basins are around  $\pm 5$  %.

In this project it cannot be checked whether the spatial changes of emissions lead to a better conformance of the observed and calculated river loads. It still has to be examined to what extent the results differ for other years for which GAUGER-data is available. Furthermore it would be sensible to test to what extent the calculated emissions are affected if the atmospheric deposition is considered completely differentiated by the use.

Table 51: The change of TN emissions into the river basins considering the deposition data according to GAUGER ET AL. (2007) for the year 1999

	<b>AD [%]</b>	<b>SR [%]</b>	<b>DR [%]</b>	<b>ER [%]</b>	<b>GW [%]</b>	<b>PS [%]</b>	<b>US [%]</b>	<b>Total [%]</b>
Danube	1.4	1.5	-10.9	0.0	-6.5	0.1	1.7	-5.2
Rhine	0.1	11.9	-7.2	0.0	-3.2	0.1	4.1	-1.3
Ems	68.2	66.1	1.4	0.0	2.7	3.3	28.6	8.0
Weser	36.4	31.7	-4.1	0.0	-0.8	1.0	8.4	2.0
Elbe	-9.2	0.8	-3.5	0.0	-3.4	-0.3	-1.4	-2.6
Odra	-4.7	13.2	-5.4	0.0	-4.1	0.9	2.7	-3.0
North Sea	50.3	49.6	0.1	0.0	1.5	1.2	16.2	3.6
Baltic Sea	42.4	38.0	2.5	0.0	3.4	1.0	16.2	6.1
Total	3.4	14.5	-4.1	0.0	-3.3	0.2	2.9	-1.2

(AD = atmospheric deposition, SR = surface runoff, DR = drainage; ER = erosion, GW = groundwater, PS = point sources, US = urban systems)

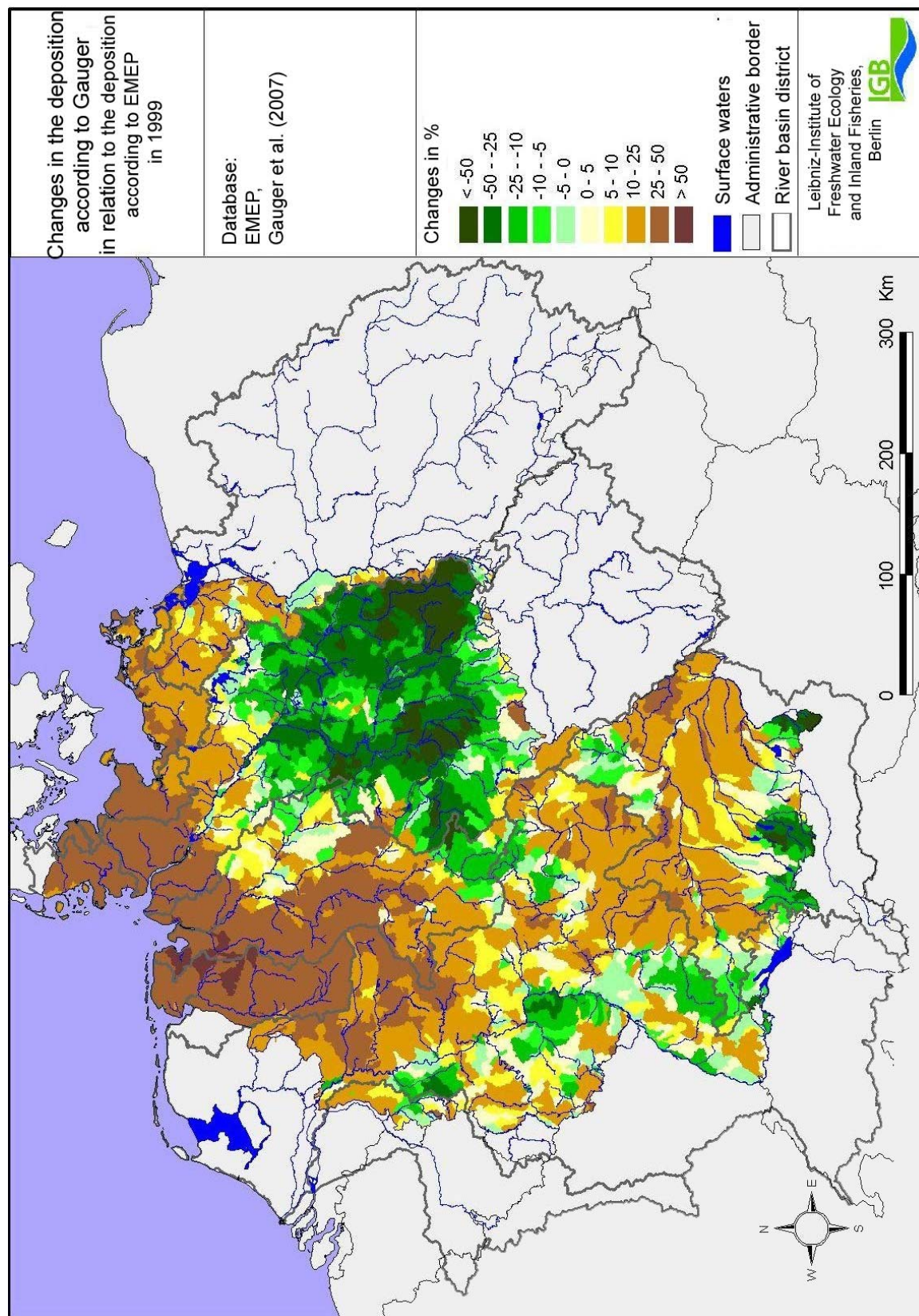


Figure 52: Comparison of the total deposition according to GAUGER ET AL. (2007) with EMEP-data for the year 1999



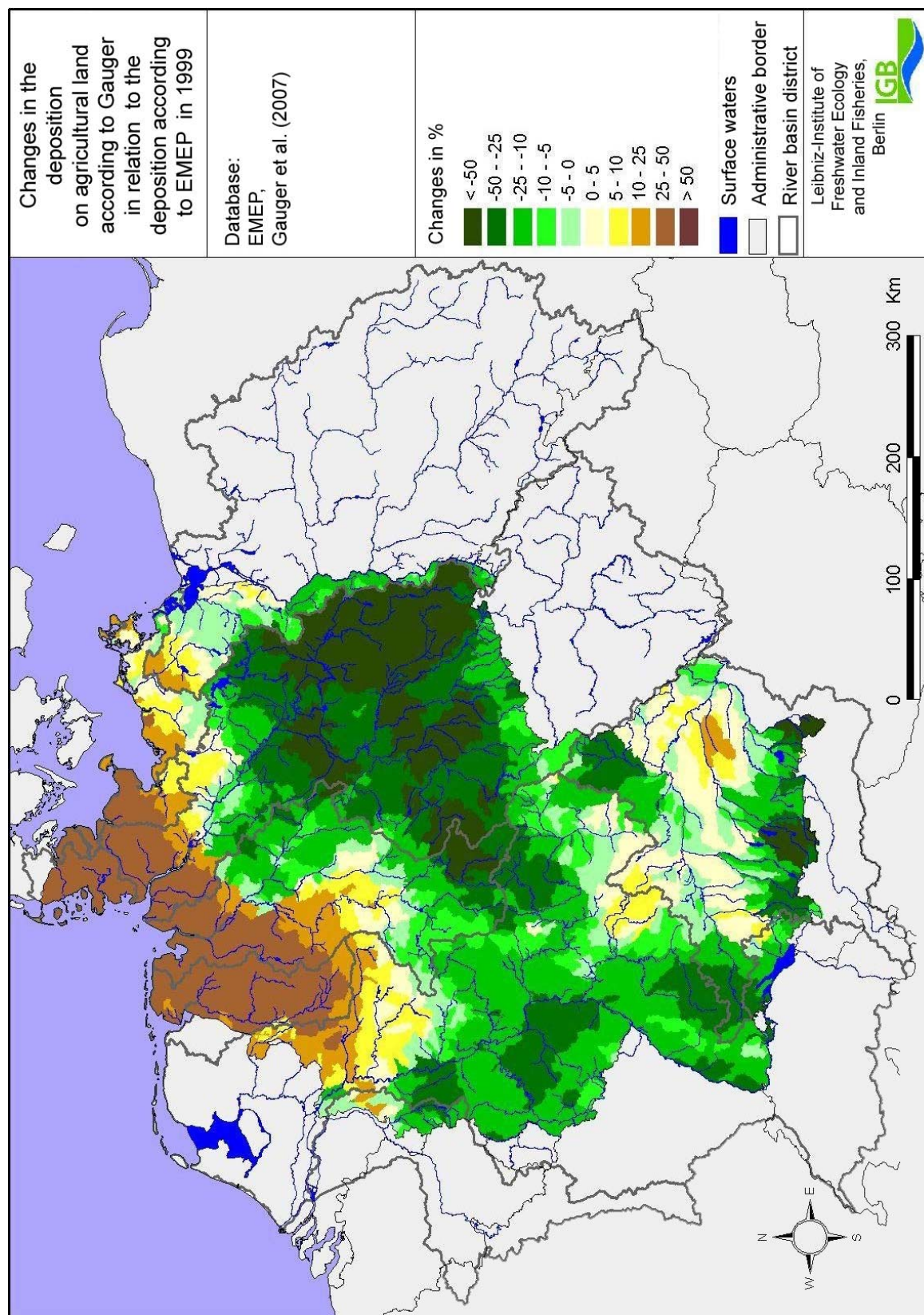


Figure 53: Comparison of the total deposition according to EMEP with the deposition onto agricultural areas according to GAUGER ET AL. (2007) for the year 1999

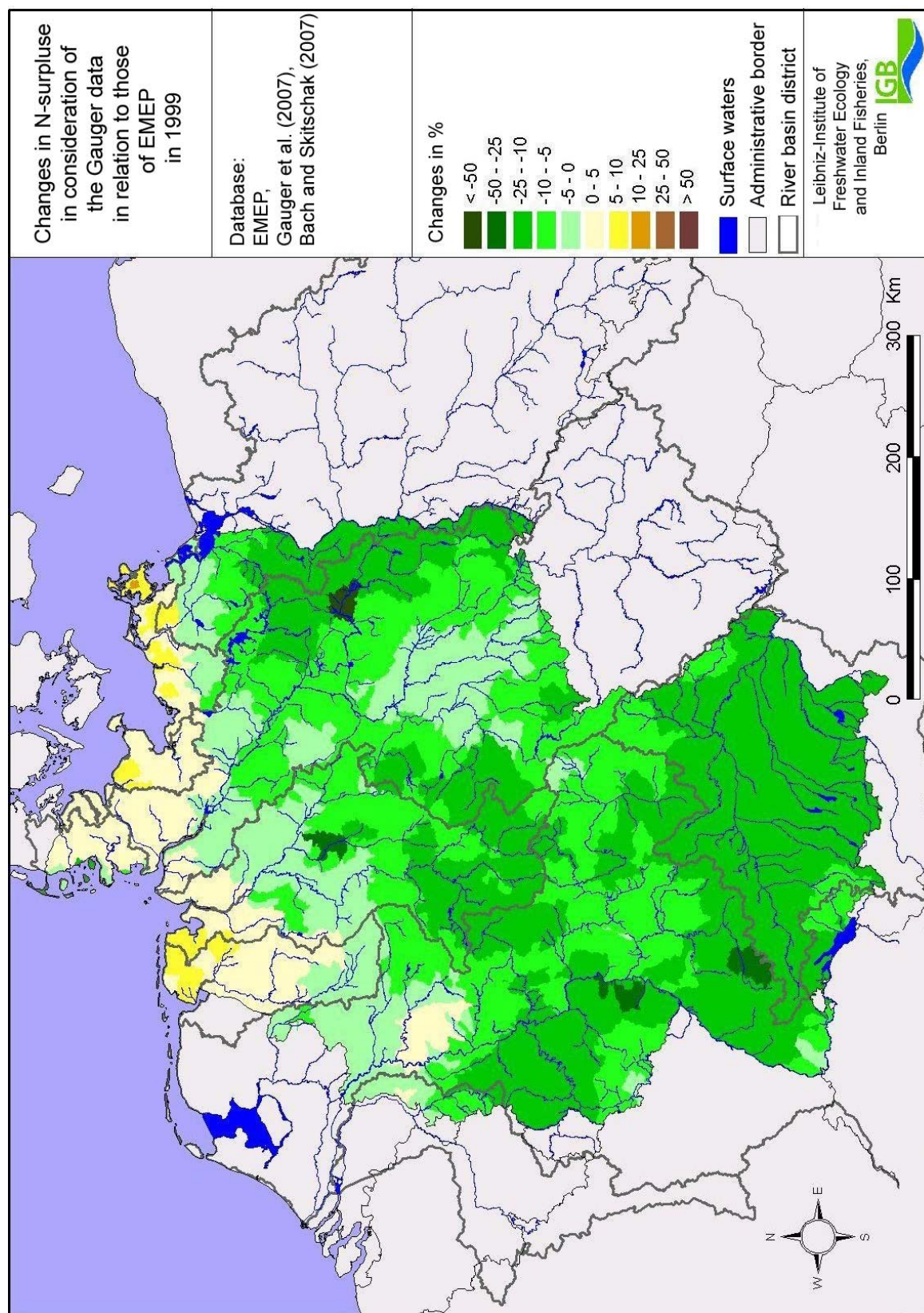


Figure 54: Comparison of the N-surplus calculated by BACH & SKITSCHAK (2007) for the year 1999 considering the deposition according to EMEP and GAUGER ET AL. (2007)



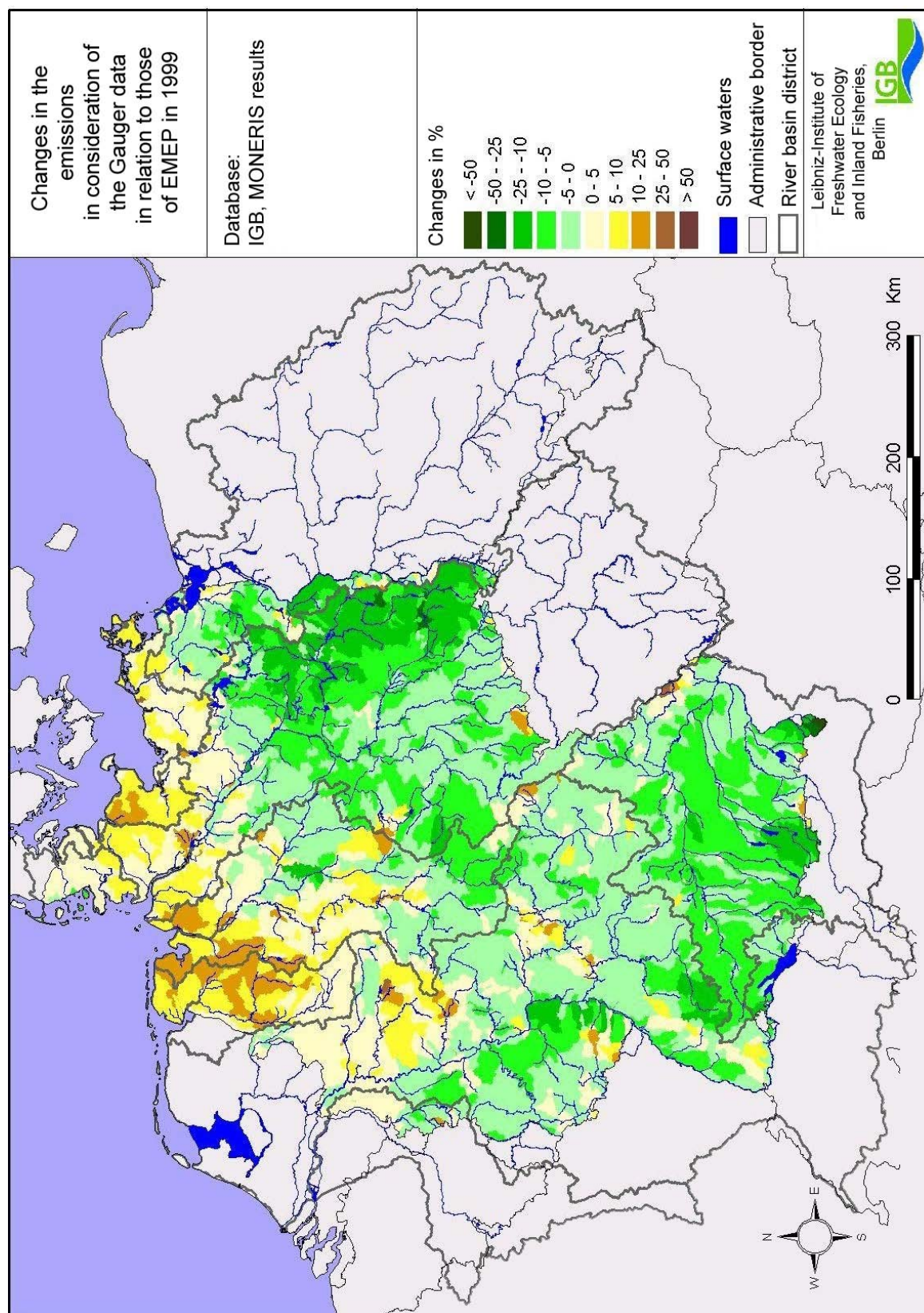


Figure 55: Comparison of the TN emissions for the year 1999 considering the N-surplus and deposition according to EMEP and GAUGER ET AL. (2007)

### 5.2.3 Comparison of the observed and modelled nutrient river loads

The comparison of the modelled and observed river loads is based on the average river loads of the individual years of a period calculated with MONERIS and the averages of the observed river loads. The time series of the concentration measurements at quality monitoring stations are partly incomplete or show discrepancies so that not all of the available data could be taken to calculate an average annual river load. To be able to compare modelled and observed river loads it was made certain that there was data on observed loads for at least three years of one period. Otherwise they were not considered for the river load comparison. The number of considered stations clearly varies between the periods but also between the observed substance fractions (total nitrogen, dissolved inorganic nitrogen, total phosphorus). Especially for the 80s there were only a few values. Figure 56 depicts the location of the considered stations.

The average deviations over the whole period between the observed and modelled river loads are 30 % for TN, 28 % for DIN and 38 % for TP and also consistently show a good statistical compliance (Table 52). In the individual periods the deviation is a little higher with a similarly good statistical compliance. In total the deviation for all substance fractions is higher in the first periods (83-87, 93-97) than in the later periods and is also above the average deviation for all years.

Table 52: Comparison of the observed and modelled TN, DIN and TP river loads for the different periods

Period	Substance/fraction	Average deviation [%]	Coefficient of determination	EF-value	Number of measurement points
1983-1987	TN	46.2	0.99	0.79	6
	DIN	34.7	0.97	0.90	125
	TP	40.6	0.91	0.91	82
1993-1997	TN	30.7	0.95	0.95	109
	DIN	27.5	0.98	0.97	213
	TP	42.2	0.80	0.76	199
1998-2002	TN	26.5	0.96	0.94	232
	DIN	28.2	0.96	0.95	366
	TP	34.0	0.94	0.92	289
2003-2005	TN	28.7	0.97	0.95	251
	DIN	31.0	0.96	0.96	304
	TP	35.6	0.86	0.77	229
Gesamt	TN	30.0	0.95	0.92	332
	DIN	28.0	0.97	0.94	413
	TP	38.4	0.89	0.89	339

As it can be seen in Figure 57, Figure 58, Figure 59 and Figure 60 the modelled river loads in all periods are scattered equally around the 1:1-line. This indicates that the modelled emissions and river loads have no systematic errors so that the cause of error can be lead back to the used input data, too. This can be explained by the fact that when the time series of

input data was derived, e.g. for wastewater treatment plants, the current condition was used as a basis to derive the earlier conditions (chapter 2). It can be assumed that the input data used from the earlier years is less precise and therefore the results have a higher error rate than the results of the later years.

In addition the comparison of the modelled and observed river loads show increasing deviations for smaller waterbodies. Like for the area runoff calibration this is lead back to the larger dynamic of the discharge and concentrations in smaller waterbodies in comparison to large rivers. In total the demand for a higher sample density and with that more samples for smaller waterbodies can be deduced. Furthermore, the input data in smaller catchment areas is more uncertain as regional differences sometimes cannot be covered in a model that represents the whole of Germany.



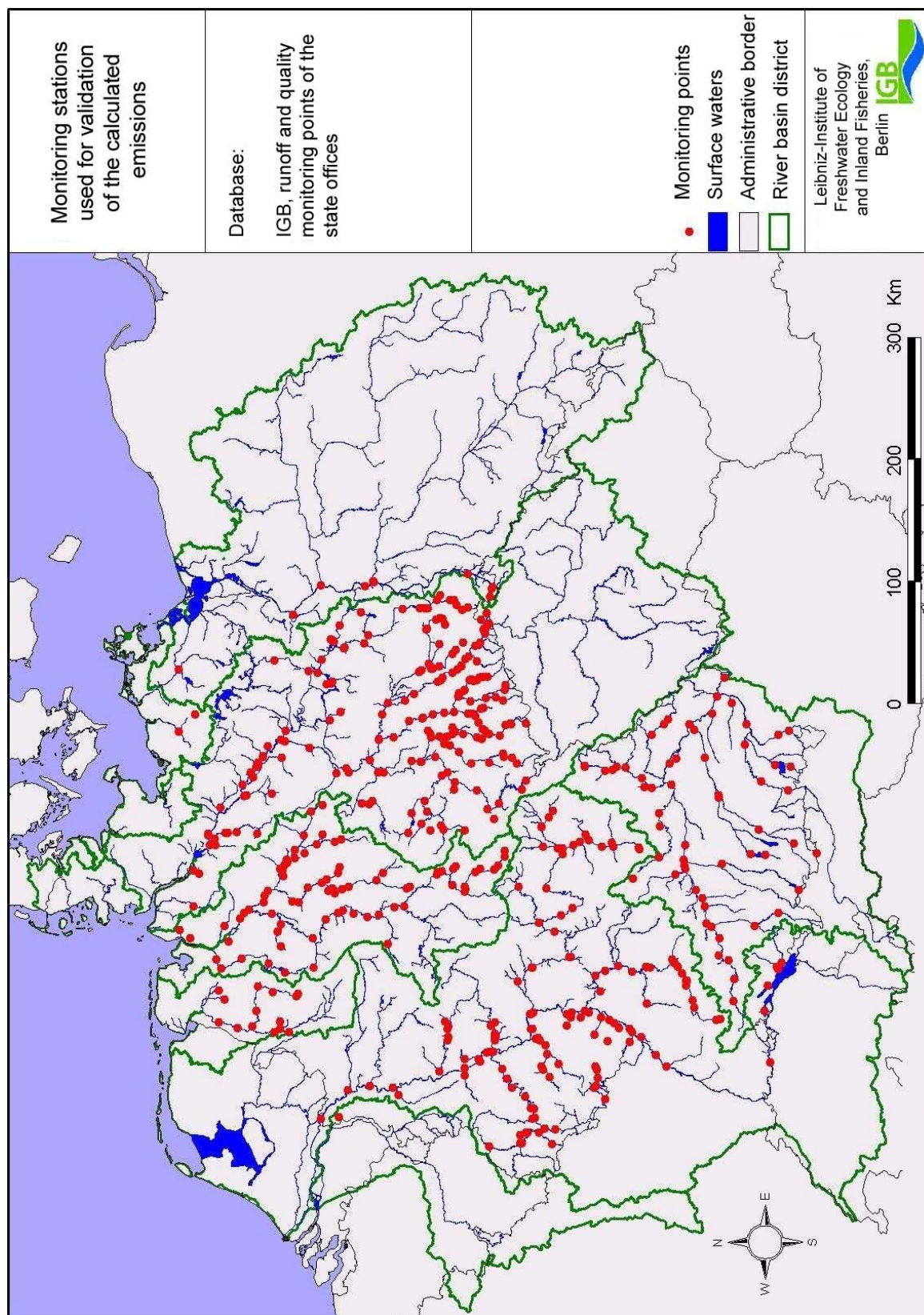


Figure 56: Quality monitoring stations used for validation of modelled river loads

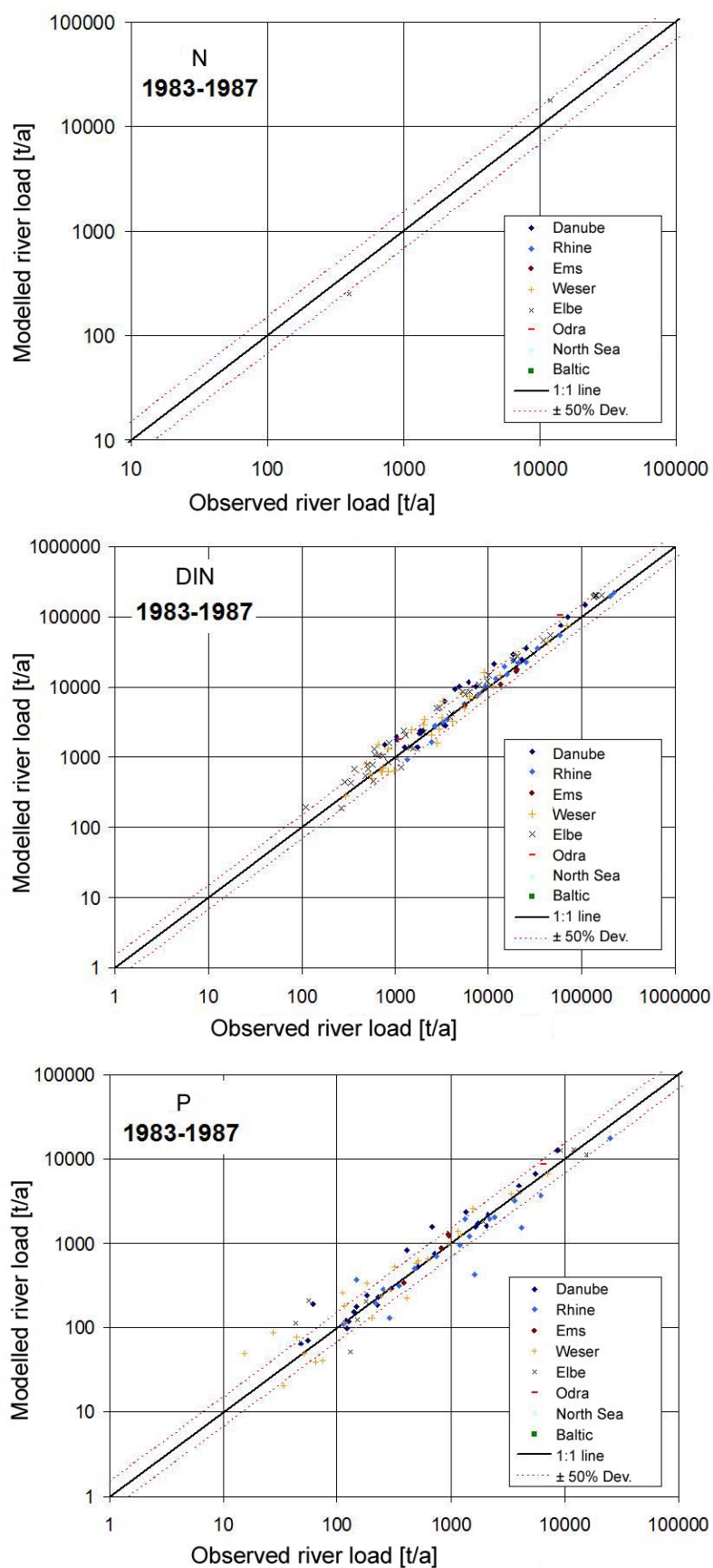


Figure 57: Comparison of modelled and observed river loads for the period 1983-1987 for TN, DIN, TP

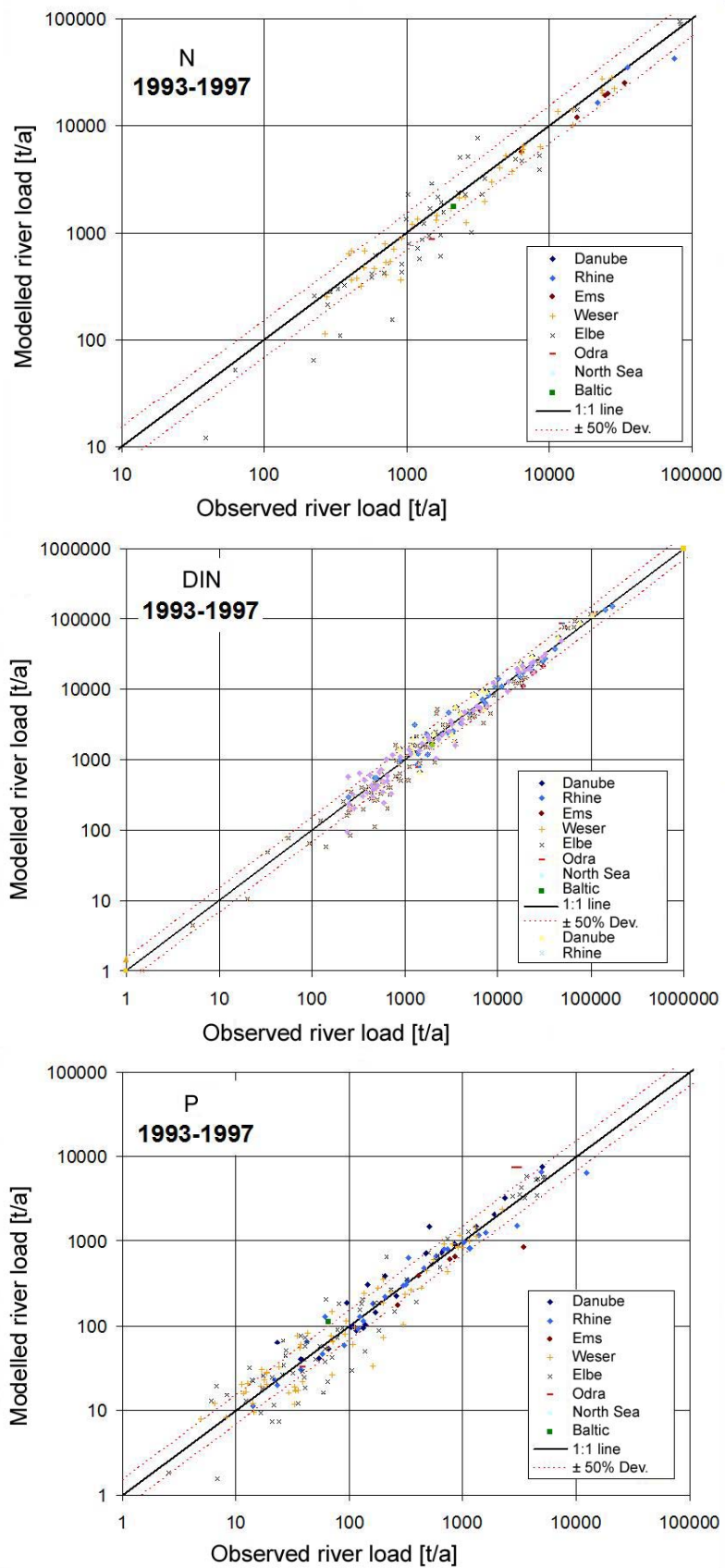


Figure 58: Comparison of modelled and observed river loads for the period 1993-1997 for TN, DIN, TP

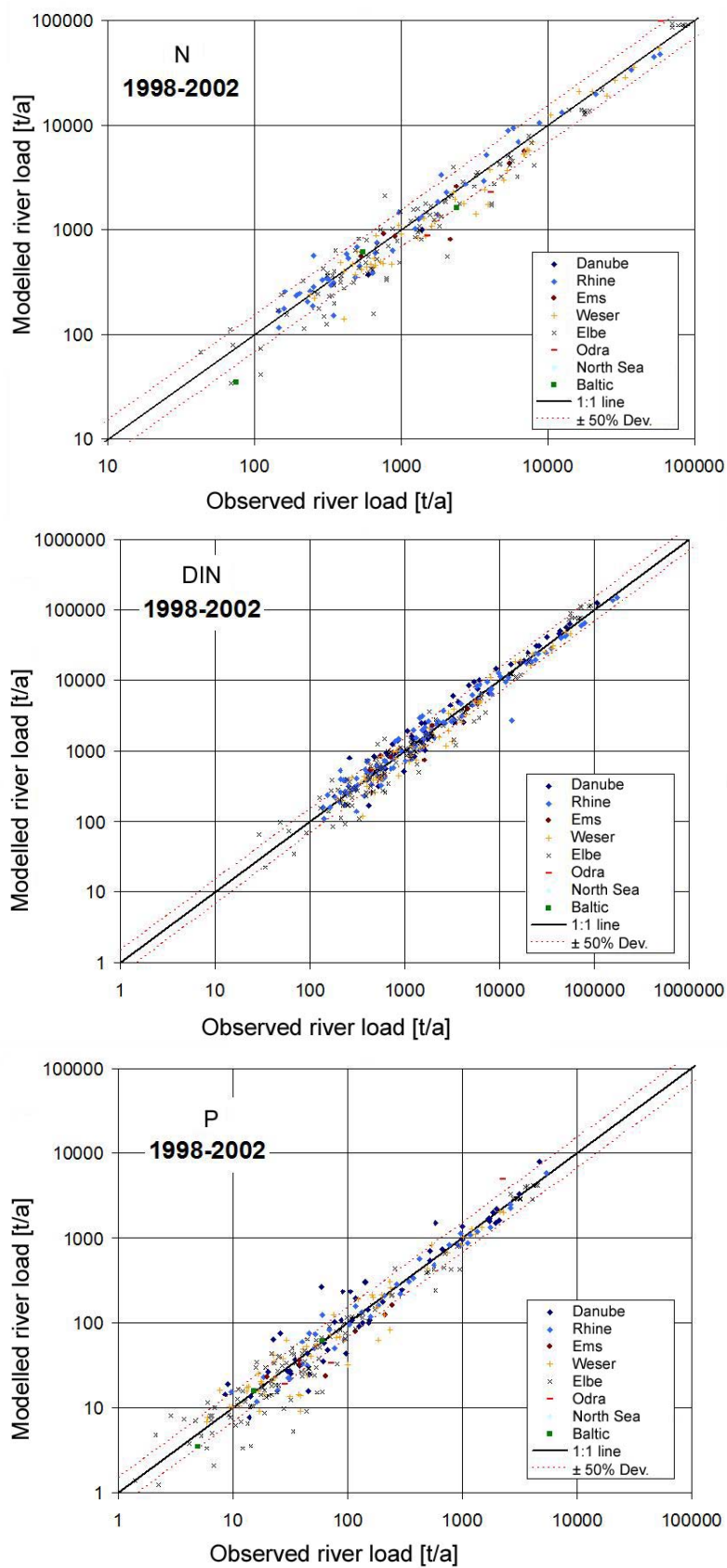


Figure 59: Comparison of modelled and observed river loads for the period 1998-2002 for TN, DIN, TP



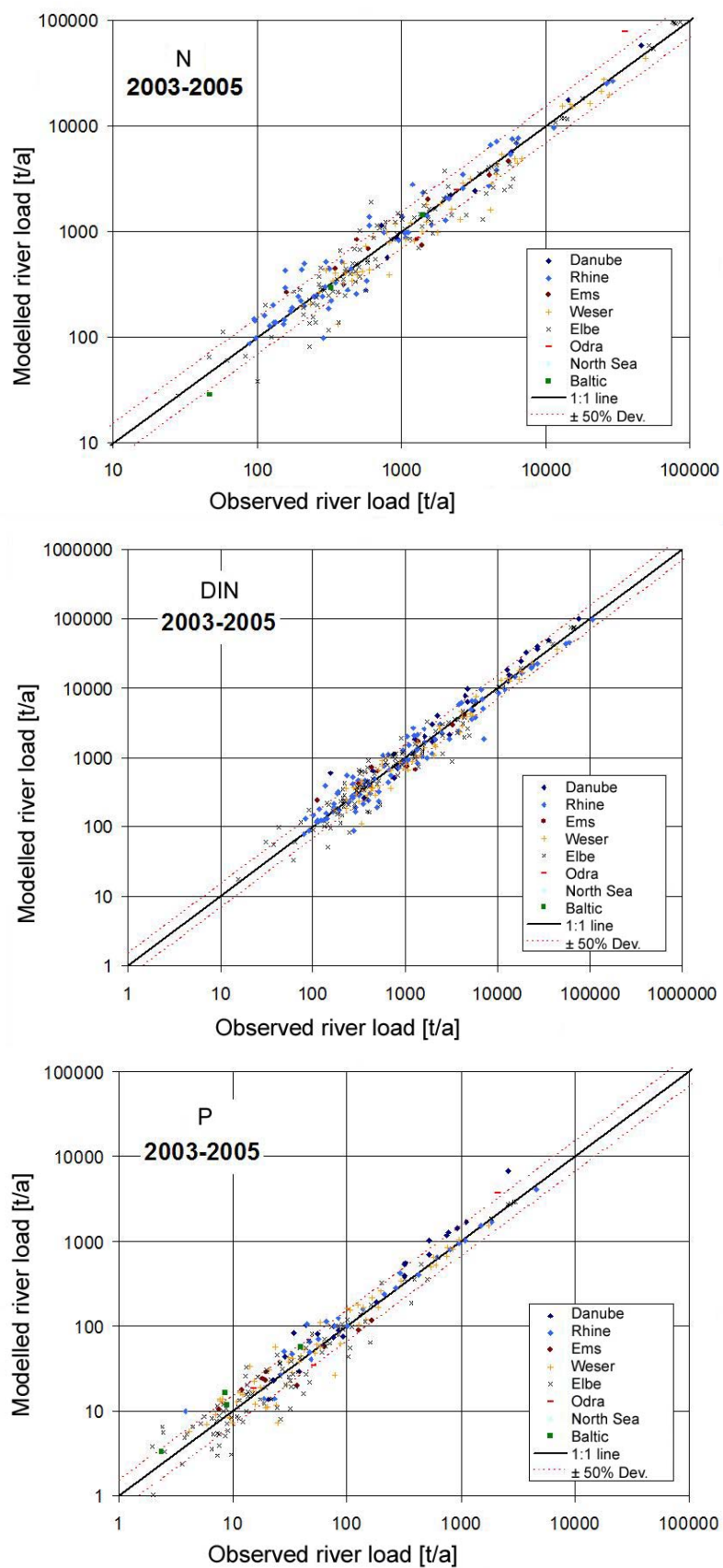


Figure 60: Comparison of modelled and observed river loads for the period 2003-2005 for TN, DIN, TP

#### **5.2.4 Cumulative retention in the river basins**

The cumulative retention describes the share of degraded and retained loads over a longer period from leaving a catchment area till flowing into a coastal zone. Loads with a longer flowing distance tend to have a higher retention rate than those with a shorter flowing distance. As Figure 61 and Figure 62 show the water surface distribution plays an essential role in the resulting cumulative retention. Five waterbodies are important for the supraregional retention: Lake Constance, the IJsselmeer, the Muggelsee, the Muritz and the Szczecin Lagoon.

Lake Constance retains a considerable share of loads and makes sure that the emissions and the resulting river loads from upstream of Lake Constance only have a small share in the total loads in the lower course of the Rhine. Being situated far back in the upper course of the Rhine, only a comparably small share of the total Rhine basin drains into Lake Constance so that Lake Constance only has a small influence on the total loads of the Rhine.

13 km downstream of Lobith/Bimmen about 11 % of the Rhine discharge is diverted through the IJssel canal. This then drains via the IJssel into the IJsselmeer and then into the North Sea. As the IJsselmeer was considered for the retention calculations the modelled loads for TN and TP were reduced more than 50%.

In the Havel and Spree the Havel lakes chain and the Muggelsee contribute to a higher retention. Upstream loads are calculated to have a retention of 40 % and more. This also means that the emissions from the city of Berlin have a reduced share in the river loads at the Elbe's estuary.

Especially the Muritz has an enormous influence on the retention and the mass balance of the surface waters connected to the Mecklenburg Lake District. A retention (TN and TP) of more than 50% was calculated for the Muritz.

As the complete Odra and all its tributaries drain into the Szczecin Lagoon it plays a central role for the retention of Odra loads before reaching the Baltic Sea.



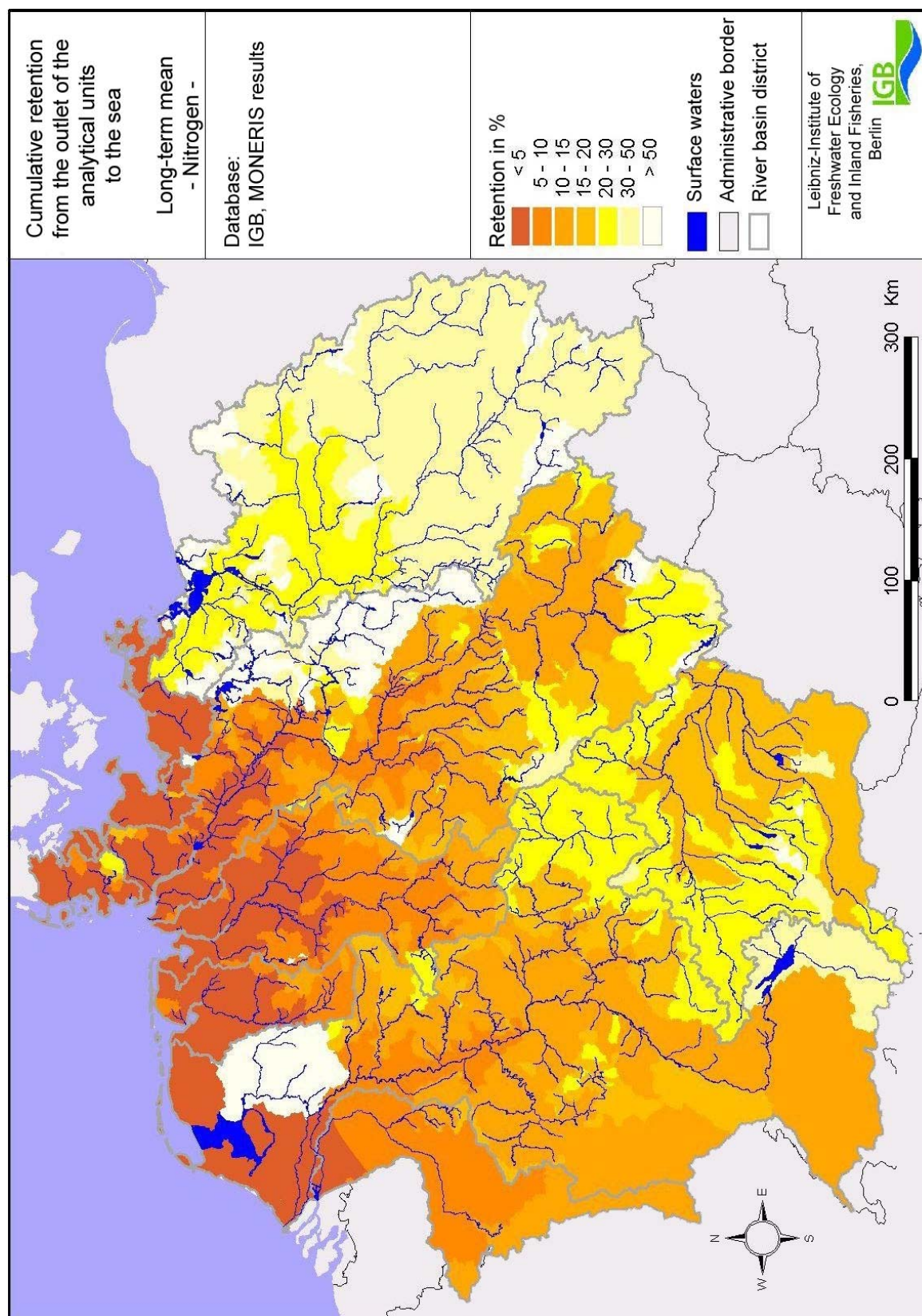


Figure 61: Average (long-term) cumulative TN-retention from the outlet of the analytical areas to the sea

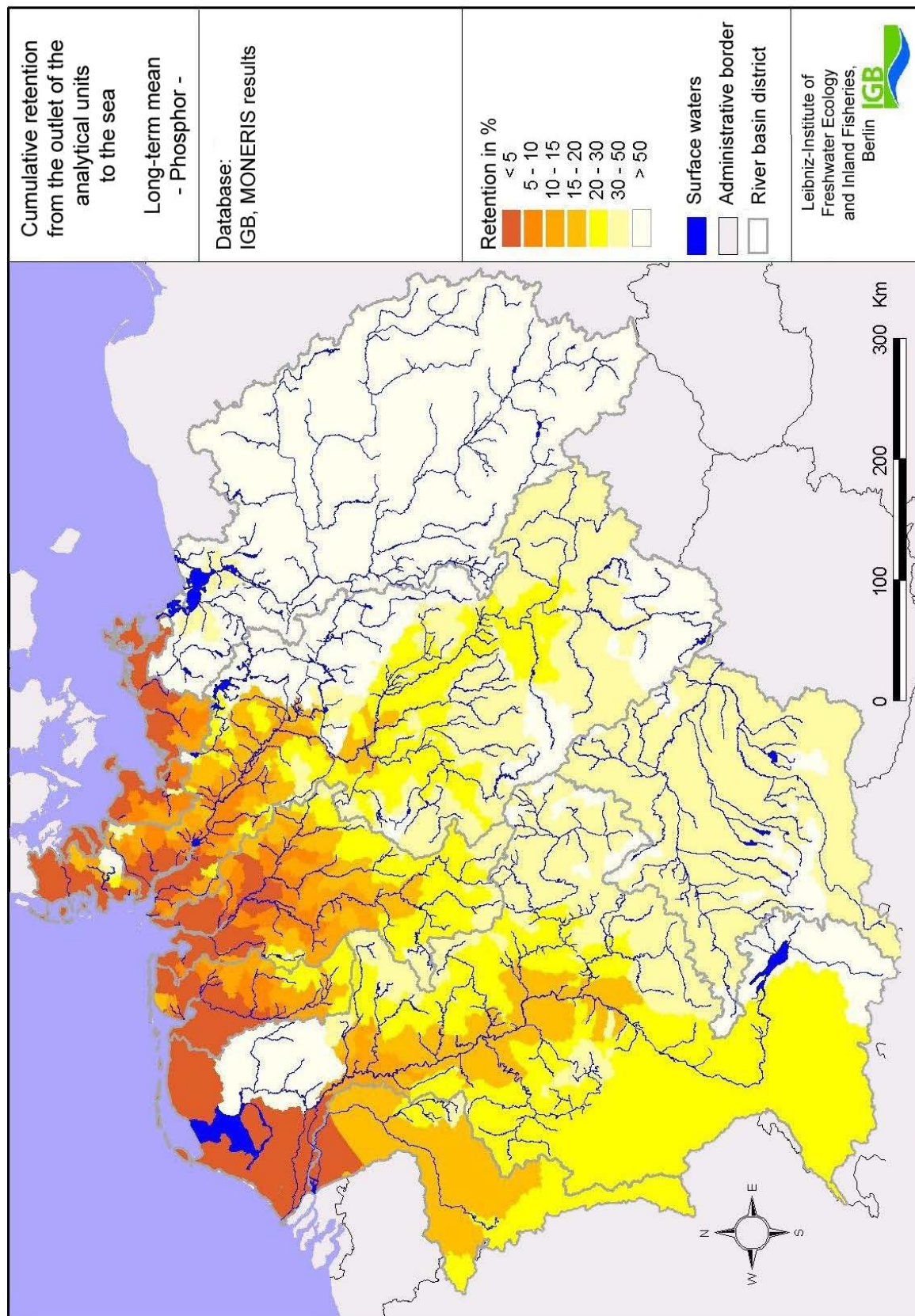


Figure 62: Average (long-term) cumulative TP-retention from the outlet of the analytical areas to the sea

## 5.3 Heavy metals

The heavy metal emissions were calculated for all the individual years (1983-2005) and subsequently accumulated to 5-year periods („1985“: 1983-1987, „1995“: 1993-1997, „2000“: 1998-2002 and „2005“: 2003-2005).

Table 53 depicts the total emissions into surface waters in Germany for the balance periods 1983-1987, 1993-1997 and 2003-2005, the share of point and diffuse pathways as well as the achieved reductions compared to the period 1985.

Table 53: Heavy metal emissions and reduction as well as share of point and diffuse pathways into the surface waters in Germany for 1983-1987, 1993-1997, 1998-2002 and 2003-2005

Metal	Emissions in t/a				Reduction to 1985		
	1983-1987	1993-1997	1998-2002	2003-2005	til 1995	til 2000	til 2005
<b>Point emissions</b>							
Cd	30.4	6.0	4.6	4.1	-80 %	-85 %	-86 %
Cr	576.9	90.9	52.9	42.9	-84 %	-91 %	-93 %
Cu	572.2	178.8	165.6	133.7	-69 %	-71 %	-77 %
Hg	24.6	2.3	1.5	1.1	-91 %	-94 %	-95 %
Ni	336.7	134.0	111.0	85.6	-60 %	-67 %	-75 %
Pb	197.5	67.4	53.0	41.9	-66 %	-73 %	-79 %
Zn	4,184.4	1,280.0	1,008.0	935.9	-69 %	-76 %	-78 %
<b>Diffuse emissions</b>							
Cd	34.5	8.8	6.8	5.1	-75 %	-80 %	-85 %
Cr	288.2	241.2	244.9	207.0	-16 %	-15 %	-28 %
Cu	483.9	409.1	418.7	327.5	-15 %	-13 %	-32 %
Hg	6.1	2.6	2.1	1.6	-57 %	-66 %	-74 %
Ni	564.3	463.7	498.3	391.2	-18 %	-12 %	-31 %
Pb	784.2	325.6	276.0	221.1	-58 %	-65 %	-72 %
Zn	3,008.9	2,248.3	2,230.4	1,819.6	-25 %	-26 %	-40 %
<b>Total emissions</b>							
Cd	64.9	14.7	11.4	9.2	-77 %	-82 %	-86 %
Cr	865.1	332.1	297.8	249.9	-62 %	-66 %	-71 %
Cu	1,056.1	587.9	584.3	461.2	-44 %	-45 %	-56 %
Hg	30.7	4.9	3.6	2.7	-84 %	-88 %	-91 %
Ni	900.9	597.6	609.3	476.8	-34 %	-32 %	-47 %
Pb	981.8	393.0	329.0	263.0	-60 %	-66 %	-73 %
Zn	7,193.3	3,528.3	3,238.5	2,755.4	-51 %	-55 %	-62 %

Figure 63 and Figure 64 depict the relative relevance of the individual pathways for the total heavy metal emissions into surface waters in Germany for the corresponding balance periods.



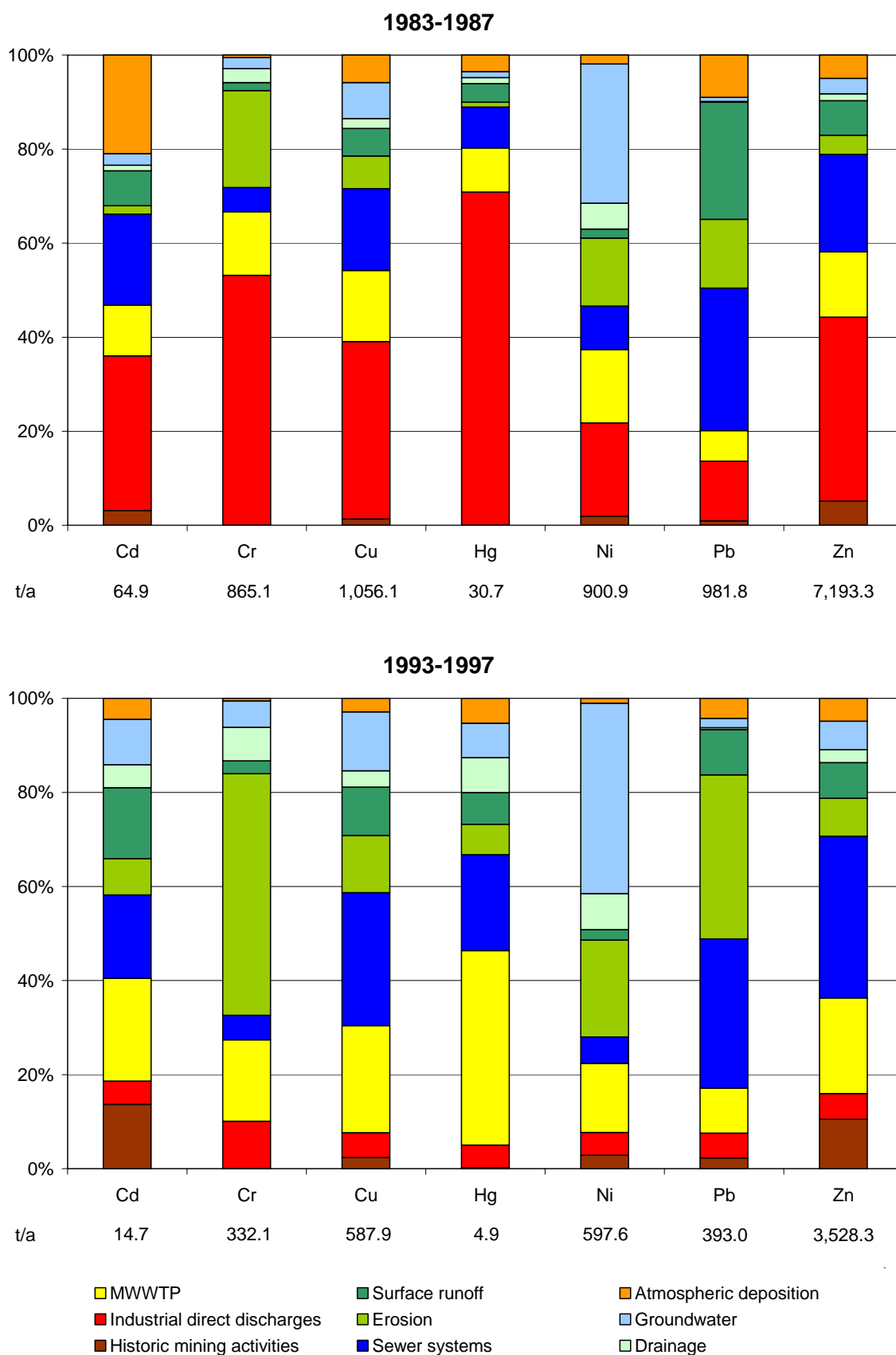


Figure 63: Relative relevance of the individual pathways for the total heavy metal emissions into surface waters in Germany for the balance periods 1983-1987 und 1993-1997

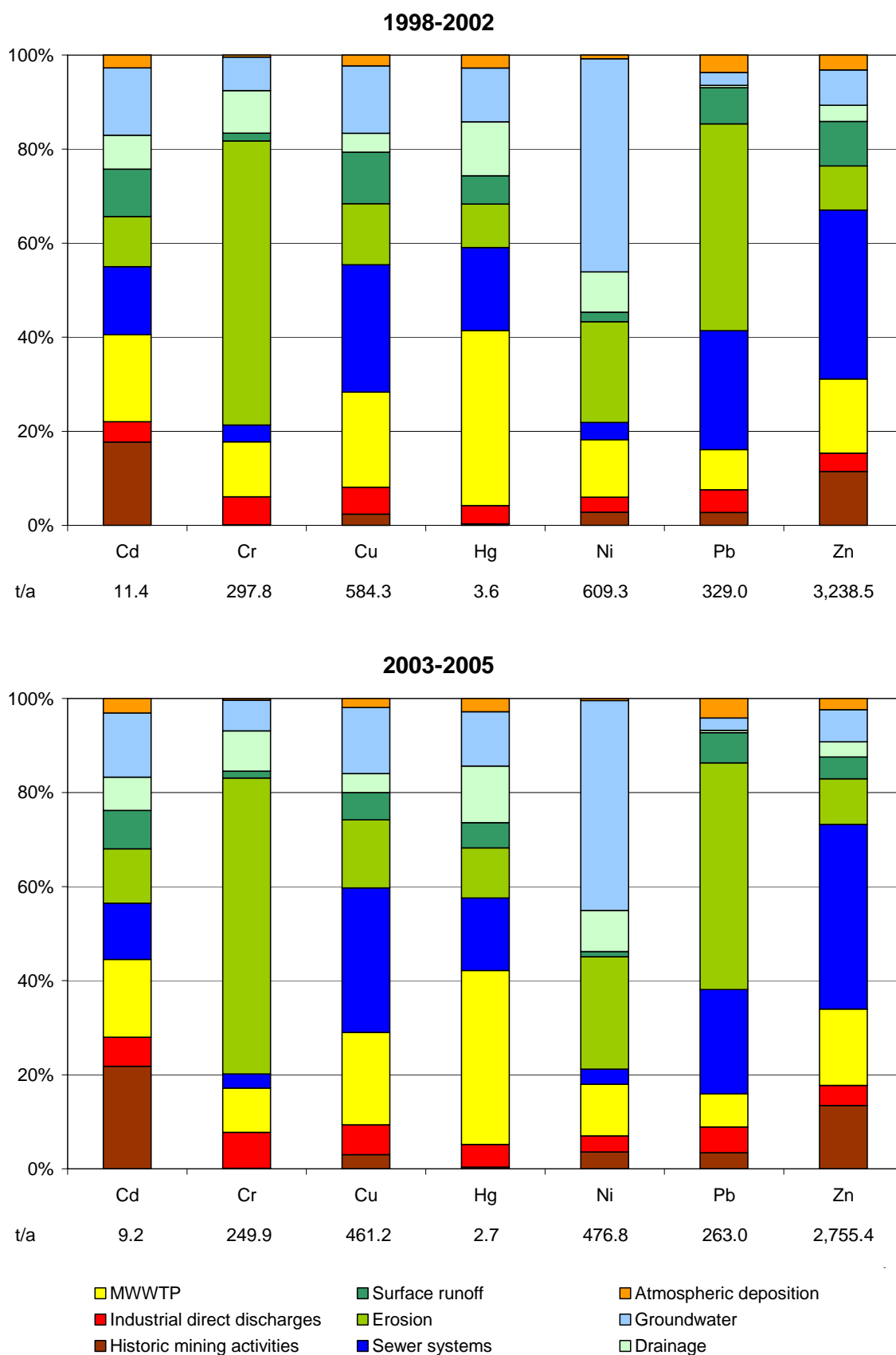


Figure 64: Relative relevance of the individual pathways for the total heavy metal emissions into surface waters in Germany for the balance periods 1998-2002 and 2003-2005

### 5.3.1 Overall view of the heavy metal emissions from Germany from 1985-2005

In the balance period 1983-1987 corpus of the emissions were caused by point pathways, especially by industrial direct dischargers. For Cr, Cu, Hg and Zn the shares for point pathways are between 54 % (Cu) and 80 % (Hg). For Cd about half of the loads are caused by point (47 %) and diffuse pathways (53 %). The most significant pathway with a share of 33 % of total Cd emissions is industrial direct discharges. Emissions from diffuse pathways prevail merely for the metals Pb (80 %) and Ni (63 %). The main pathways for Pb are the sewer systems with 30 % of the total emissions. For Ni the largest share (30 %) results from groundwater inflow (Table 53, Figure 63, Figure 64).

From 1985 to 2005 heavy metal emissions from point pathways were considerably reduced. This is mainly due to the reduction of industrial direct discharges and shows reduction rates of between 75 % for Ni and 95 % for Hg, whereby the main part of the reduction took place in the early 90s (Table 53). The reasons for this is an improved wastewater treatment, higher connection rate of direct dischargers to the public sewer system, emigration of wastewater-intensive industries (e.g. textile industry, leather tanneries), reduction of the amount of used water by reusing the process water and especially by the dismantling of industry by closing several factories in the new states of Germany since the 90s. Nowadays the share of direct industrial discharges is in average below 10 % of the total emissions for all metals (Figure 64).

Emissions from municipal wastewater treatment plants were also significantly reduced from 1985 to 2005, which lies between 43 % for Cu and 80 % for Cr. Reasons being the measures taken in the catchment areas (indirect dischargers, reduction of heavy metal concentrations in stormwater runoff from impervious areas due to a lower atmospheric deposition) as well as the state of process technology achieved in municipal wastewater treatment plants. Due to the further elimination of nutrients also the average efficiency for heavy metal removal was improved. Especially the biological sewage treatment and the phosphate precipitation led to higher removal for heavy metals (FUCHS ET AL., 2002). Except for Cd the emissions from municipal wastewater treatment plants are nowadays the most important point pathway.

The emissions from historical mining activities were assumed constant over the total observation period. Especially for Cd and Zn the balance period 2003-2005 shows a considerable share on the total emissions of 22 % for Cd and 14 % for Zn. For Cd the emissions from historic mining activities are the most important point source. As already described in chapter 4.2.1.3 it must be assumed that the emissions from this source are even higher in reality as not all emissions could be recorded.

Due to the reduction of point emissions since the mid 1990s the major part of the heavy metal emissions into surface waters in Germany is caused by diffuse pathways. In the balance period 2003-2005 the share of diffuse emissions is between 55 % for Cd and 84 % for Pb.

The direct atmospheric deposition onto water surface in the balance period 2003-2005 amounts in average for all metals to less than 5% of the total emissions into surface waters in Germany. The reduction rates for this pathway since 1985 are between 82 % (Cr, Zn) and 98 % (Cd) due to the improved purification of exhaust gas of industrial emissions into the



atmosphere and the introduction of unleaded petrol (comp. paragraph 4.2.2.1). The reduction of atmospheric deposition rates is also especially important for pathways that are characterised by stormwater runoff processes from the unsealed and impervious surfaces, such as sewer systems and surface runoff from unsealed areas.

In the balance period 2003-2005 emissions from sewer systems are the most important pathway for Cu and Zn with a share of 31 % resp. 39 %. Emissions from sewer systems also are important for Cd, Hg and Pb with shares between 12 % (Cd) and 22 % (Pb) in the total emissions. From 1985-2005 the emissions from sewer systems were drastically reduced for Cd, Cr, Hg, Ni and Pb by at least 80 %. This is mainly due to the already described reduction of atmospheric deposition rates. For Cu and Zn the decrease is much lower and is 23 % for Cu and 27 % for Zn within the period 1985-2005. The main sources for Cu and Zn in urban areas are the corrosion of metal surfaces (roofs and rain gutters as well as zinc-plated products) and traffic (tyre and brake pad) (comp. paragraph 4.2.2.6). HILLENBRAND ET AL. (2005) used a source-specific approach to calculate the emissions of Cu, Pb and Zn from urban areas into surface waters in Germany. Figure 65 shows the results of the calculated source-specific emissions from storm sewers according to HILLENBRAND ET AL. (2005) compared to the emissions from storm sewers in Germany calculated with MONERIS.

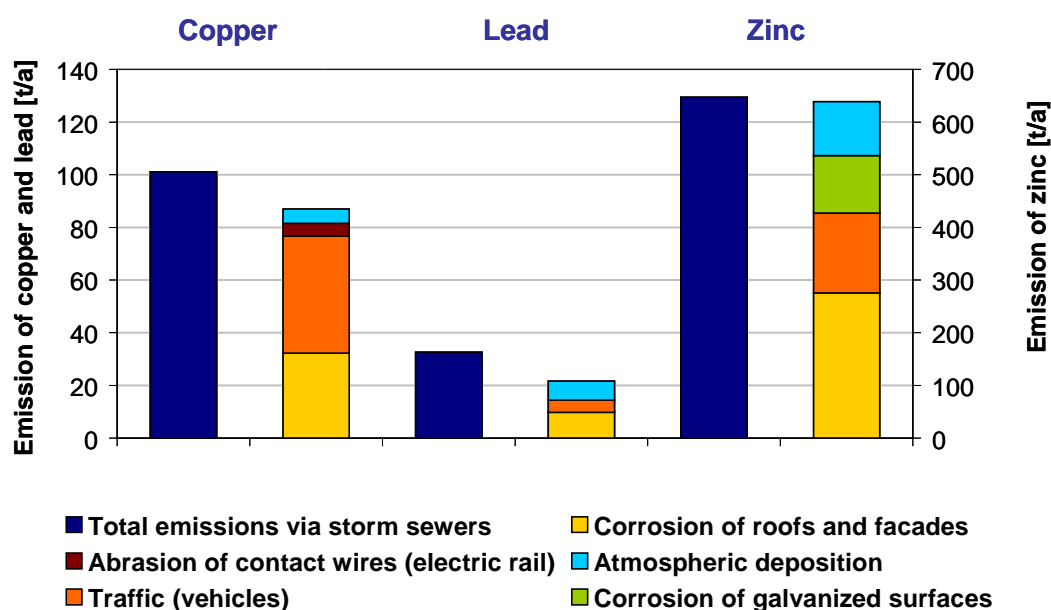


Figure 65: Comparison of the calculated emissions from storm sewers for copper, lead and zinc with the source-specific emission approach according to HILLENBRAND ET AL. (2005)

Even though different input data and calculation methods were used by the both different quantification approaches, they both show very similar results. The source-specific emission calculation point out the relevance of corrosion from metal surfaces and of traffic for the emissions of Cu, Pb and Zn via sewer systems. For Cu and Zn, the atmospheric deposition only plays a minor role for the pollution of impervious urban areas as well as for Pb meanwhile, too.

Emissions via surface runoff from unsealed areas are significantly affected by the heavy metal concentration in precipitation. According to this high reduction rates of 82 % (Cr, Zn)

and 98 % (Cd) were achieved for this pathway. The share of emissions via surface runoff from unsealed areas in the total emissions for the balance period 2003-2005 is between 1 % for Ni and 8 % for Cd. Furthermore, apart from the heavy metals contained in precipitation the fertilisers containing heavy metals can be washed off unsealed areas (comp. paragraph 4.2.2.3). The share of emissions resulting from washed off fertilisers of the total emissions via surface runoff is below 15 % for Cd, Hg, Ni and Pb in the current balance period. For Cr 48 % of emissions resulting from surface runoff are from washed out fertilisers. This is mainly due to the marginal relevance of atmospheric deposition compared to emissions from other sources. For Zn a share of 22 % and for Cu of 29 % was calculated. Especially pig slurry shows very high amounts of Cu and Zn that can be lead back to the mineral feed enriched with Cu and Zn.

For the metals Cr and Pb erosion is the most important pathway in the balance period 2003-2005 with a share of 63 % (Cr) and 48 % (Pb) in the total emissions. For Ni the share is 24 % and for the rest of the metals about 10 % of the total emissions. The loads from erosion only slightly changed throughout the whole observation period, whereby the precipitation pattern had the most influence. For the period with a high precipitation level, the period 1998-2002, an increase was observed and for the low precipitation period 2003-2005 a decrease in erosion was observed compared to 1983-1987.

Emissions from the pathway groundwater inflow are especially important for Ni. In the balance year 2003-2005 45 % of the Ni emissions resulted from groundwater. For the other metals the share in the total emissions is between 3 % (Pb) and 14 % (Cu, Cd). Changes during the observation period from 1985 to 2005 are also significantly influenced by the hydrology.

The share of drainage for all heavy metals with the exception of Hg is below 10 % of the total emissions. For Hg a share of 12 % was calculated that is quite uncertain as there were only measurement readings from seepage water that were below the limit of quantification and therefore half of the limit of quantification was used (paragraph 4.1.2.5).

In Figure 66 the total emissions for the four balance periods is graphically depicted, differentiated in point and diffuse pathways (data from Table 53). For the diffuse pathways the reduction over the complete observation period varies between 28 % for Cr and 85 % for Cd. It can generally be said that the main reason for the reduction of diffuse emissions lies in the reduction of atmospheric deposition rates. Metals that had diffuse emissions in the balance period 1983-1987 which were mainly caused by pathways that had atmospheric deposition as main contribution source (direct deposition onto water surface, surface runoff from unsealed and impervious areas) such as Cd, Hg and Pb therefore have the highest reduction rates. The emissions from the most important diffuse pathways for Cr (erosion), Ni (groundwater inflow) as well as Cu and Zn (sewer systems) was barely reduced as already described.

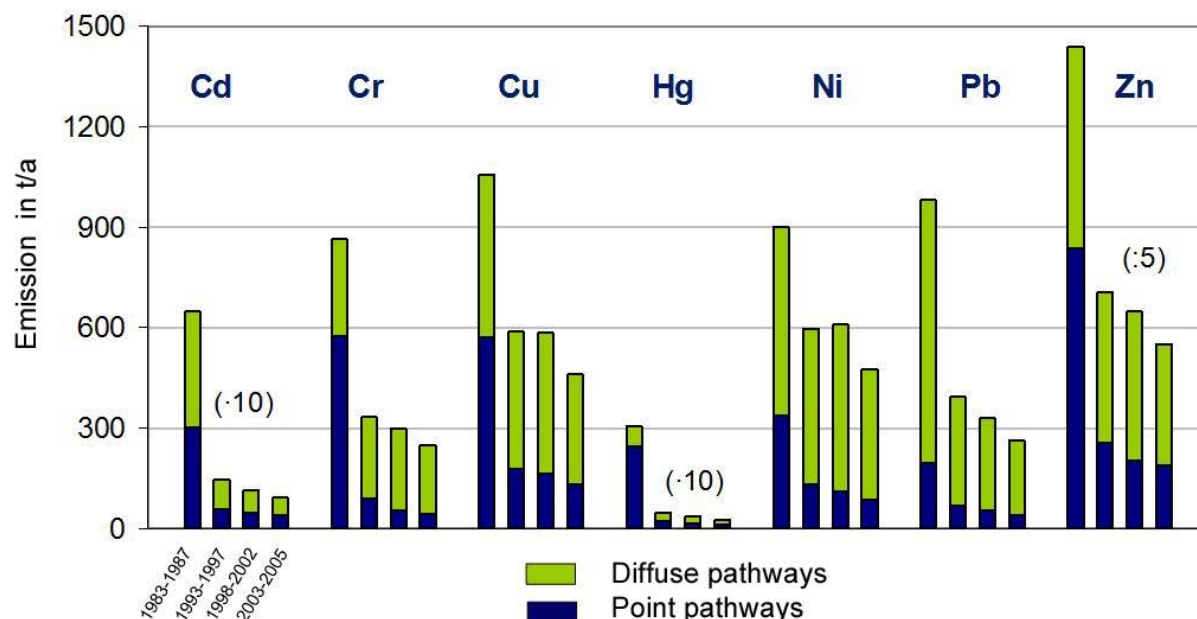


Figure 66: Reduction of point and diffuse heavy metal emissions into Germany's surface waters for 1985, 1995, 2000 and 2005

The reduction of the total heavy metal emissions from Germany into surface waters between 1985 and 2005 depending on the considered metal is between 47 % for Ni and 91 % for Hg (Table 53 and Figure 66).

### 5.3.2 Regional importance of the pathways

Figure 67 to Figure 80 show the spatial distribution of the heavy metal emissions in the sub-basins as well as the importance of the pathways in the river basin districts for the current balance period 2003-2005. In Table 54 to Table 60 the point sources and diffuse emissions for all metals for the four balance periods and the river basin districts are depicted.

In the balance period 2003-2005 the emissions for Cd from historic mining activities are the most important pathway with 22 % in average for Germany. Most of the recorded emissions from this pathway are from the Erzgebirge ("Ore Mountains") and cause 56 % of the total emissions in the river basin district of the Elbe.

Erosion is the most important pathway for Cr. The main loads are from the Alps and the agronomically used regions around the Mittelgebirge (low mountain ranges). The main pressures at the Elbe estuary as well as in Hamburg and Berlin result from industrial direct dischargers and municipal wastewater treatment plants.

Cadmium

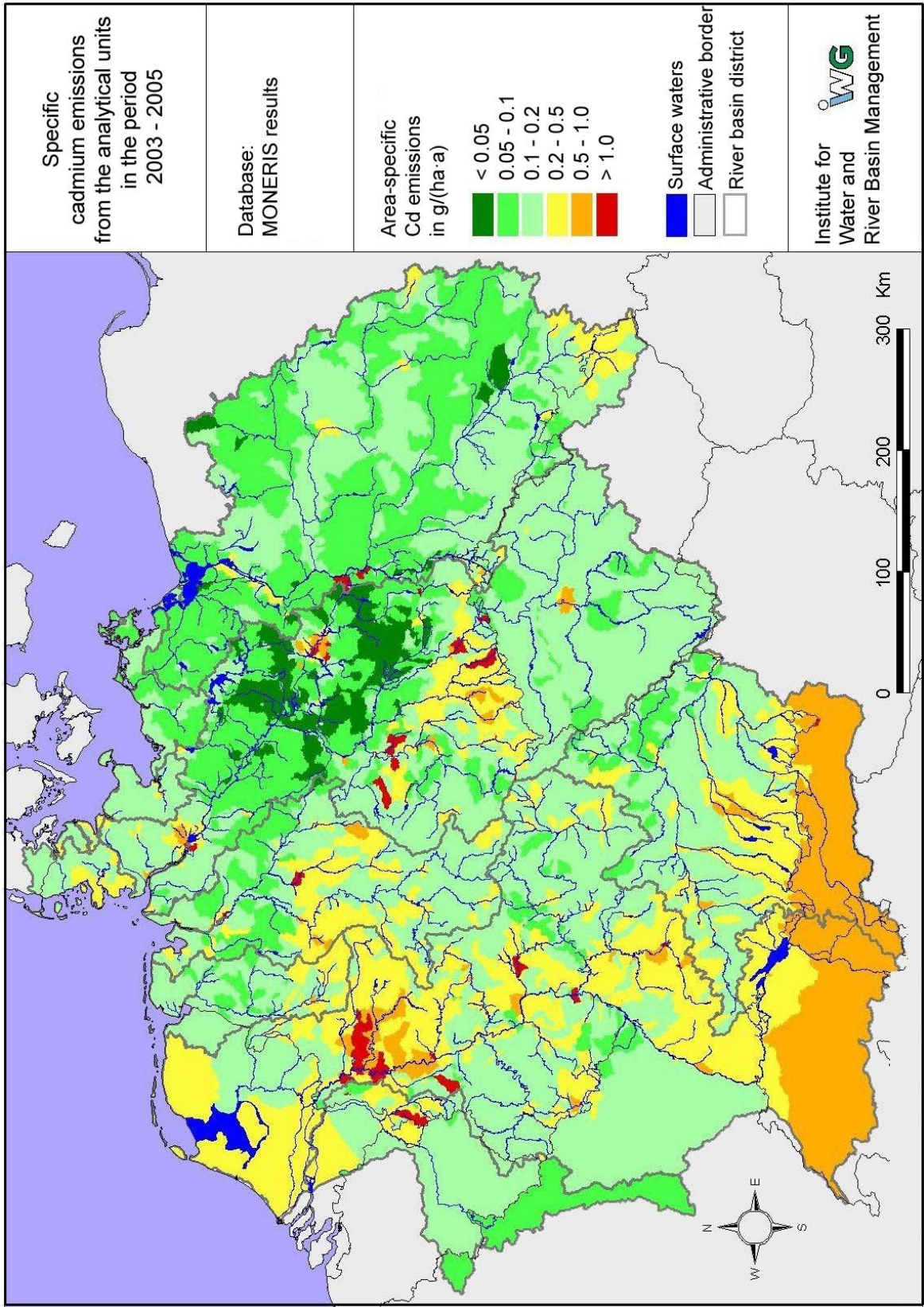


Figure 67: Area-specific Cd-emissions from the analytical units into the surface water bodies in the period 2003-2005

Table 54: Point and diffuse cadmium emissions from Germany into the river basin districts and seas in 1983-1987, 1993-1997, 1998-2002 and 2003-2005

Cadmium	Point emissions in t/a				Diffuse emissions in t/a			
Period	83-87	93-97	98-02	03-05	83-87	93-97	98-02	03-05
Danube	0.44	0.29	0.24	0.19	3.09	1.70	1.32	0.98
Rhine	4.60	2.17	1.55	1.23	6.26	3.03	2.41	1.72
Meuse	0.13	0.06	0.05	0.07	0.22	0.09	0.07	0.05
Ems	0.13	0.07	0.06	0.04	0.54	0.31	0.22	0.18
Weser	1.64	0.36	0.25	0.18	2.43	1.14	0.88	0.68
Elbe	23.01	2.87	2.41	2.35	14.20	1.82	1.35	1.09
Odra	0.10	0.04	0.03	0.05	4.30	0.20	0.13	0.11
Eider	0.01	0.00	0.00	0.00	0.23	0.12	0.11	0.09
Schlei/Trave	0.02	0.01	0.01	0.00	0.39	0.15	0.11	0.09
Warnow/Peene	0.29	0.08	0.01	0.00	2.85	0.22	0.16	0.14
North Sea	29.51	5.54	4.32	3.87	23.87	6.51	5.04	3.81
Baltic Sea	0.41	0.13	0.05	0.05	7.54	0.57	0.40	0.33
Black Sea	0.44	0.29	0.24	0.19	3.09	1.70	1.32	0.98
Germany	30.37	5.96	4.61	4.11	34.50	8.77	6.76	5.12

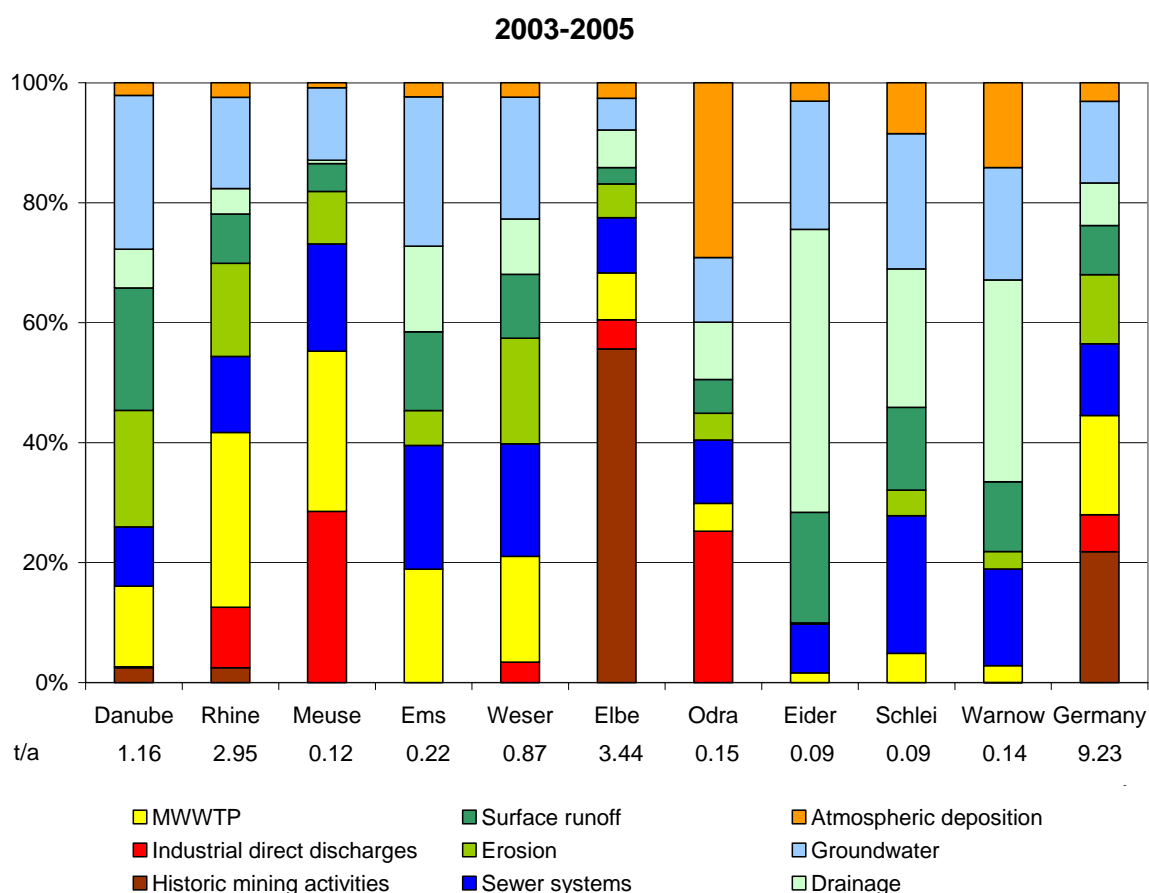


Figure 68: Relative importance of the pathways for cadmium emissions into the river basin districts of Germany in the balance period 2003-2005



Chromium

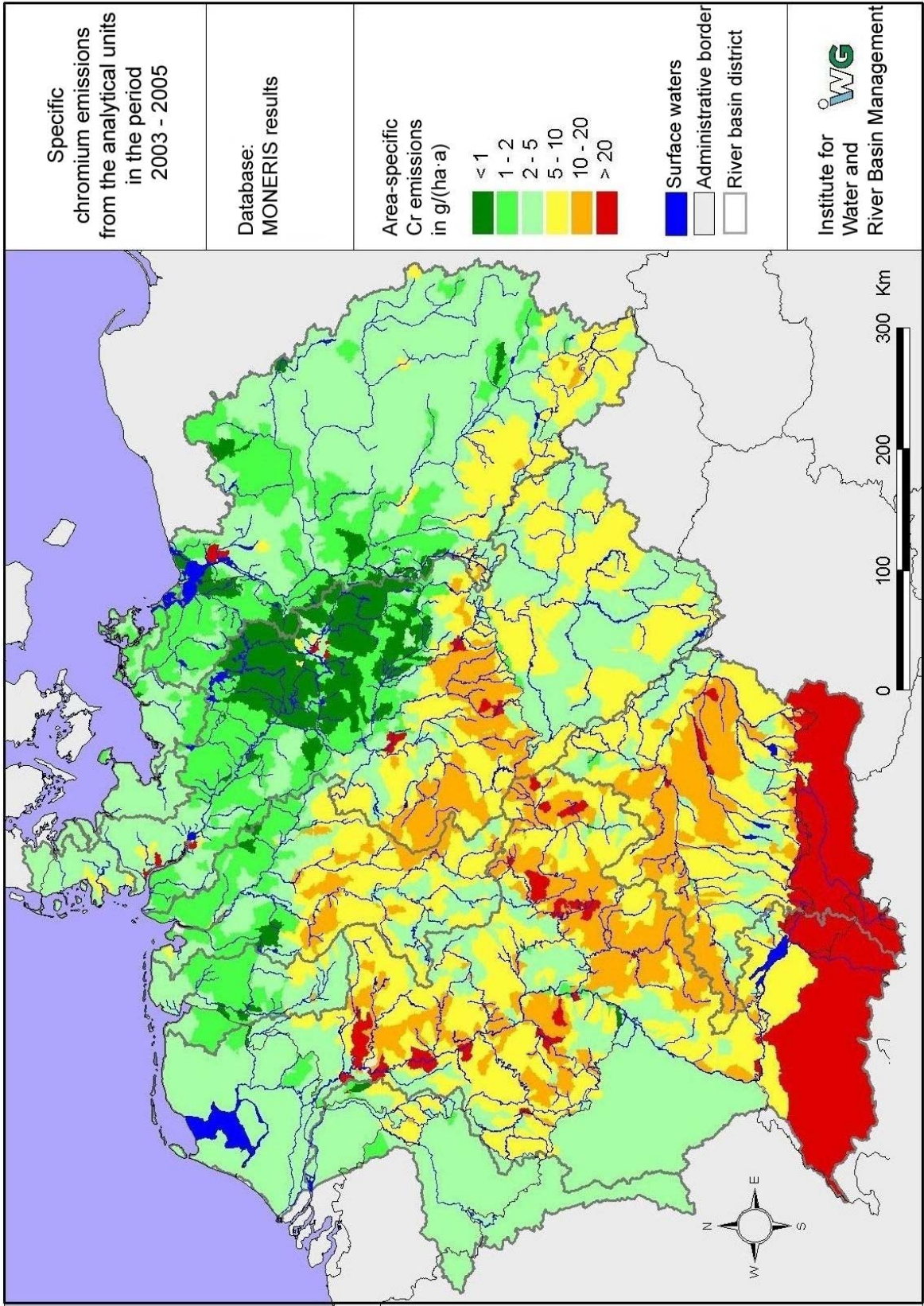


Figure 69: Area-specific Cr-emissions from the analytical units into the surface water bodies in the period 2003-2005



Table 55: Point and diffuse chromium emissions from Germany into the river basin districts and seas in 1983-1987, 1993-1997, 1998-2002 and 2003-2005

Chromium	Point emissions in t/a				Diffuse emissions in t/a			
Period	83-87	93-97	98-02	03-05	83-87	93-97	98-02	03-05
Danube	8.29	6.55	5.21	2.26	66.19	60.98	62.49	53.52
Rhine	248.02	54.79	32.60	24.66	114.93	94.28	97.91	80.56
Meuse	2.99	1.34	0.77	0.57	2.17	1.40	1.38	1.17
Ems	2.28	1.53	1.00	0.63	5.04	4.00	3.84	3.27
Weser	30.23	10.40	5.32	5.99	30.27	25.40	26.07	22.18
Elbe	263.29	15.34	7.56	8.46	56.13	45.69	43.66	38.41
Odra	19.16	0.34	0.24	0.21	3.32	2.02	1.85	1.53
Eider	0.75	0.09	0.03	0.02	2.66	2.02	2.30	1.89
Schlei/Trave	0.26	0.15	0.07	0.05	2.86	2.11	2.10	1.74
Warnow/Peene	1.58	0.36	0.08	0.04	4.64	3.29	3.32	2.76
North Sea	547.58	83.49	47.28	40.33	211.21	172.80	175.15	147.47
Baltic Sea	21.00	0.85	0.39	0.31	10.83	7.42	7.26	6.03
Black Sea	8.29	6.55	5.21	2.26	66.19	60.98	62.49	53.52
Germany	576.87	90.89	52.88	42.89	288.22	241.21	244.90	207.02

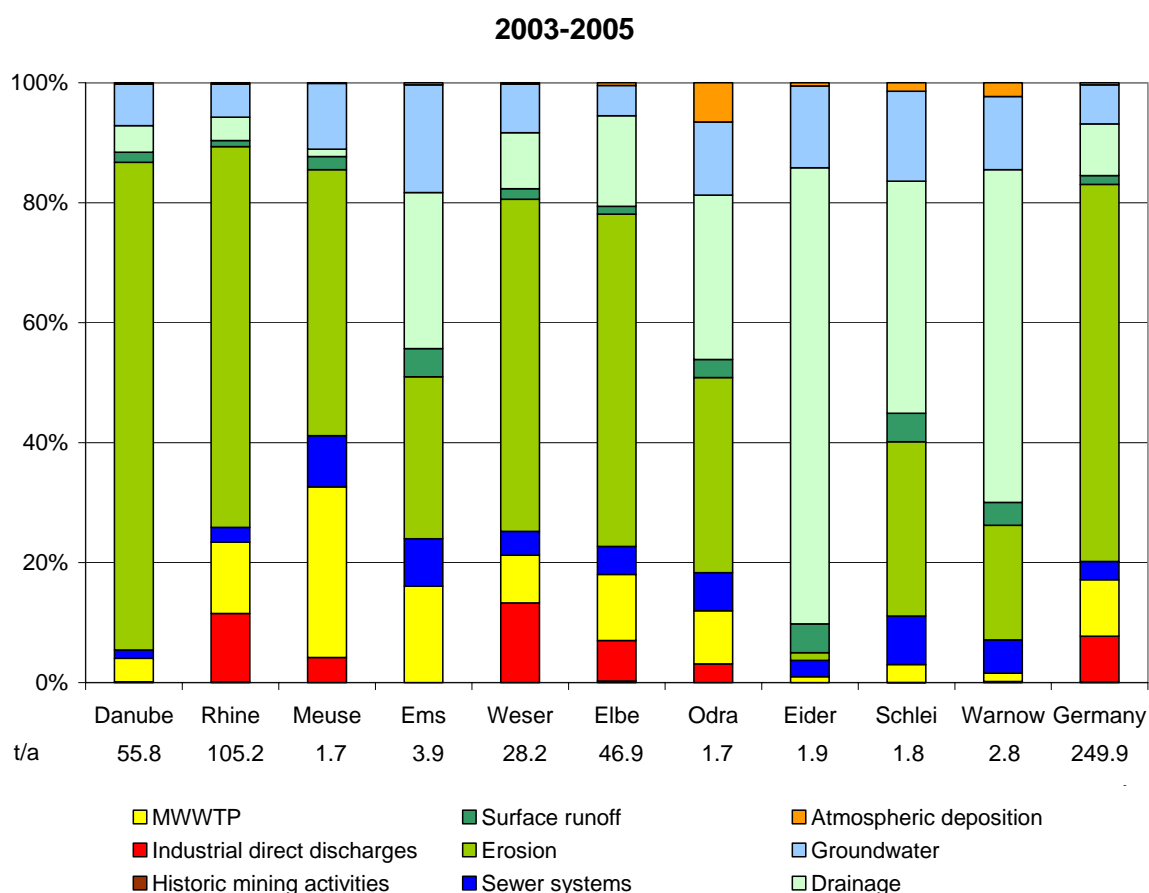


Figure 70: Relative importance of the pathways for chromium emissions into the river basin districts of Germany in the balance period 2003-2005

Copper

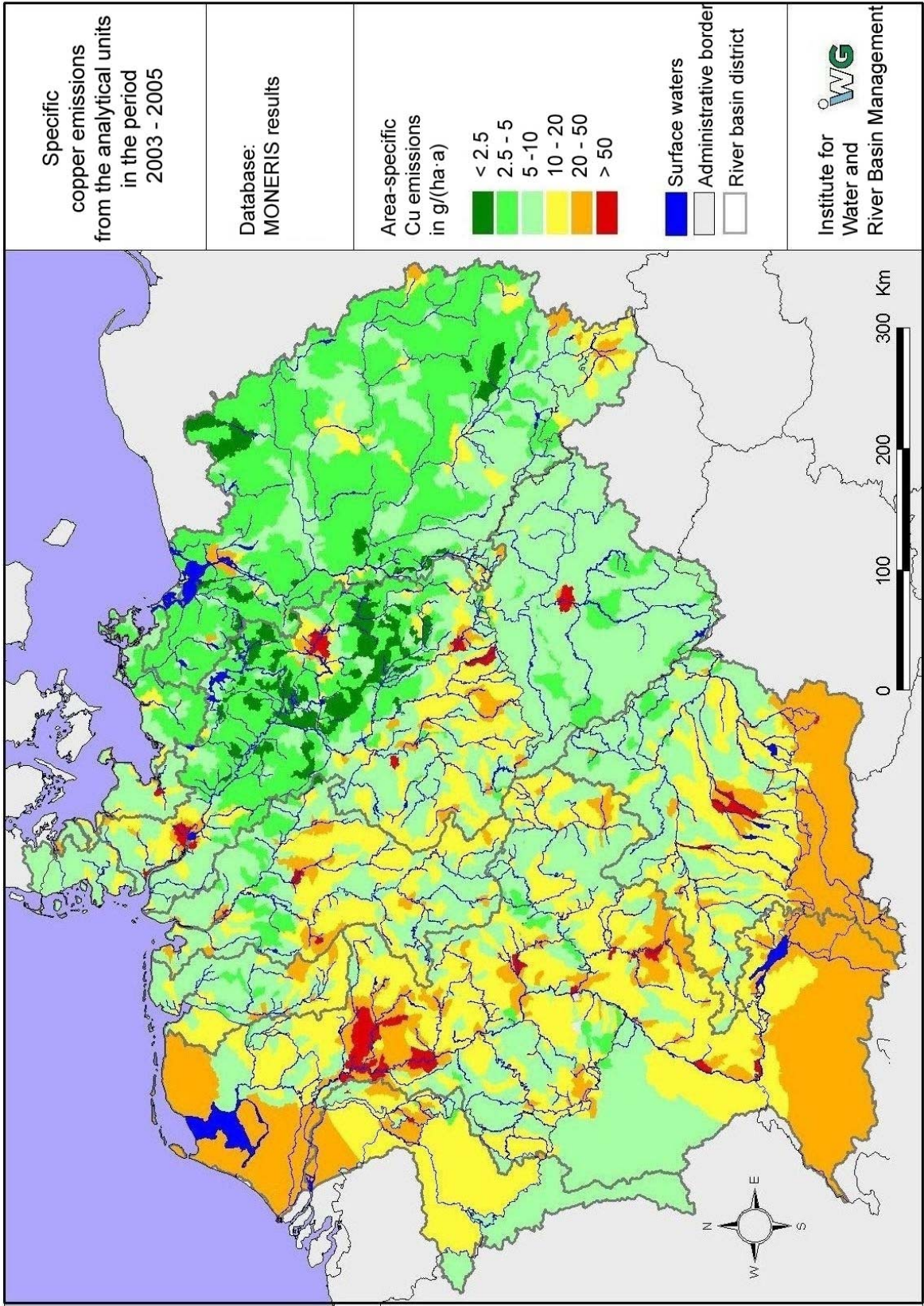


Figure 71: Area-specific Cu-emissions from the analytical units into the surface water bodies in the period 2003-2005

Table 56: Point and diffuse copper emissions from Germany into the river basin districts and seas in 1983-1987, 1993-1997, 1998-2002 and 2003-2005

Copper	Point emissions in t/a				Diffuse emissions in t/a			
Period	83-87	93-97	98-02	03-05	83-87	93-97	98-02	03-05
Danube	21.6	18.5	16.8	11.7	80.2	74.7	80.3	57.0
Rhine	185.9	110.6	103.2	85.2	181.5	150.5	156.8	116.3
Meuse	3.8	3.1	3.1	2.2	6.2	4.9	5.0	4.2
Ems	3.9	3.3	3.2	3.0	14.2	13.8	13.5	11.4
Weser	25.5	12.1	9.6	7.7	56.9	53.4	54.0	44.6
Elbe	326.7	27.4	26.7	21.3	98.9	85.6	82.4	72.1
Odra	0.4	0.5	0.5	0.7	17.9	6.9	6.4	5.1
Eider	1.6	0.7	0.4	0.3	5.5	4.3	5.1	4.0
Schlei/Trave	2.3	1.9	1.3	0.9	8.2	6.5	6.6	5.5
Warnow/Peene	0.6	0.7	0.7	0.7	14.4	8.7	8.7	7.3
North Sea	547.3	157.3	146.2	119.7	363.2	312.3	316.7	252.6
Baltic Sea	3.3	3.1	2.5	2.2	40.5	22.1	21.7	17.9
Black Sea	21.6	18.5	16.8	11.7	80.2	74.7	80.3	57.0
Germany	572.2	178.8	165.6	133.7	483.9	409.1	418.7	327.5

### 2003-2005

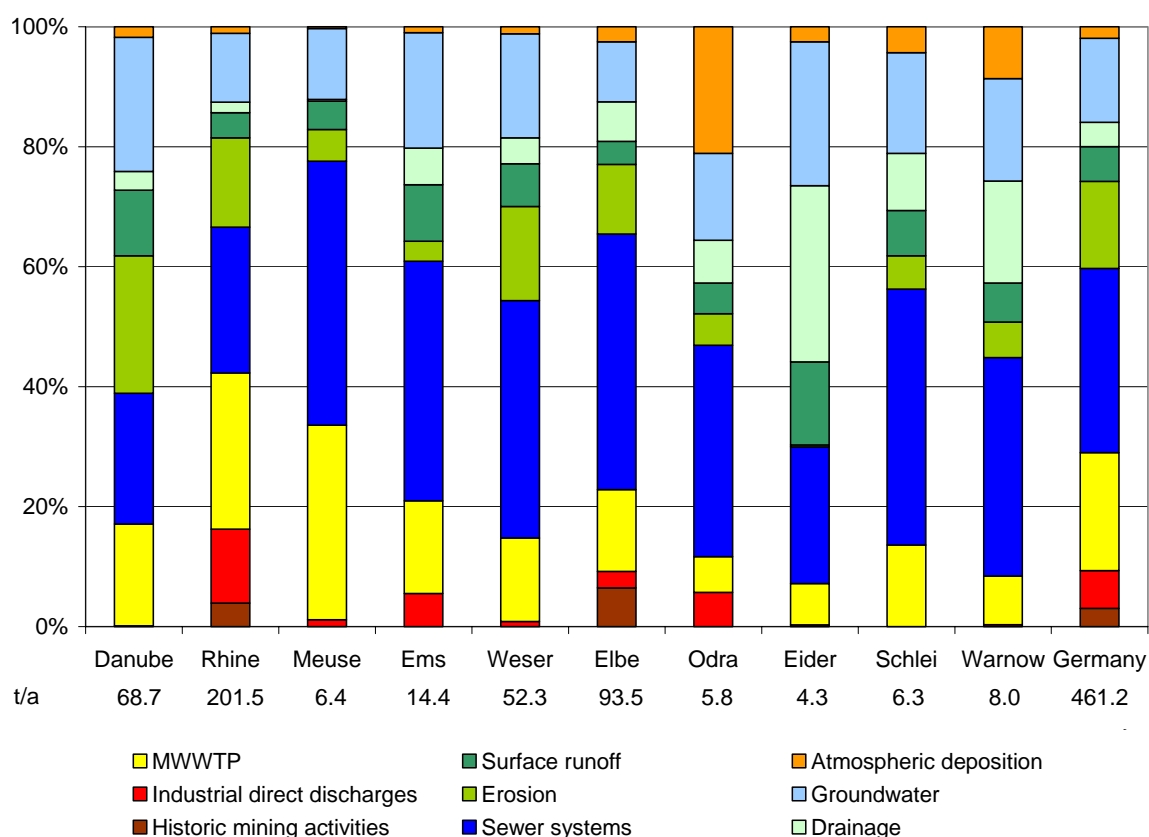


Figure 72: Relative importance of the pathways for copper emissions into the river basin districts of Germany in the balance period 2003-2005



Mercury

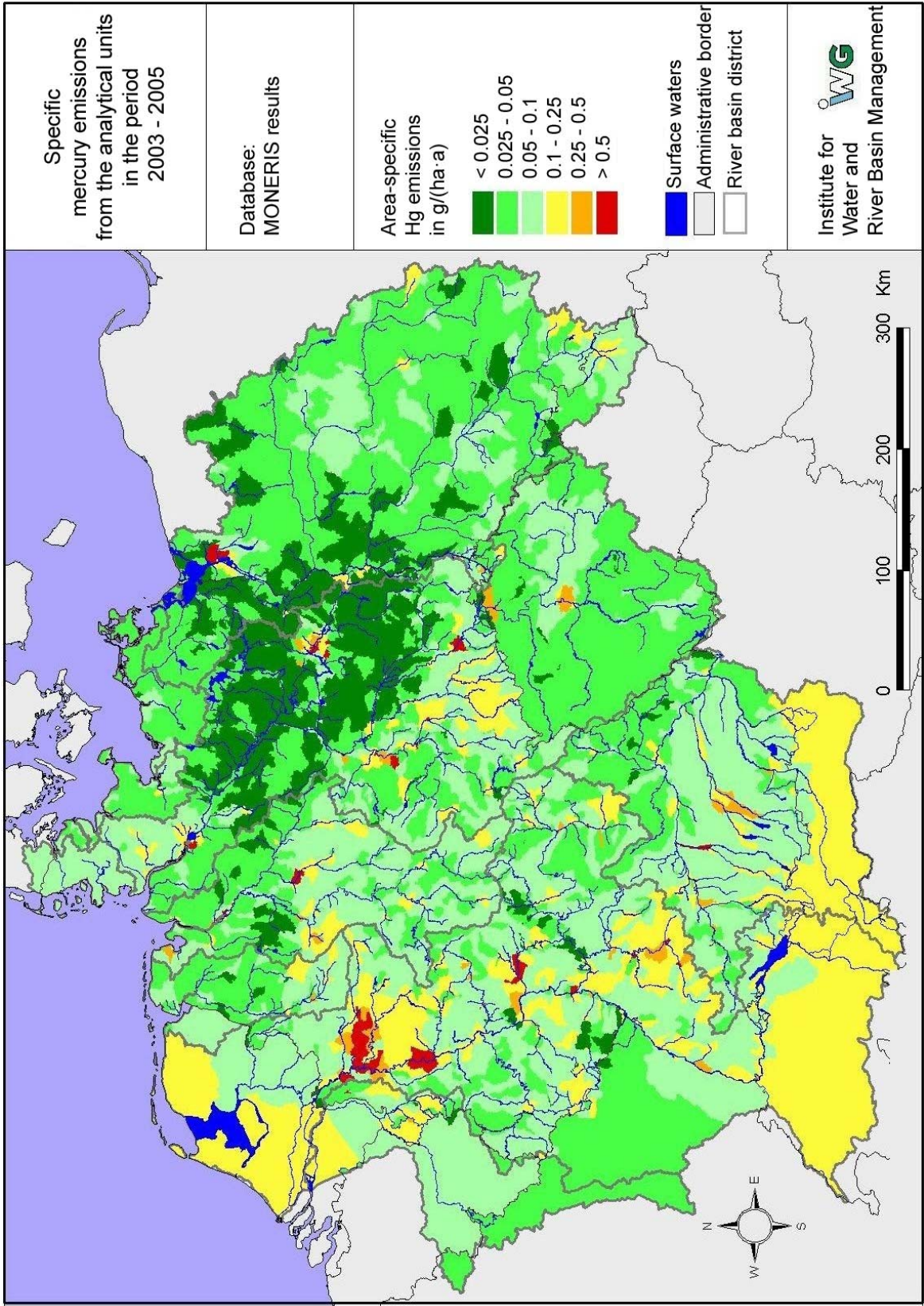


Figure 73: Area-specific Hg-emissions from the analytical units into the surface water bodies in the period 2003-2005

Table 57: Point and diffuse mercury emissions from Germany into the river basin districts and seas in 1983-1987, 1993-1997, 1998-2002 and 2003-2005

Mercury	Point emissions in t/a				Diffuse emissions in t/a			
Period	83-87	93-97	98-02	03-05	83-87	93-97	98-02	03-05
Danube	0.296	0.212	0.149	0.133	0.924	0.429	0.375	0.280
Rhine	2.246	1.255	0.827	0.653	2.001	0.870	0.733	0.519
Meuse	0.055	0.038	0.028	0.029	0.070	0.027	0.021	0.015
Ems	0.072	0.052	0.038	0.027	0.170	0.084	0.068	0.054
Weser	0.491	0.256	0.163	0.108	0.651	0.311	0.254	0.196
Elbe	21.380	0.407	0.258	0.186	1.541	0.637	0.462	0.367
Odra	0.008	0.006	0.006	0.006	0.288	0.080	0.040	0.031
Eider	0.014	0.004	0.001	0.001	0.079	0.039	0.042	0.034
Schlei/Trave	0.013	0.009	0.004	0.003	0.109	0.045	0.037	0.029
Warnow/Peene	0.020	0.013	0.003	0.002	0.229	0.083	0.059	0.048
North Sea	24.258	2.013	1.315	1.003	4.512	1.968	1.578	1.186
Baltic Sea	0.041	0.028	0.013	0.011	0.626	0.209	0.136	0.108
Black Sea	0.296	0.212	0.149	0.133	0.924	0.429	0.375	0.280
Germany	24.596	2.253	1.477	1.147	6.062	2.606	2.089	1.573

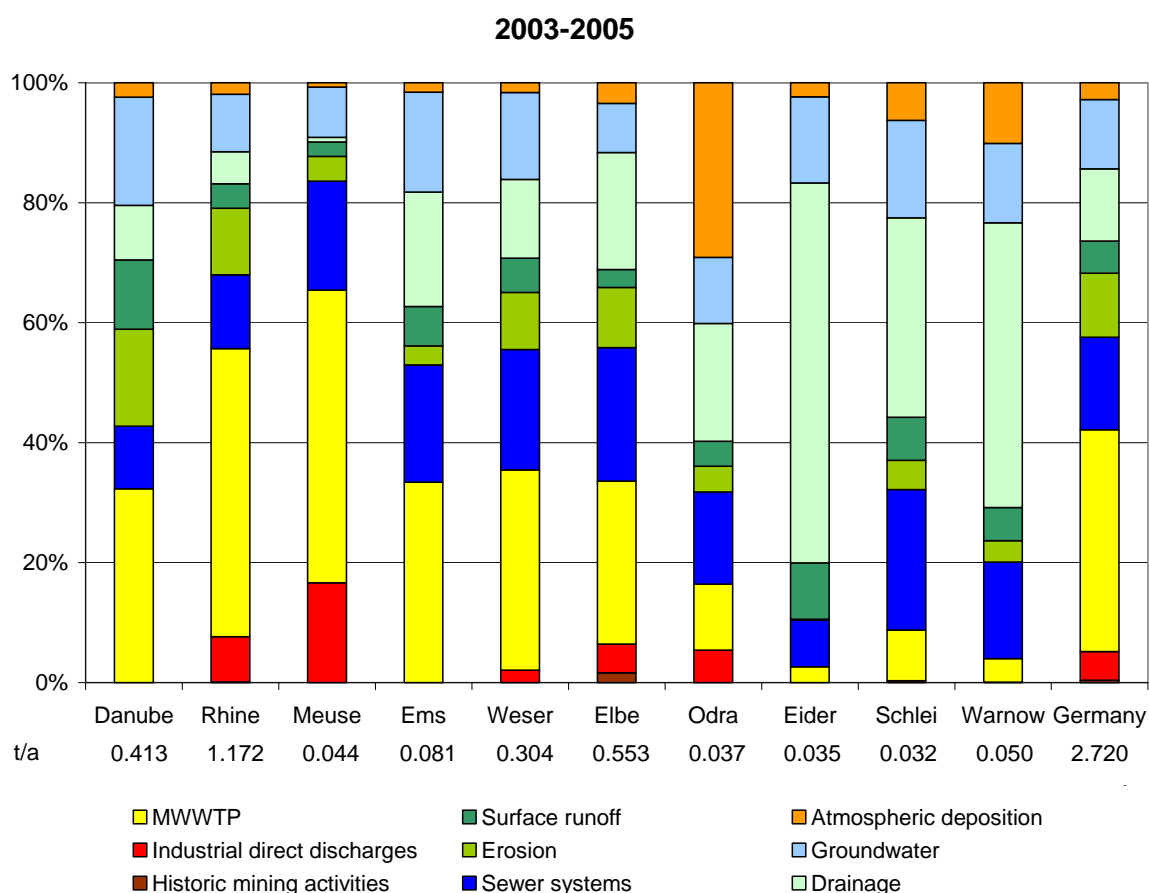


Figure 74: Relative importance of the pathways for mercury emissions into the river basin districts of Germany in the balance period 2003-2005



Nickel

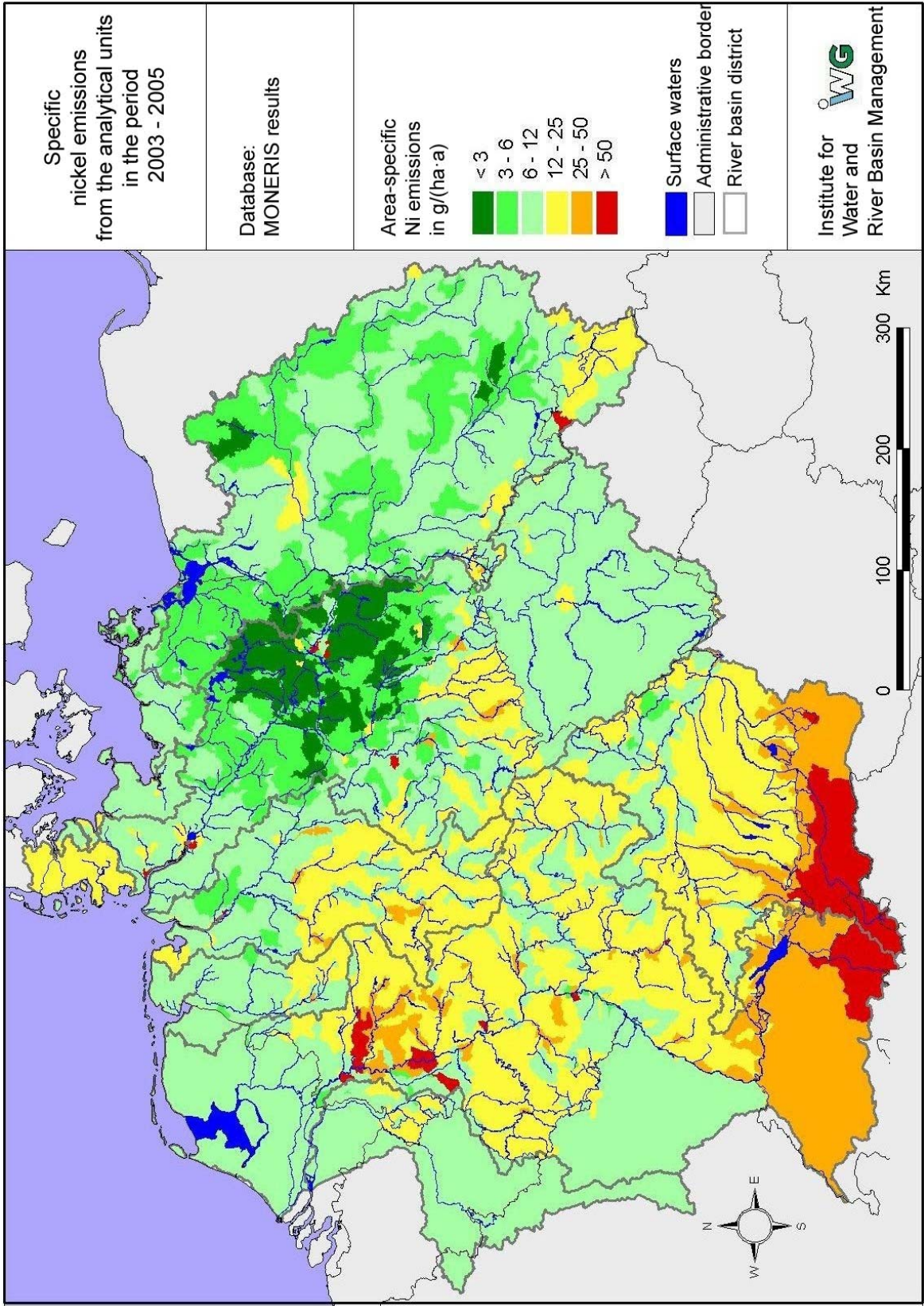


Figure 75: Area-specific Ni-emissions from the analytical units into the surface water bodies in the period 2003-2005



Table 58: Point and diffuse nickel emissions from Germany into the river basin districts and seas in 1983-1987, 1993-1997, 1998-2002 and 2003-2005

Nickel	Point emissions in t/a				Diffuse emissions in t/a			
Period	83-87	93-97	98-02	03-05	83-87	93-97	98-02	03-05
Danube	11.5	8.9	7.0	4.9	108.8	99.3	112.9	87.2
Rhine	150.7	82.4	74.2	56.4	215.4	174.8	194.1	145.3
Meuse	3.2	2.3	2.5	1.4	5.6	3.7	4.1	3.4
Ems	3.3	2.2	2.4	1.4	16.4	15.3	15.1	12.6
Weser	38.4	9.1	6.7	5.5	71.5	62.8	65.8	53.4
Elbe	124.9	26.9	16.7	14.8	102.4	79.0	76.1	64.3
Odra	1.1	0.6	0.5	0.4	13.5	6.3	5.9	4.5
Eider	0.7	0.2	0.2	0.1	8.0	6.2	7.7	6.5
Schlei/Trave	0.7	0.5	0.5	0.3	8.6	6.6	6.8	5.6
Warnow/Peene	2.2	0.8	0.3	0.3	14.1	9.5	9.7	8.4
North Sea	321.1	123.3	102.7	79.7	419.3	341.9	362.9	285.4
Baltic Sea	4.1	1.9	1.3	1.0	36.2	22.5	22.5	18.6
Black Sea	11.5	8.9	7.0	4.9	108.8	99.3	112.9	87.2
Germany	336.7	134.0	111.0	85.6	564.3	463.7	498.3	391.2

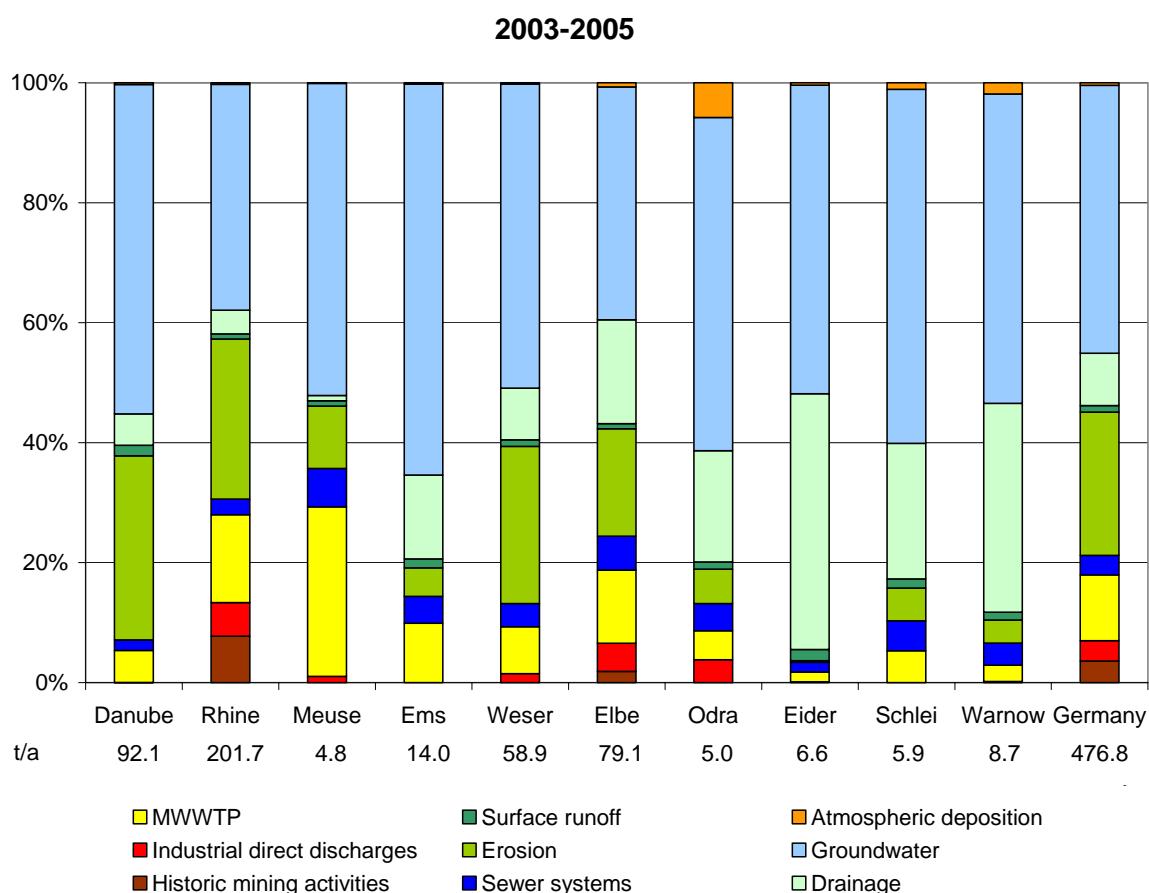


Figure 76: Relative importance of the pathways for nickel emissions into the river basin districts of Germany in the balance period 2003-2005

Lead

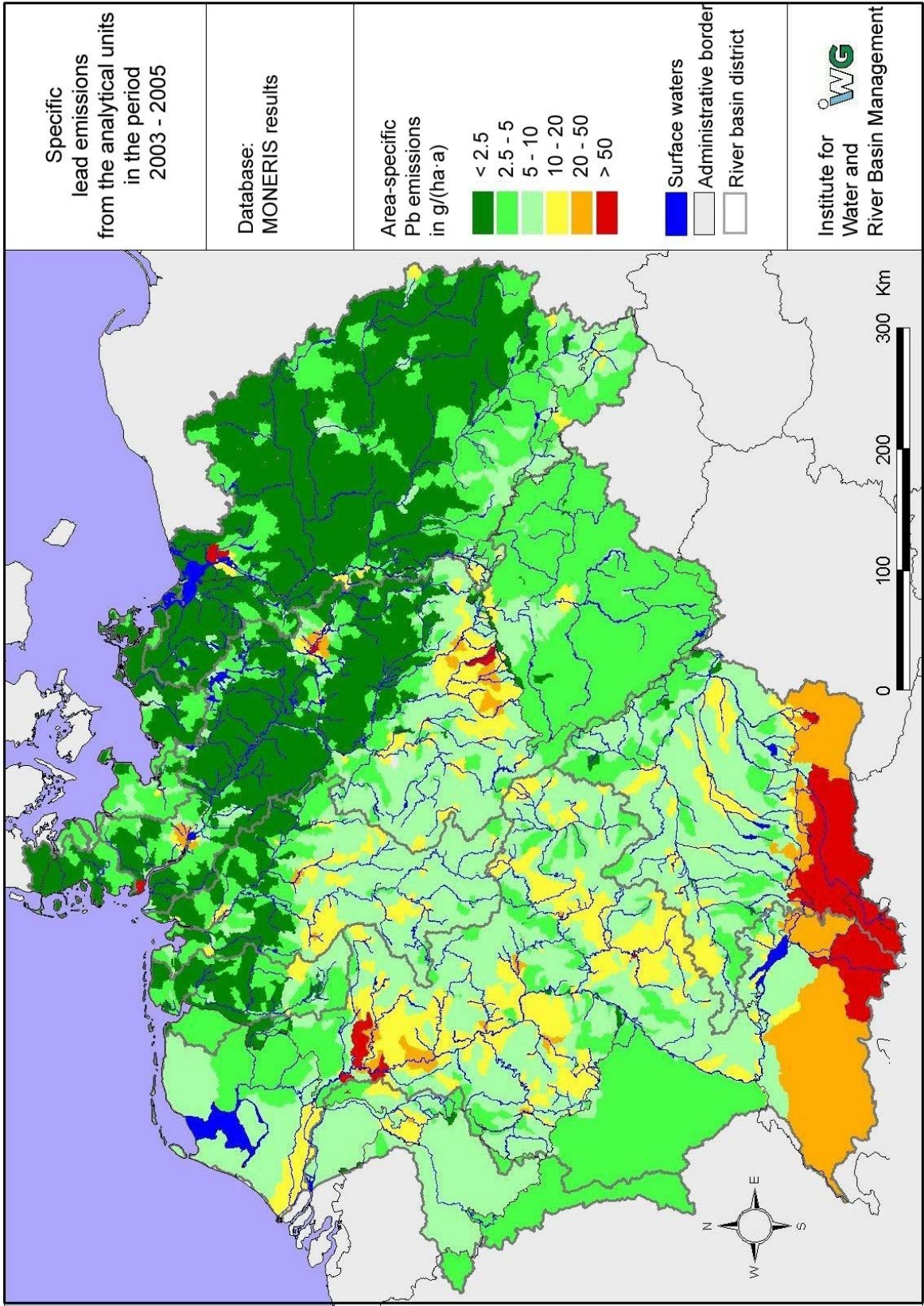


Figure 77: Area-specific Pb-emissions from the analytical units into the surface water bodies in the period 2003-2005

Table 59: Point and diffuse lead emissions from Germany into the river basin districts and seas in 1983-1987, 1993-1997, 1998-2002 and 2003-2005

Lead	Point emissions in t/a				Diffuse emissions in t/a			
Period	83-87	93-97	98-02	03-05	83-87	93-97	98-02	03-05
Danube	10.71	6.28	3.78	2.33	149.54	62.69	55.35	44.99
Rhine	110.19	36.26	28.02	22.15	290.86	124.78	109.49	85.05
Meuse	1.32	0.77	0.61	0.50	9.33	3.66	2.87	2.21
Ems	1.78	1.10	0.94	0.58	21.01	8.12	5.91	4.62
Weser	26.78	5.01	4.21	2.19	90.34	40.13	33.87	27.24
Elbe	44.39	16.47	14.21	13.62	159.22	70.35	56.53	47.13
Odra	0.51	0.38	0.97	0.43	25.08	5.19	3.94	3.35
Eider	0.16	0.06	0.03	0.02	7.43	1.74	1.41	1.13
Schlei/Trave	0.35	0.20	0.08	0.06	12.49	3.98	2.97	2.38
Warnow/Peene	1.35	0.82	0.16	0.05	18.94	4.98	3.67	3.04
North Sea	184.61	59.68	48.01	39.05	578.18	248.79	210.08	167.37
Baltic Sea	2.21	1.40	1.21	0.53	56.52	14.15	10.58	8.77
Black Sea	10.71	6.28	3.78	2.33	149.54	62.69	55.35	44.99
Germany	197.53	67.35	52.99	41.92	784.24	325.63	276.01	221.12

### 2003-2005

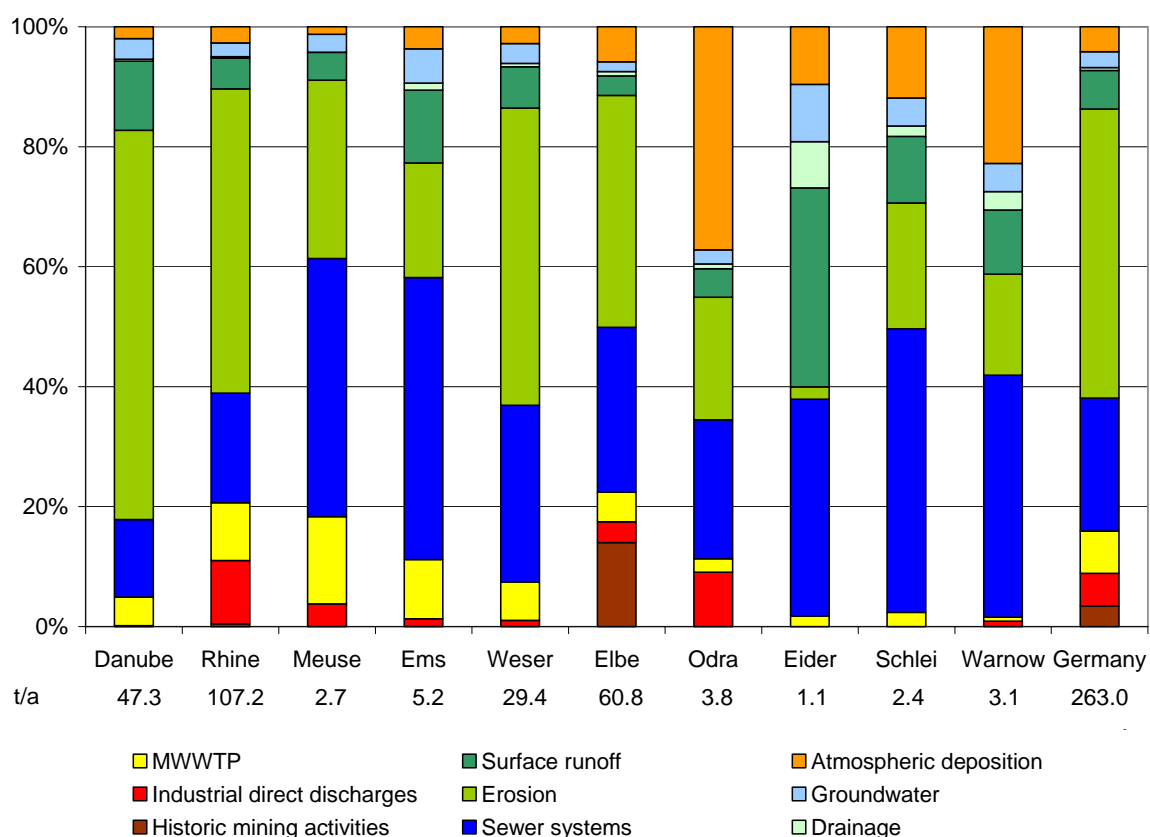


Figure 78: Relative importance of the pathways for lead emissions into the river basin districts of Germany in the balance period 2003-2005



Zinc

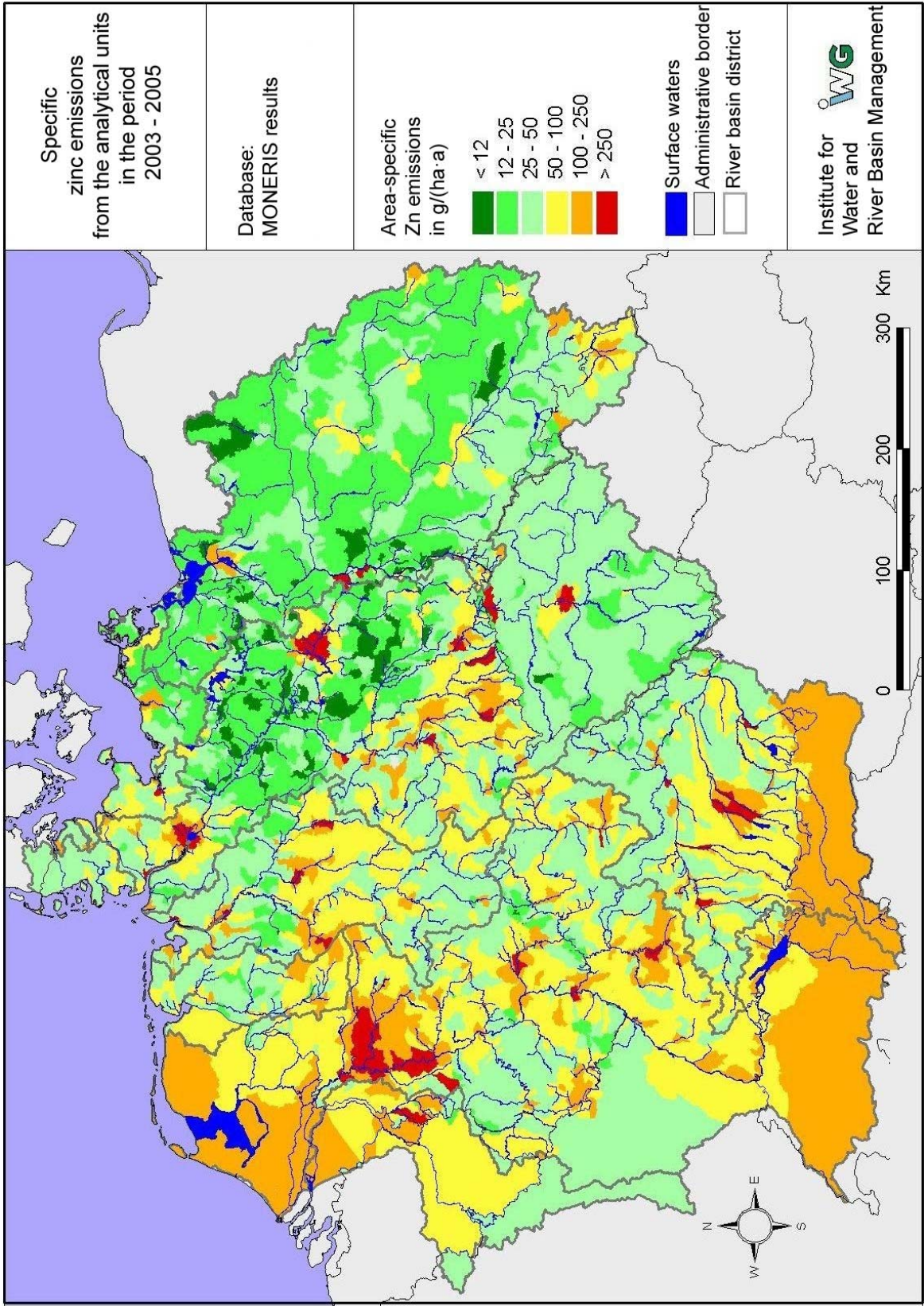


Figure 79: Area-specific Zn-emissions from the analytical units into the surface water bodies in the period 2003-2005

Table 60: Point and diffuse zinc emissions from Germany into the river basin districts and seas in 1983-1987, 1993-1997, 1998-2002 and 2003-2005

Zink	Point emissions in t/a				Diffuse emissions in t/a			
Period	83-87	93-97	98-02	03-05	83-87	93-97	98-02	03-05
Danube	168.0	133.2	104.1	93.4	466.3	357.8	379.2	278.7
Rhine	1039.7	536.9	396.7	321.9	1065.7	795.8	802.9	625.0
Meuse	45.9	21.2	13.0	12.1	40.5	30.6	30.6	27.1
Ems	26.7	19.1	13.4	12.2	88.1	77.9	76.3	66.9
Weser	1573.5	106.6	58.4	44.8	348.9	296.3	295.1	252.4
Elbe	1318.5	449.4	406.6	427.5	717.0	523.2	492.4	441.5
Odra	3.1	7.8	10.7	19.1	108.5	50.3	40.1	32.3
Eider	2.2	0.8	0.8	0.8	32.0	22.6	25.4	20.4
Schlei/Trave	2.4	1.9	2.1	2.0	50.9	39.2	38.5	33.0
Warnow/Peene	4.4	3.2	2.2	2.0	90.9	54.6	50.0	42.3
North Sea	4006.5	1134.0	888.9	819.4	2292.2	1746.4	1722.7	1433.2
Baltic Sea	9.9	12.9	15.0	23.1	250.4	144.1	128.6	107.6
Black Sea	168.0	133.2	104.1	93.4	466.3	357.8	379.2	278.7
Germany	4184.4	1280.0	1008.0	935.9	3008.9	2248.3	2230.4	1819.6

### 2003-2005

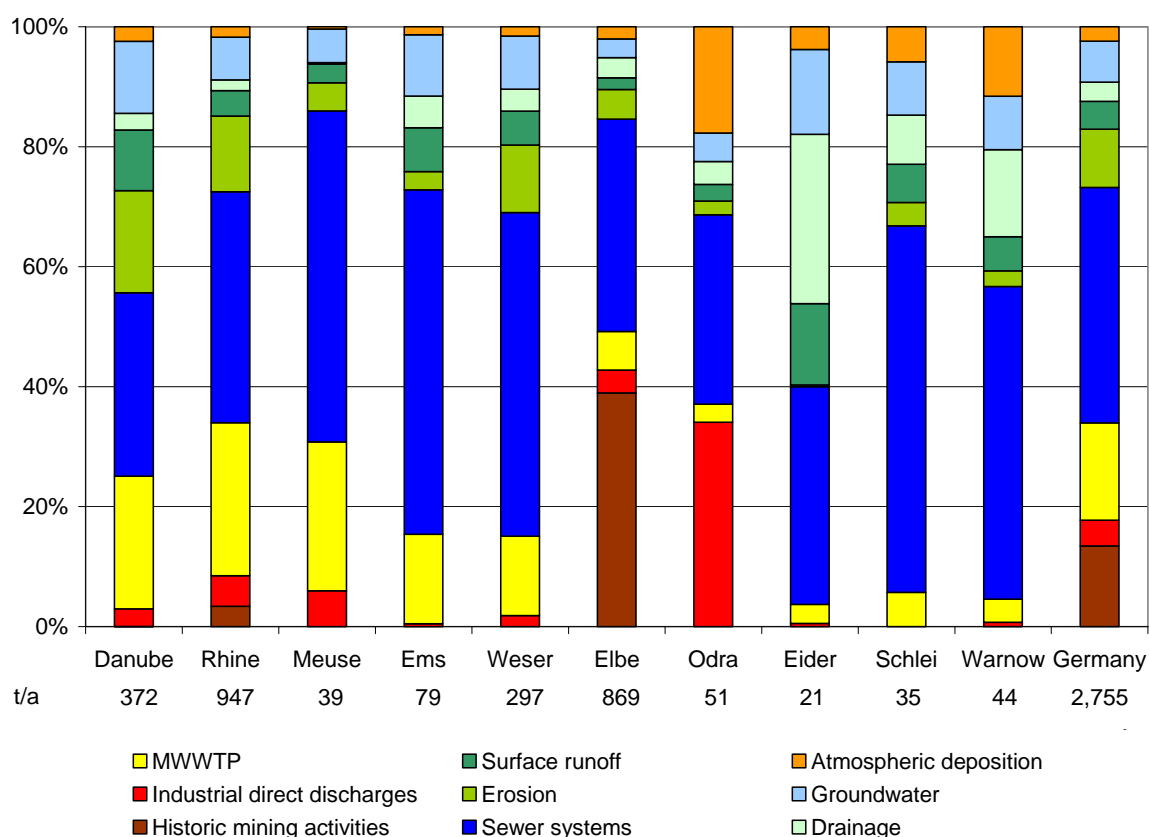


Figure 80: Relative importance of the pathways for zinc emissions into the river basin districts of Germany in the balance period 2003-2005

The highest emissions of Cu, Hg and Zn are from urban areas (sewer systems and municipal wastewater treatment plants). Main focus points are urban agglomerations such as the Ruhr area as well as Hamburg, Berlin, Munich and Stuttgart. The area-specific emissions from sewer systems are higher in the north of Germany due to the predominant use of separate sewer systems (comp. paragraph 3.3.9.1). In addition, Zn emissions from historic mining activities are important in the Elbe basin, analogue to Cd.

Groundwater inflow is the most important pathway for Ni. Due to the low groundwater recharge rate in the east of Germany the area-specific emissions are lower in those areas.

Emissions for Pb are mainly caused by erosion and sewer systems. High area-specific emissions are found in the Alps and Mittelgebirge (low mountain ranges) regions as well as in urban agglomerations.

### **5.3.3 Comparison of the observed and modelled river loads**

The plausibility of the total emissions can be verified with the help of river loads that were balanced from quality and discharge readings (comp. paragraph 3.2). Quality data on heavy metals was collected from the year 1998 onwards by the relevant authorities. Depending on the metal between 18540 (Hg) and 36332 (Cu) individual values were reported. First the annual loads were modelled and then the averages for the balance periods 1998-2002 and 2003-2005 were calculated. At the time of the requests the federal states could only deliver data up until 2004, therefore in average only two years could be considered for the current period. Hence the data for this period is much more uncertain than for the period 1998-2002 which was calculated as average of five years. To check the plausibility all monitoring stations were used that had balanced river loads from both periods. In Figure 81 to Figure 87 the comparison between observed and modelled river loads at the quality monitoring stations is depicted for both periods. The amount of considered monitoring stations changes between 121 (Cd) and 219 (Cu) as not from all monitoring stations data for all metals was available. The statistical quality parameters considered are the systematic deviation (BIAS), the root mean squared error (RMSE) as well as the model efficiency coefficient according to Nash-Sutcliffe (NASH. & SUTCLIFFE, 1970) (EF). Furthermore, the average observed and modelled annual river load was defined for the considered monitoring stations.

For the comparison of the modelled river loads with the observed river loads at the quality monitoring stations the retention within the waterbody was estimated. With a power function (Equation 5-1) based on the specific runoff of the sub-basins the heavy metal retention was estimated by adjusting the empirical factors a and b (FUCHS ET AL., 2002). The condition for the adjustment was that the model efficiency according to Nash-Sutcliffe is to be as high as possible (EF=1). This adjustment was made for the balance period 1998-2002 as there were more annual loads available. Subsequently, the factors were transferred to the period 2003-2005 so that both periods were comparable regarding the statistical quality parameters.



Equation 5-1 
$$L = \frac{1}{1 + R_L} \cdot E \quad \text{mit} \quad R_L = a \cdot q^b$$

L	modelled river load (emission – retention) [kg/a]
E	emission [kg/a]
R <sub>L</sub>	load weighted retention [-]
q	specific runoff [l/(km <sup>2</sup> ·s)]
a,b	empirical factors

Figure 81 to Figure 87 depict the comparison between the observed and modelled river loads at the quality monitoring stations for the periods 1998-2002 and 2003-2005. Apart from the 1:1-line the range of the deviation  $\pm 50\%$  was illustrated. The data for all metals is spread beyond the deviation range of  $\pm 50\%$ . The reasons for the spread between observed and modelled river loads are amongst others the uncertainties in the quality data that is used to balance the observed river loads. Some of the measured concentrations are below the limit of quantification. In these cases, analogue to the input data for the emission calculations, half of the value of the limit of quantification was used. Compared to the other metals, the model efficiency for Hg with an EF value of 0.25 (comp. Figure 84) is low. This can be lead back to the high share of reported concentrations below the limit of quantification of 73 % (referring to all reported individual values). For Cd, Cr and Pb 57 %, 55 % resp. 47 % of the reported measurement readings are below the limit of quantification. The loads for Cu, Ni and Zn have a share of 13 %, 15 % resp. 22 % of measurement readings below the limit of quantification and can be seen as reliable.

It can generally be said that the deviations between observed and modelled river load for smaller catchment areas are increasing. The heavy metal emissions quantification had to be made with the help of average emission factors due to the available data base and the scale of the large river basins that had to be covered. This procedure does not allow an adequate consideration of characteristics of smaller river basins such as increased geogenously caused heavy metal contents and regional and local contamination hotspots due to industrial, agricultural and urban use. Especially emissions from historic mining activities must be mentioned in this context. This project was able to record numerous locations but for some of them there was no data on the emissions. For example this applies to the abandoned mining in the Harz which has no data on emissions (comp. paragraph 4.2.1.3). Accordingly the Cd, Pb and Zn emissions into the Weser tributaries (e.g. Innerste, Oker, Leine) and in the following in the Aller and Weser are severely underestimated. Large differences were also found in the river basin of the Elbe for the catchment areas of the Mulde and Saale that show high geogenous heavy metal concentrations due to the Erzgebirge and Thuringian Slate Mountains. And it also has to be assumed that not all emissions from abandoned mining locations in eastern Germany were covered. The deviations are especially visible for Zn in the Triebisch which drains pit waters from the abandoned mining area in Freiberg (comp. Figure 87).

Erosion plays an important role for Cr and Pb (comp. paragraph 5.3.2). Erosion events are especially caused by seldom and often local heavy rainfall. Due to the existing scaling level of the large river basins and the temporal resolution of one year the emissions from this pathway cannot be quantified reliably in small catchment areas.

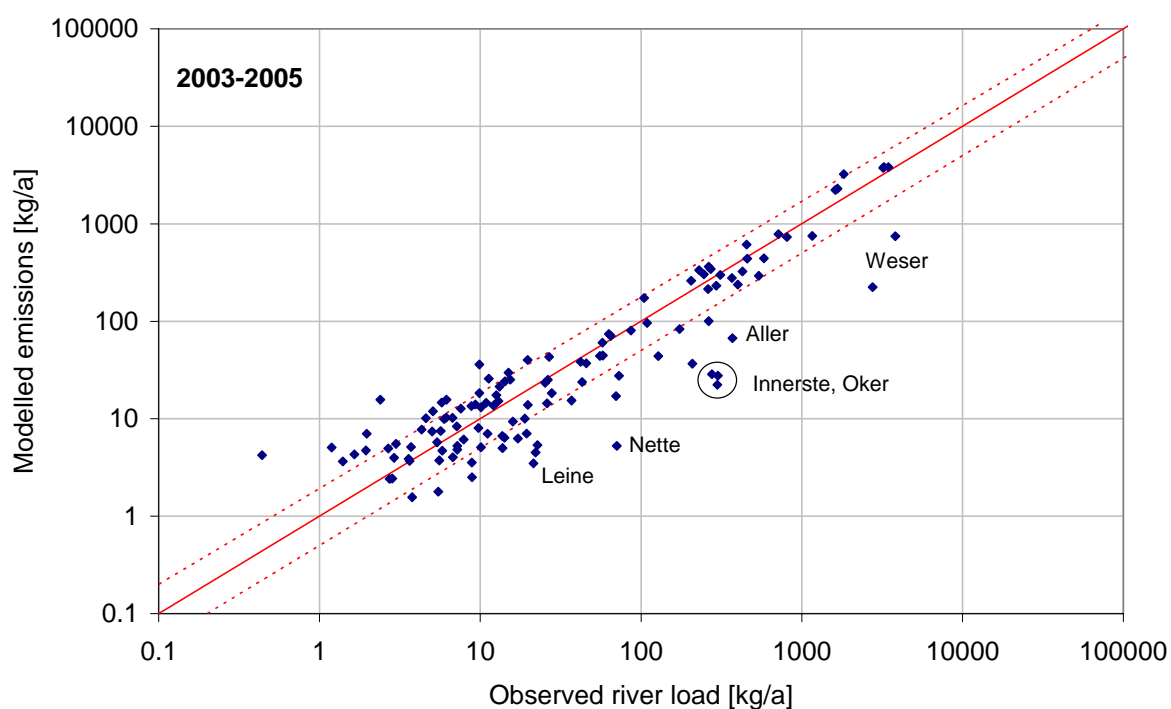
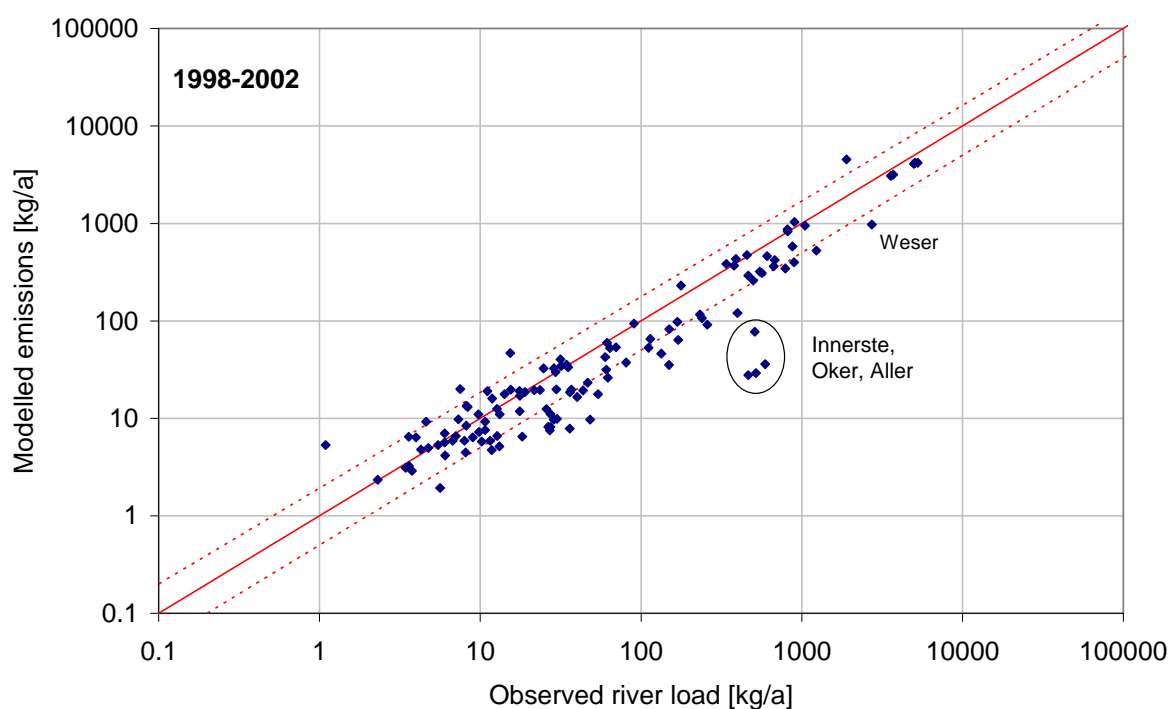
Some river basins show modelled emissions that are much higher than the observed river load. One reason for this is for example that the quality monitoring station is situated directly behind a sea or a barrage. The retention rate of the water in these catchments is often underestimated using the relations derived from flowing waters. Furthermore, river systems with canals and weirs (e.g. Spree, Grosse Roder) cannot be represented with the underlying approaches.

For the balance period 1998-2002 very high model efficiency coefficients between  $EF = 0.77$  (Pb) and  $EF = 0.94$  (Ni, Cr) were reached, with the exception of Hg. In the period of 2003-2005 the model efficiency is much lower with values between  $EF = 0.44$  (Pb) and  $EF = 0.89$  (Cu). In this period there was a lot less data (2-3 years) available for the emission calculations as well as for the balancing of observed river loads. Errors that arise from input data that is not adequately exact concerning the spatial and temporal resolution as well as from the underlying simplified model approaches are generally averaged for longer observation periods (and larger catchment areas) so that in this case better results can be achieved.

Regarding the systematic deviation (BIAS) it can be seen that the observed river loads compared to the modelled river loads were slightly underestimated for the period 1998-2002 while in the period 2003-2005 some were significantly overestimated, resp. in the case of Cd and Pb not quite as badly underestimated. Comparing both periods the observed river load decreased between 21 % (Pb) and 39 % (Cr), mainly due to the hydrological differences. This strong decline could not be represented by the model to the same extent for the observed monitoring stations. But it must be considered that the heavy metal retention in the waterbodies can only be estimated on the basis of the data from 1998-2002 and was taken as constant for both periods. A detailed, non-constant illustration of the retention processes within the waterbodies is needed as well as an observation of the complete five-year cycle from 2003-2007 for a final validation of the current period.

For Cd and Hg no retention within the waterbodies was considered because the quantified emissions have already been underestimated in a direct comparison with the observed river load (comp. BIAS in Figure 81 and Figure 84). For Cd the reasons can be mainly seen in underestimated emissions from former mining activities (comp. Figure 81). For Hg the data is very uncertain for the emission calculations as well as for observed river load. But the comparison (Figure 84) shows that the quantified emissions for Hg are within the right dimension. In average retention within the waterbodies for the whole investigation area was calculated as being 56 % resp. 39 % especially for Cr und Pb which are both strongly affected by erosion. For Ni the average retention is at 51 %. For Cu and Zn the retention is much lower at 13 % resp. 10 % which is plausible as these metals mainly come from urban point and diffuse sources (municipal wastewater treatment plants, storm sewers, combined sewer overflows etc.) and are therefore transported mainly as dissolved or very fine particulate matter.

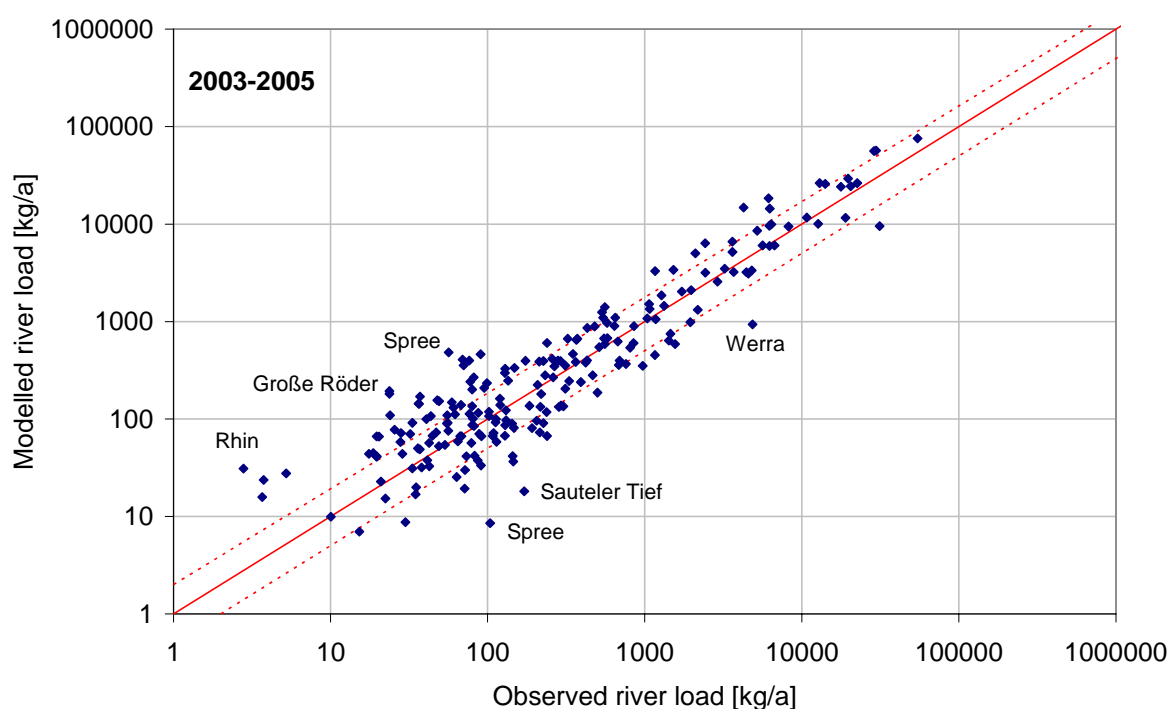
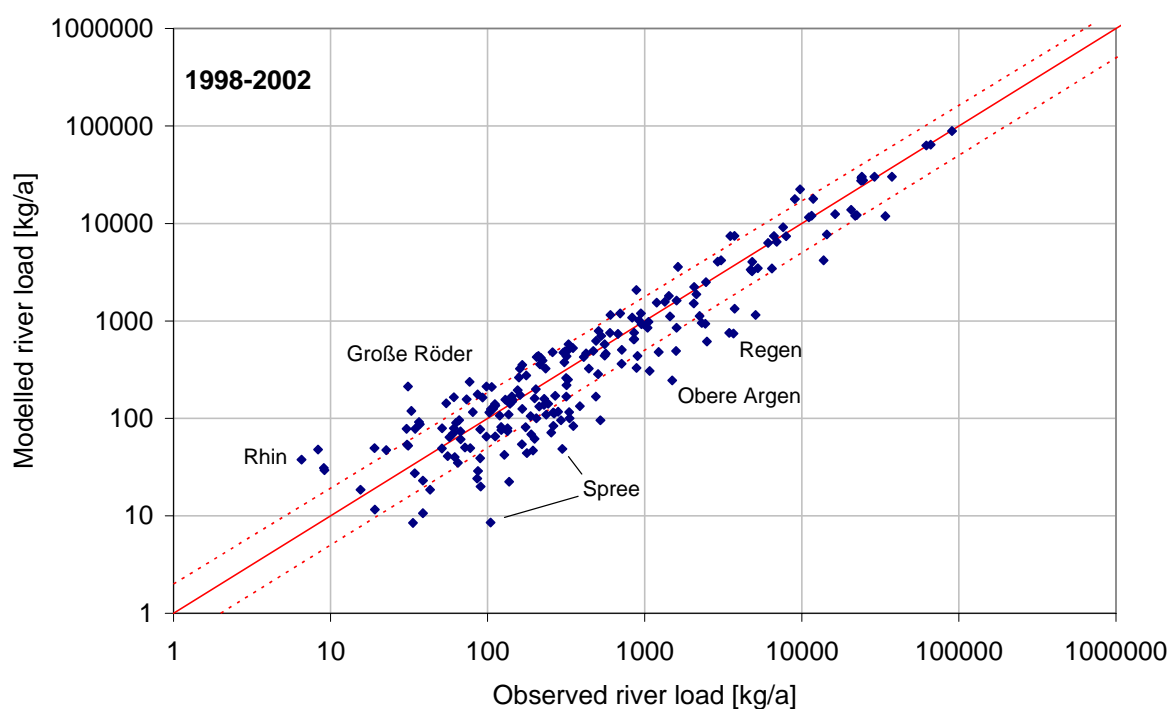
## Cadmium



Period	Number gauges	Mean, observed	Mean, modelled	BIAS	RMSE	Nash-Sutcliffe
1998-2002	121	381.1 kg	301.4 kg	- 21.3 %	335.0 kg	EF = 0.88
2003-2005	121	275.5 kg	242.3 kg	- 12.0 %	408.1 kg	EF = 0.71

Figure 81: Comparison of modelled emissions and observed river loads for the periods 1998-2002 und 2003-2005 for cadmium

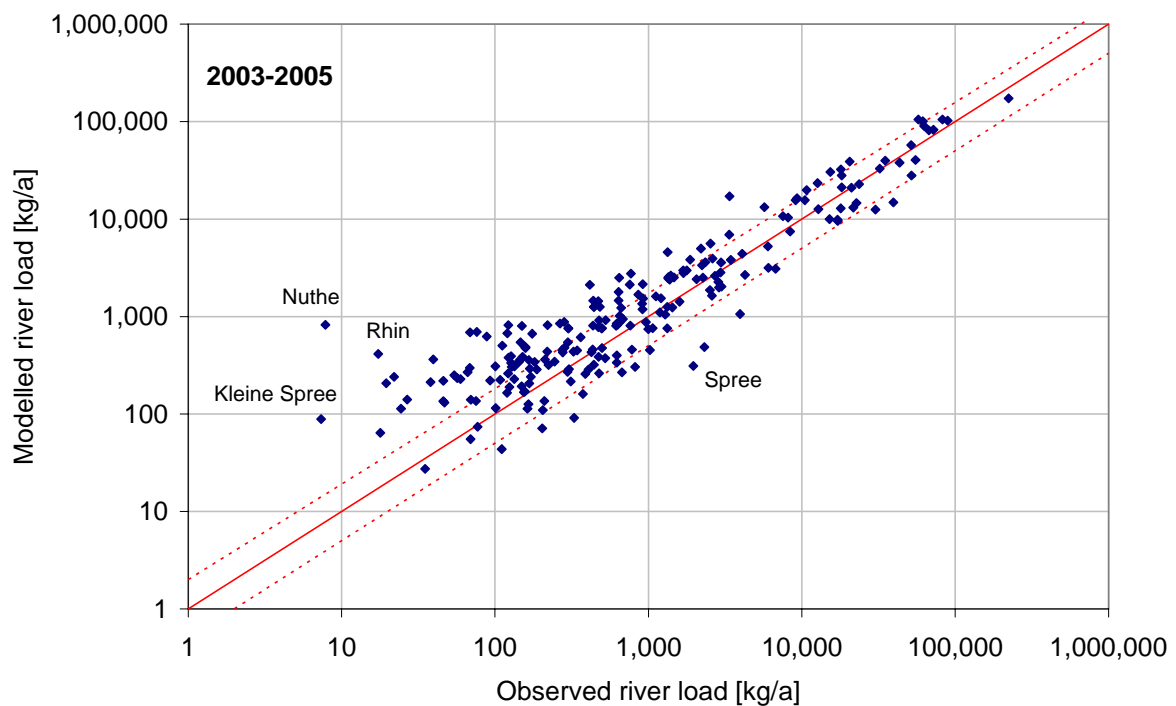
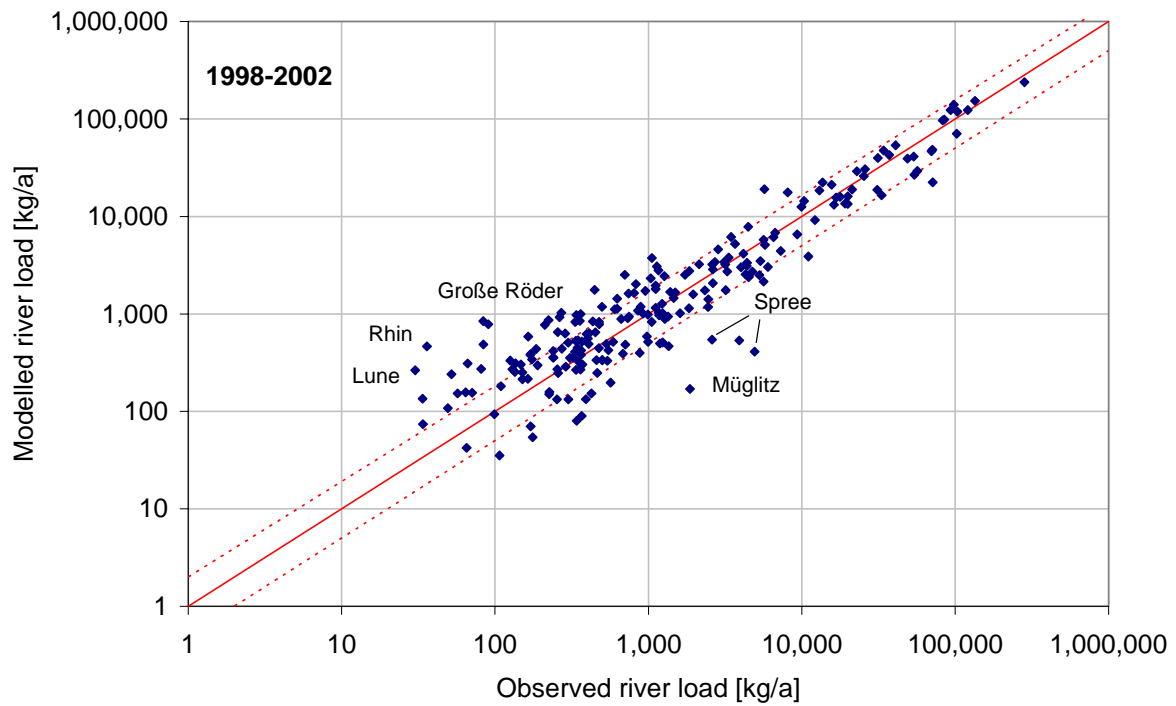
## Chromium



Period	Number gauges	Mean, observed	Mean, modelled	BIAS	RMSE	Nash-Sutcliffe
1998-2002	211	3,514 kg	3,284 kg	- 6.6 %	2,599 kg	EF = 0.94
2003-2005	211	2,141 kg	2,833 kg	+ 32.3 %	4,006 kg	EF = 0.62

Figure 82: Comparison of the observed and modelled river loads for the periods 1998-2002 und 2003-2005 for chromium

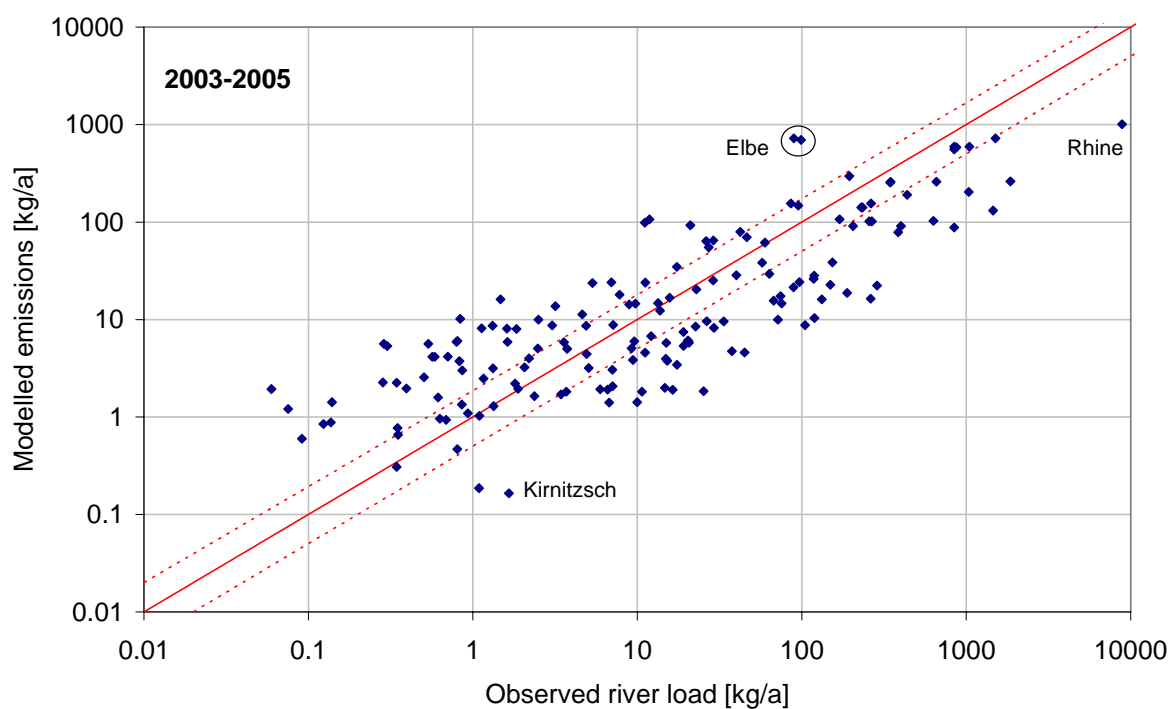
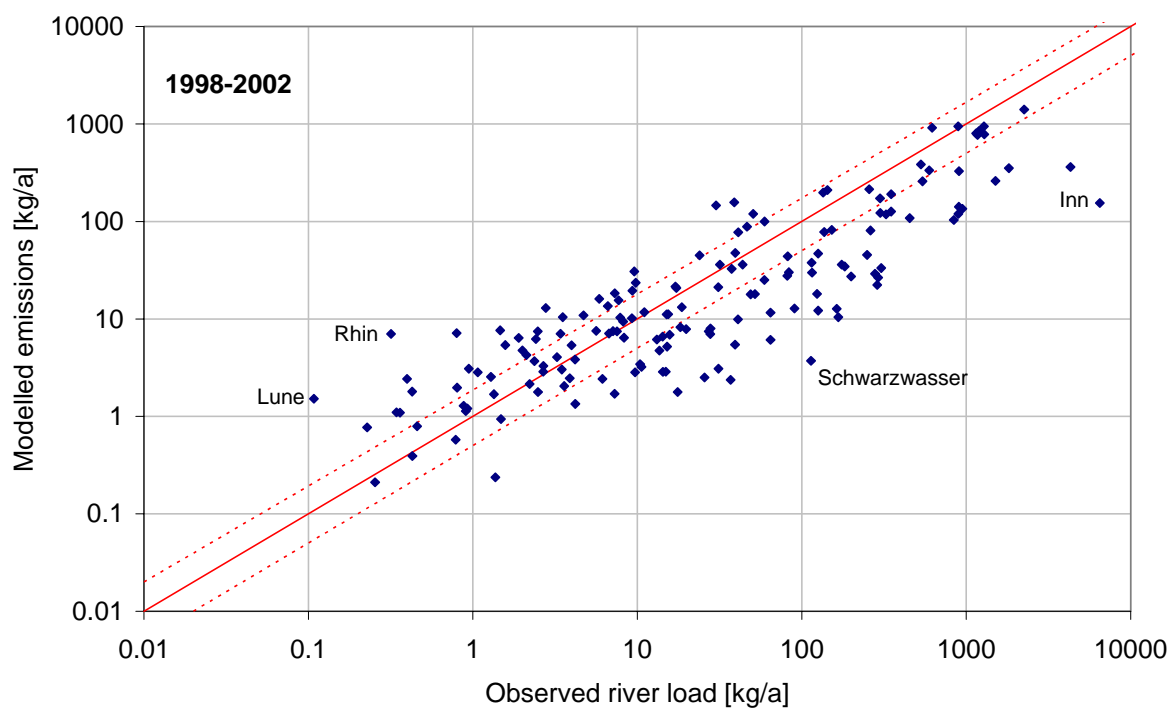
## Copper



Period	Number gauges	Mean, observed	Mean, modelled	BIAS	RMSE	Nash-Sutcliffe
1998-2002	219	10,389 kg	10,030 kg	- 3.5 %	7,869 kg	EF = 0.93
2003-2005	219	7,085 kg	7,875 kg	+ 11.2 %	7,326 kg	EF = 0.89

Figure 83: Comparison of the observed and modelled river loads for the periods 1998-2002 und 2003-2005 for copper

## Mercury

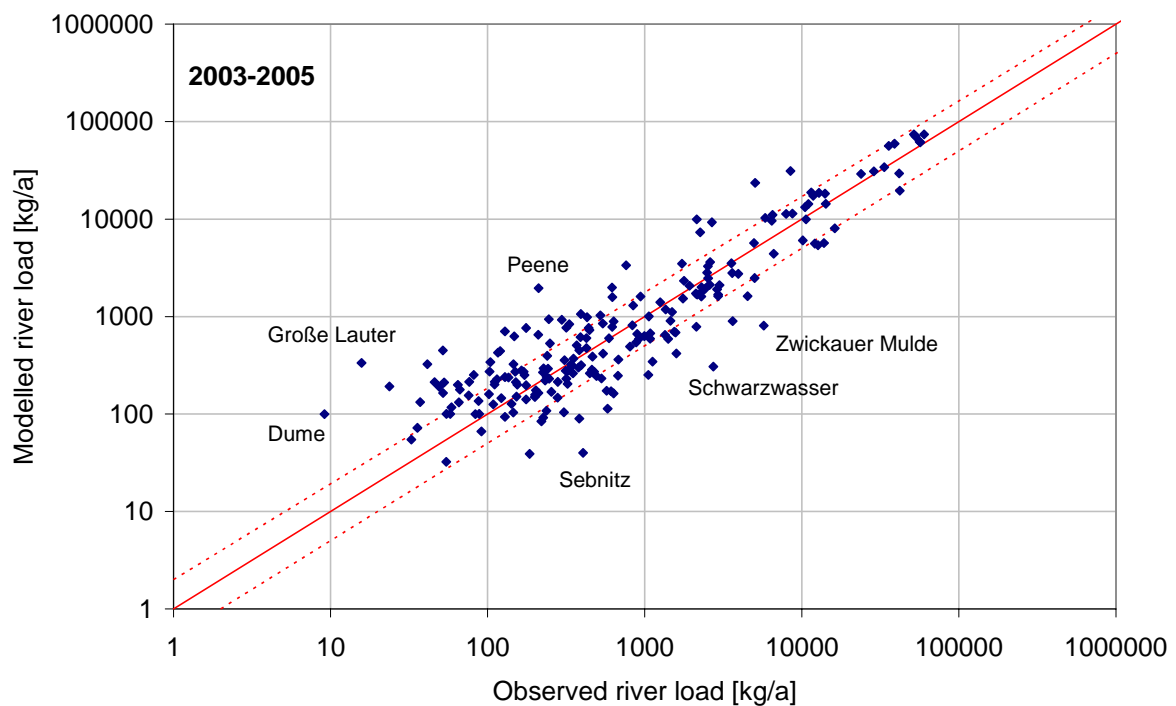
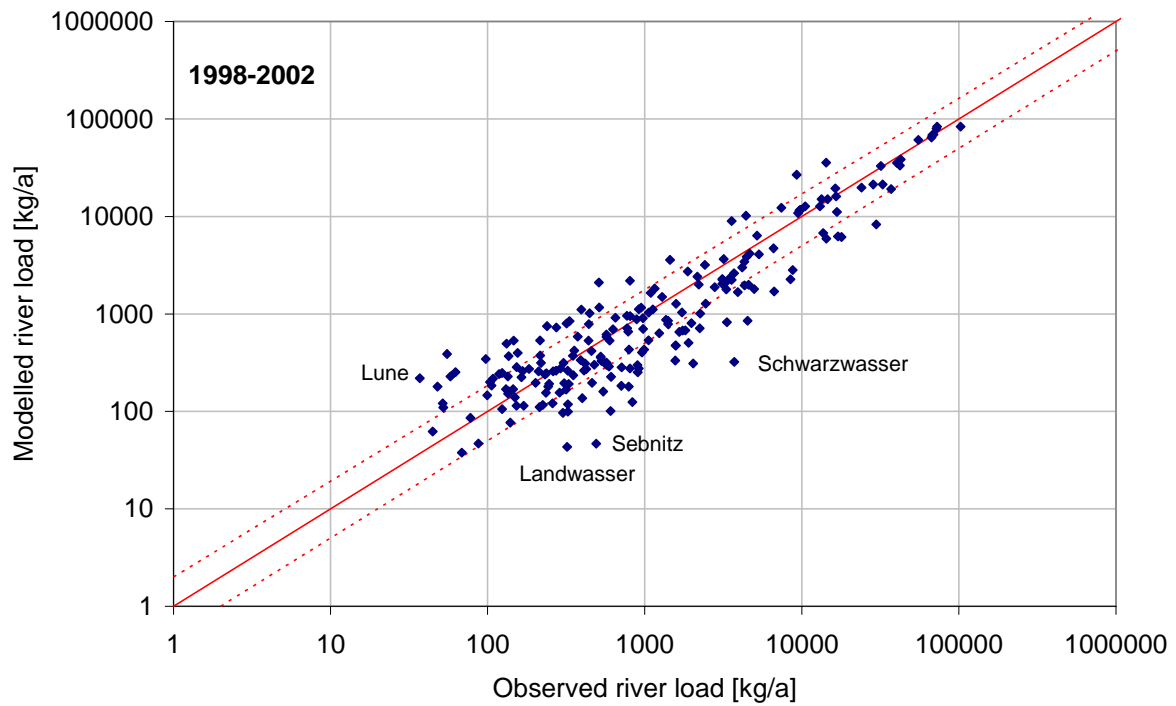


Period	Number gauges	Mean, observed	Mean, modelled	BIAS	RMSE	Nash-Sutcliffe
1998-2002	158	241.6 kg	89.4 kg	- 63.0 %	640.6 kg	EF = 0.25
2003-2005	158	176.4 kg	66.8 kg	- 62.1 %	667.3 kg	EF = 0.25

Figure 84: Comparison of modelled emissions and observed river loads for the periods 1998-2002 und 2003-2005 for mercury



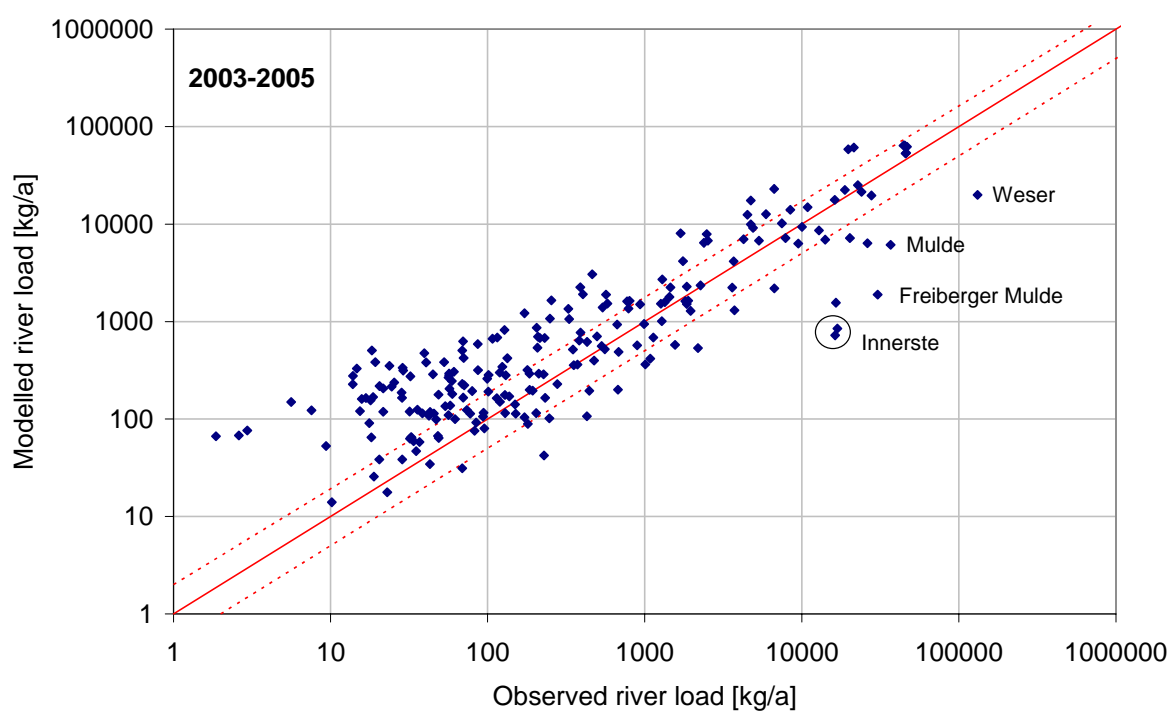
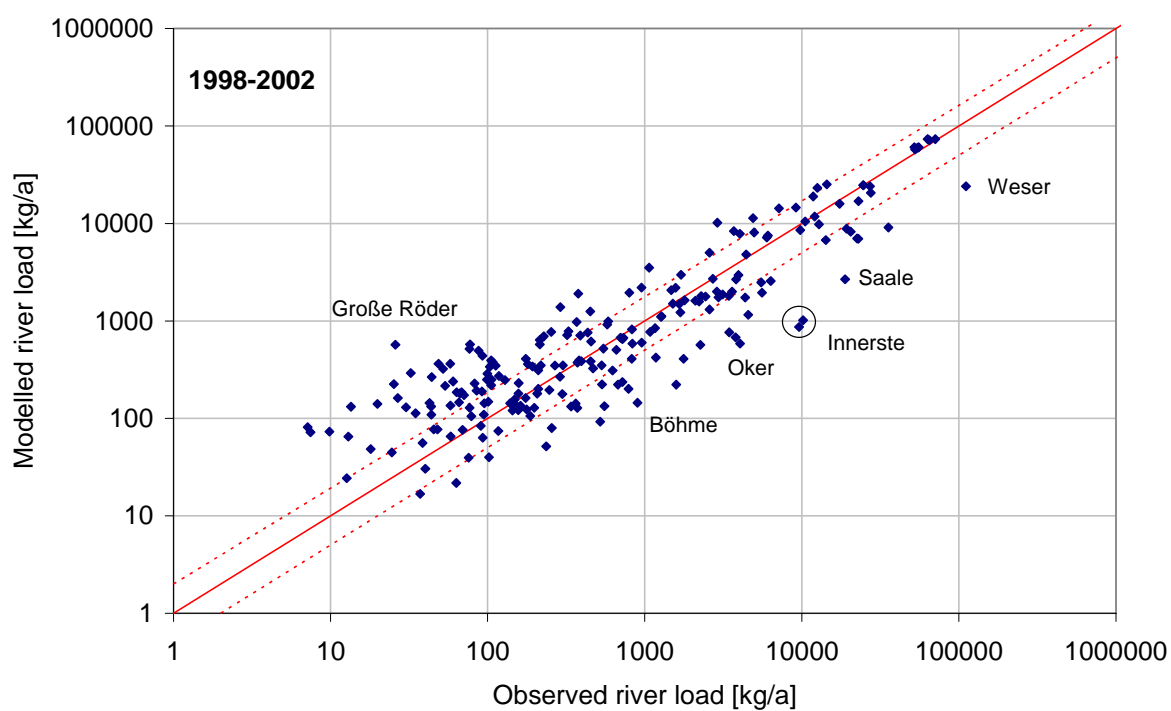
## Nickel



Period	Number gauges	Mean, observed	Mean, modelled	BIAS	RMSE	Nash-Sutcliffe
1998-2002	211	5,984 kg	5,422 kg	- 9.4 %	3,885 kg	EF = 0.94
2003-2005	211	4,228 kg	4,852 kg	+ 14.8 %	4,421 kg	EF = 0.85

Figure 85: Comparison of the observed and modelled river loads for the periods 1998-2002 und 2003-2005 for nickel

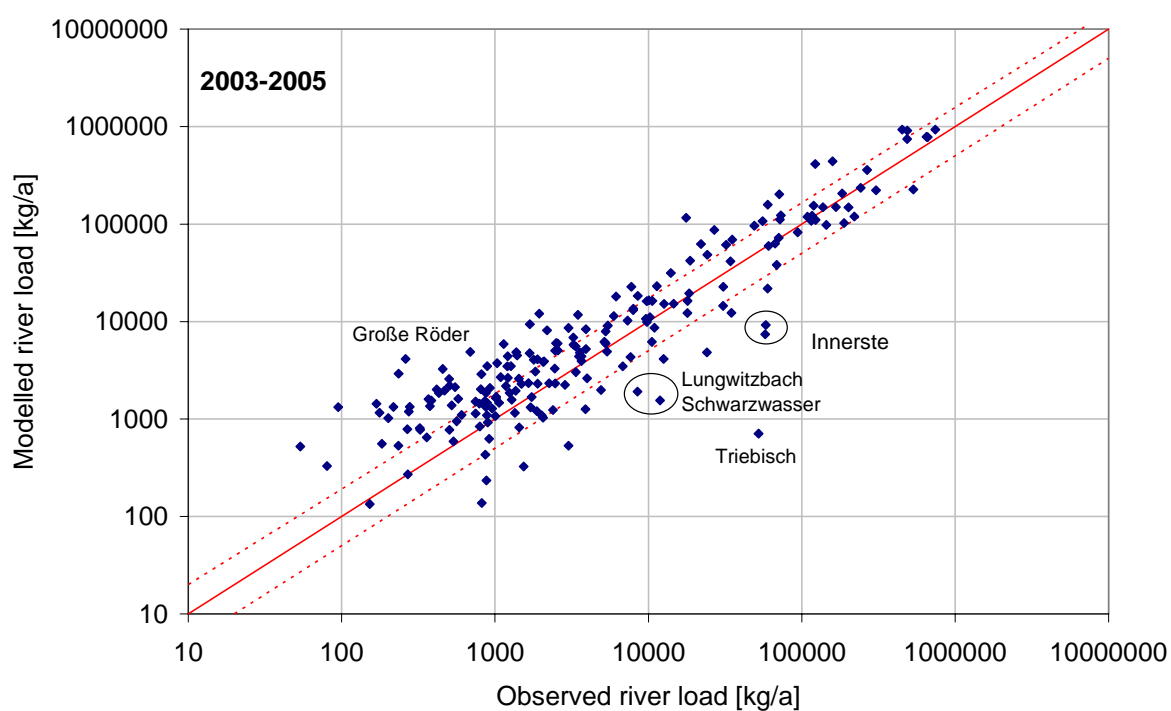
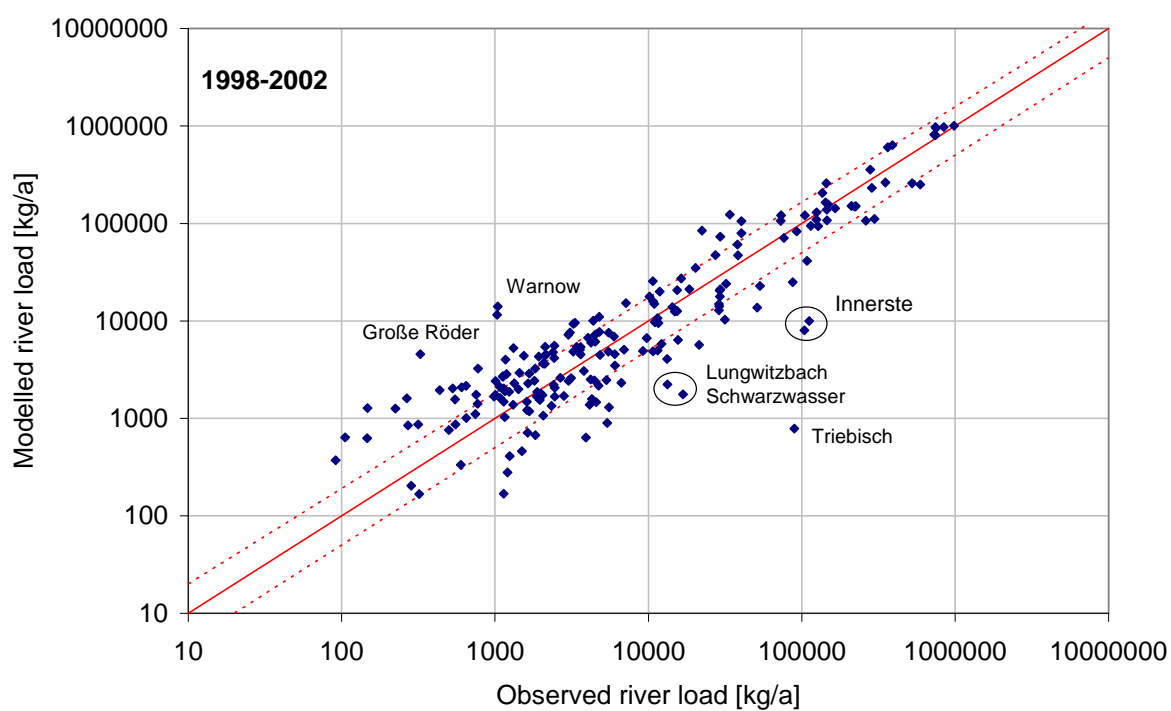
## Lead



Period	Number gauges	Mean, observed	Mean, modelled	BIAS	RMSE	Nash-Sutcliffe
1998-2002	212	5,260 kg	4,574 kg	- 13.0 %	7,042 kg	EF = 0.77
2003-2005	212	4,140 kg	4,041 kg	- 0.9 %	9,865 kg	EF = 0.44

Figure 86: Comparison of the observed and modelled river loads for the periods 1998-2002 und 2003-2005 for lead

## Zinc



Period	Number gauges	Mean, observed	Mean, modelled	BIAS	RMSE	Nash-Sutcliffe
1998-2002	202	61,783 kg	61,759 kg	- 0.04 %	54,682 kg	EF = 0.89
2003-2005	202	43,156 kg	54,459 kg	+ 26.2 %	67,136 kg	EF = 0.70

Figure 87: Comparison of the observed and modelled river loads for the periods 1998-2002 und 2003-2005 for zinc

## 5.4 Polycyclic aromatic hydrocarbons (PAH)

### 5.4.1 Pathways

The description for the individual pathways clearly shows the existing uncertainties in quantifying the PAH emissions discharged into surface waters. This especially concerns the groundwater inflow for which there is only a small amount of monitoring data above the limit of quantification, as well as for the erosion pathway for which there has been no research on possible accumulation of PAH due to the preferred transport of fine soil particles. For the pathway sewer systems it has to be considered that the available data from urban areas has a wide fluctuation range and in addition measurements are often only available for traffic areas. Furthermore, the analytical problems that accompany the substance group of PAH have to be considered: there are a large number of different individual substances so they are hard to compare as the types of analysis differ, too. The emission ratios between the individual PAH substances can vary spatially and temporally due to the dependence on the emission sources. A conversion of individual substances to the PAH sum is therefore additionally affected by errors. Nevertheless, a first assessment of the relevance of the individual pathways can be made on the basis of available data (comp. Figure 88 and Table 61).

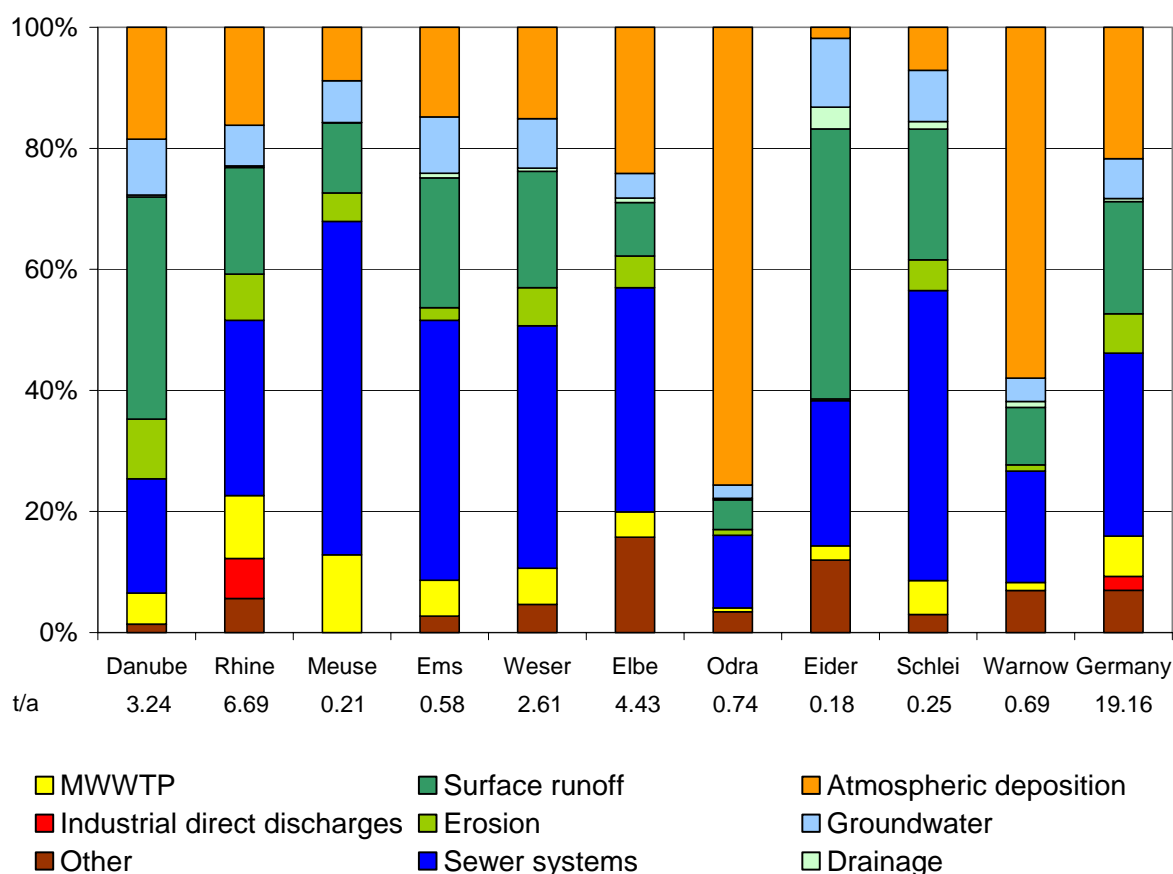


Figure 88: Relative relevance of the individual pathways for the  $\Sigma \text{PAH}_{16}$ - emissions into surface waters in the river basin district of Germany for the balance period 2003-2005

Even though there is still research needed for some of the pathways (e.g. inland navigation, steel construction for hydraulic engineering, erosion) the overall view with the help of the resulting relevance distribution shows clearly that it can be assumed that diffuse atmospheric emissions have a dominating influence on the total emissions of PAH in waterbodies. Apart from direct deposition onto water surfaces (pathway „deposition“) the atmospheric deposition also indirectly essentially contributes to the emissions into waterbodies from urban areas as well as from erosion and surface runoff. In addition it can be assumed that the emissions from wastewater treatment plant effluents are strongly influenced by atmospheric deposition onto areas in the catchment area of the wastewater treatment plants. Therefore more than 80 % of PAH emissions into waterbodies can be classified as „influenced by atmospheric deposition“.

Table 61: Modelled annual  $\Sigma$  EPA-PAH<sub>16</sub>-emissions into waterbodies

Pathway	$\Sigma$ EPA-PAH <sub>16</sub> [kg/a]
MWWTP	1,269
Industrial direct discharges	443
Inland navigation	1,341
Atmospheric deposition	4,165
Erosion	1,248
Surface runoff	3,556
Drainage	93
Groundwater inflow	1,256
Sewer systems	5,794
<b>Total</b>	<b>19,164</b>

#### 5.4.1.1 Verification of the emissions via wastewater treatment plant

According to paragraph 4.3.1.1 assuming that the treatment efficiency in WWTP for PAH is > 90 % (IVASHECHKIN, 2005) an annual inflow load of 12.9 t/a  $\Sigma$  EPA-PAH<sub>16</sub> to the WWTP in Germany can be calculated with the help of sewage sludge concentrations. If the amount of treated wastewater in Germany in 2004 is said to be 9,410 million m<sup>3</sup> annually (DESTATIS, 2004) this leads to an average inflow concentration in wastewater treatment plants of 1.37 µg/l  $\Sigma$  EPA-PAH<sub>16</sub>. For a large-scale investigation it is therefore a good match to the average dominating concentration range of stormwater runoff according to WELKER (2004) of approx. 1.3 µg/l (comp. paragraph 4.3.2.7). But it has to be considered that the inflow loads into wastewater treatment plants are influenced by the components „domestic wastewater“, „commercial wastewater“ as well as „sewer infiltration water“. In practise strong regional differences will be encountered (e.g. depending on the share of separate and combined sewer system). A validation of the inflow concentrations of the wastewater treatment plants in Germany, calculated from sewage sludge concentrations and efficiencies of the wastewater treatment, the known loads for the most important components in combined sewers are put in contrast and compared.

## **Wastewater**

In Germany in the year 2004 an annual wastewater amount of 5,271 million cubic metres was discharged into the public sewer system. The main part, 98.7% (5,204 million cubic metres), was treated in public wastewater treatment plants (DESTATIS, 2004). For domestic wastewater (e.g. from human excrements, washing processes and detergents, etc.) there is hardly any data for a large-scale investigation available. BEIER (2008) detected 0.6 to 1 µg/l  $\Sigma$  EPA-PAH<sub>16</sub> in sewage. Therefore, for a first assessment the average PAH concentration in wastewater due to the anthropogenic use was estimated to be 0.8 µg/l. Based on this value an annual load of 2 t/a  $\Sigma$  EPA-PAH<sub>16</sub> reaches the inflow of the wastewater treatment plants. In the year 2004 according to EPER an additional 3.4 t/a  $\Sigma$  EPA-PAH<sub>16</sub> from indirect industrial dischargers (comp. paragraph 4.2.1.2) are to be considered. This leads to a calculated total input load of 7.7 t/a.

## **Sewer infiltration water and stormwater runoff**

Apart from wastewater a total of 4,205.7 million m<sup>3</sup> of sewer infiltration water and stormwater were treated in wastewater treatment plants in the year 2004 (DESTATIS, 2004). 2,393.7 million m<sup>3</sup> are allotted to precipitation which leads to an input load into the wastewater treatment plants of 3.1 t/a at an average  $\Sigma$  EPA-PAH<sub>16</sub> concentration (paragraph 4.3.2.70) of 1.3 µg/l (WELKER, 2004). The remaining 1,812 million m<sup>3</sup>/a are sewer infiltration water. According to DIN 4045 (DIN, 2003) the sewer infiltration water is defined as „groundwater infiltration due to leakages in the sewer system, illegally discharged water via faulty connections as well as stormwater runoff into wastewater sewers e.g. via manhole covers“. Due to the many possible sources for sewer infiltration water there is no measuring data that can be used over a large area concerning PAH. In addition, the information on the share of the different components in the sewer infiltration water (groundwater infiltration, scheduled and unscheduled discharge of stream, spring, drainage and surface water) can currently only be estimated (DOHMANN, 2008). Considering the partly calculated and partly estimated share of stormwater, stream and drainage water the share of infiltrated groundwater in sewer infiltration water is approx. 55 % (DOHMANN, 2008). For a first assessment for this share a PAH groundwater concentration is assumed to be 0.02 µg/L (comp. paragraph 4.3.2.7). For the remaining 45 % a concentration of 3.5 µg/l according to IWG (2008) is assumed as the sources are comparable especially for unscheduled discharged water via faulty connections and via manhole covers. This leads to a calculated inflow load of 2.9 t/a in wastewater treatment plants from sewer infiltration water.

## **Comparison of the PAH loads in the inflow of wastewater treatment plants**

On the basis of the annual loads, calculated with the help of the water amounts and PAH concentrations, of 7.7 t in wastewater, 3.1 t in stormwater as well as 2.9 t in sewer infiltration water the calculated inflow load of wastewater treatment plants is 13.7 t/a. Therefore, this matches the annual  $\Sigma$  EPA-PAH<sub>16</sub> input loads of 12.9 t in the inflow of wastewater treatment plants in Germany which were calculated with the help of sewage sludge concentrations (comp. paragraph 4.3.1.1). Due to the regionally and temporally widely varying input parameters (e.g. sewer infiltration water) it can be counted with a considerable fluctuation range in



practise. Based on two completely independent ways to calculate the PAH inflow loads of wastewater treatment plants the comparative value can be seen as an affirmation of the general dimension of the emissions.

#### **5.4.2 Comparison of the observed and modelled PAH river loads**

On the basis of the collected data and of an assessment of the PAH-retention within the waterbodies a comparison of observed and modelled river loads can be made. With the help of this comparison a plausibility check can be done for the result. Figure 89 shows the correlation between modelled river load (emission - retention) and the observed river load (calculated from the monitoring stations data). Additionally the 50% deviation for the 1:1 line is depicted as dotted line. The trend at quality monitoring stations with large PAH river loads shows significantly lower deviations from the 1:1 line than monitoring stations with lower loads.

Correspondingly, the PAH emissions can be calculated as far as possible with a deviation of less than 50 %, especially for the large river basins like e.g. the Rhine and Elbe. For smaller observed river loads and small river basins the modelled loads are normally much higher than the ones observed. One possible cause for this on the one hand are the demanding analytics for PAH. PAH measuring data from monitoring stations is therefore not available in the same quality (temporal resolution, amount of recorded PAH compounds) and quantity (number of monitoring points) for all river basins. If the modelled river loads are significantly higher than the observed river loads then often the location of the quality monitoring points is below seas or barrages that have a special influence on the retention.

In addition further uncertainties have to be considered for PAH. This provides the necessity to partially assume average emission factors for emission calculations on the basis of the available data and the scale that has to be covered for the river basins in Germany. Regional or local characteristics often can't be illustrated completely. Spatially and temporally variable characteristics (meteorology, type of landscape, land use) can also not be adequately detected for large-scale observations.

Further uncertainties for the emission estimations are traced back to substance-specific characteristics of PAH. Due to the multitude of possible compounds measurements in general have to be confined to the toxicologically or quantitatively most important representatives of that group of elements of 16 ( $\Sigma$  EPA-PAH<sub>16</sub>) or less compounds. But even within this choice uncertainties arise for further evaluations due to the different physico-chemical characteristics. Apart from the different water solubilities and varying sorption characteristics the different possible degradation processes have to be mentioned in this context. The potential degradation of organic compounds additionally enhances the effect also proven for other substances (e.g. heavy metals) that lower observed river loads are detected compared to the modelled river loads. In these cases the retention within the waterbody in the affected river basins is underestimated with the used retention correlations.

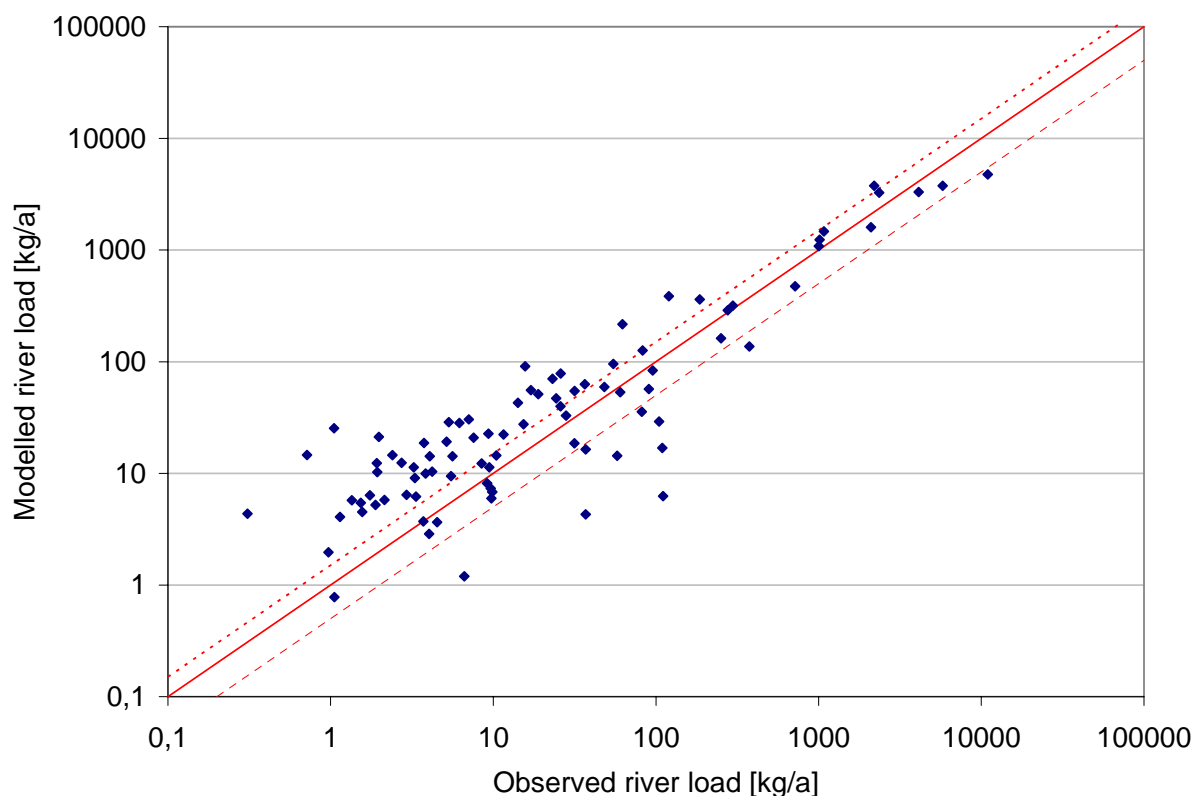


Figure 89: Comparison of the observed and modelled  $\Sigma$  EPA-PAH<sub>16</sub> river loads in the balance period 2003 – 2005

The mentioned causes for potential uncertainties show that more research and development is needed for PAH. Even though large-scale trends emerged from the available data, further steps should be strived for improving the results, especially for average and smaller river basins. To optimise this, further research on the emissions as well as on the waterbodies is needed with a higher spatial and temporal resolution. In addition, due to the special characteristics of organic compounds, a further examination of the retention under special consideration of the sorption and degradation processes is necessary.

### 5.4.3 The relevance of PAH in the protection of waterbodies

According to BMU (2006) the PAH compounds exceeded in the period of 2002-2004 at approx. 10 to 25 % of the sampled monitoring stations of German waterbodies the environmental quality standards for the ecological and chemical state according to EU-WFD. According to a survey in the federal states on the water pollution by PAH in Germany, several PAH representatives as well as PAH as sum parameter were classified as „relevant“ in the overall assessment (LEHMANN & VIETORIS, 2006). In order to reduce the exceeding of environment quality standards in waterbodies urgent action is needed especially for diffuse atmospheric emissions due to their high relevance.

#### 5.4.4 Initial solution

The current emission balancing concerning the sources of atmospheric PAH emissions implies a large relevance for PAH emissions due to complex matter transfer processes for waterbodies especially from older, inefficient or badly regulated small wood domestic combustion plants. Due to the prospective price development for oil and gas and the promotion of CO<sub>2</sub>-neutral fuels a further strong increase in wood domestic combustion plants is to be assumed.

A first rough estimation of the further emission development for small wood domestic combustion plants can be made on the basis of scenarios of the German Federal Environment Agency (UBA, 2007b) and the assumption of a proportional development of the PAH emissions.

According to a prognosis relating to the amendment to the act 1st Ordinance for the Implementation of the Federal Immission Control Act (BimSchV) without any stricter requirements (comp. Figure 90, „no amendment“) for small wood domestic combustion plants till the year 2025 the particulate matter and PAH emissions compared to the year 2005 will rise by approx. 30 %. If requirements are only made for new plants (comp. Figure 90, „regulations only for new plants“) the maximum in the year 2015 (+12.5 % compared with 2005) will be followed by similar values in the year 2025 as they were in 2005. If the regulations are made for new and existing plants (comp. Figure 90, „regulations for new plants and exchange programme“) the dust and PAH emissions will be reduced in 2025 by about 58% towards the value of 2005.

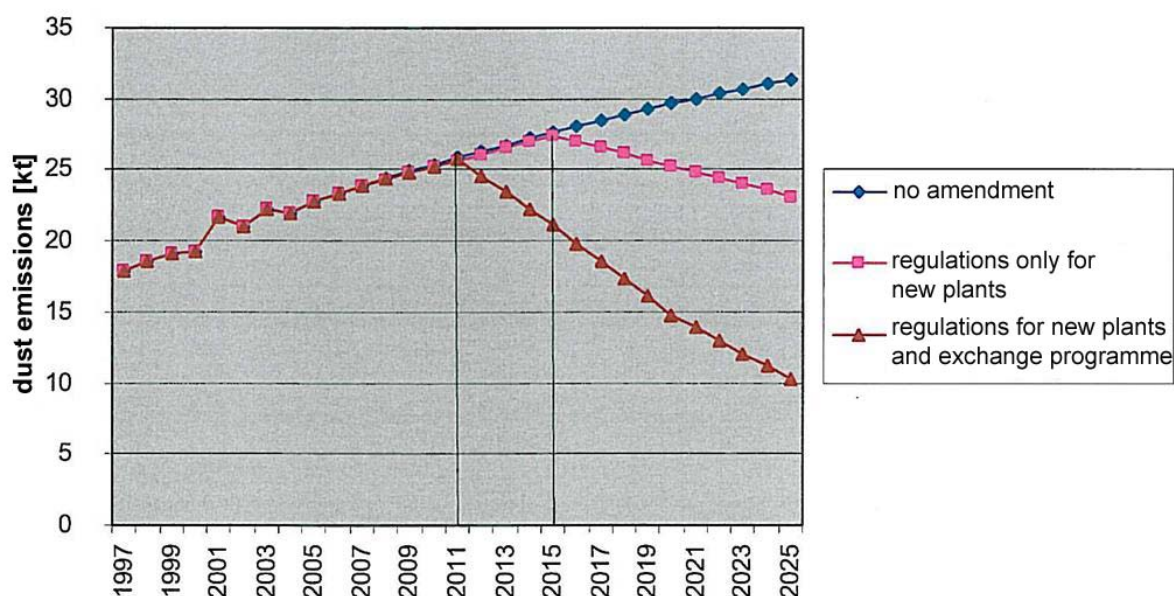


Figure 90: Prognosis of the development of dust emissions - amendment of the first Ordinance for the Implementation of the Federal Immission Control Act (BMU, 2007)

With the help of current data on PAH emissions and scenarios of the German Federal Environment Agency on the development of emissions for wood domestic combustion within the

framework of the amendment of the first Ordinance for the Implementation of the Federal Immission Control Act a first estimation of possible reduction potentials of PAH emissions into waterbodies by atmospheric emissions can be made. According to the specifications given by the PRTR (2007) for the period 1990 to 2005 domestic fires in Germany quantitatively have the largest share of the registered diffuse PAH total emissions into the atmosphere with in average > 80 %. On the basis of the German UN ECE CLRTAP emission report 2008 for the report year 2006 (UBA, 2008b) 33.4 t are allotted to the category „households“ and therefore 91 % of the registered atmospheric emissions of the PAH indicator substance benzo(a)pyrene being 36.7 t.

Compared to the sum parameters from four individual PAH compounds, which were also registered, annually 87.9 t (corresponds to 87 %) of the total registered 101.1 t are allotted to wood domestic combustion plants. For the further calculations a share of the domestic fires of 80 % is assumed for the  $\Sigma$  EPA-PAH<sub>16</sub> sum parameter. On the basis of the available data on waterbody emissions the possible impact on the pathways directly and indirectly influenced by the atmospheric deposition is derived.

The scenario „regulations for new plants and exchange programme“ acts on the assumption that  $\Sigma$  EPA-PAH<sub>16</sub>-emissions from domestic firing plants will be reduced by 58 % compared to the value in 2005 by the year 2025. This leads to a calculated reduction potential for the total emissions into waterbodies of 6.3 t annually which would correspond to a reduction of 32.5 % compared to 2005 (comp. Table 62).

Table 62: Scenarios: the development of  $\Sigma$  EPA-PAH<sub>16</sub> in waterbody emissions for the year 2025

Pathway	Emissions 2005	Emissions 2025	Emissions 2025
		Scenario „ regulations for new plants and ex- change programme “	Scenario „no amendment”“
	$\Sigma$ EPA-PAH <sub>16</sub> [kg/a]	$\Sigma$ EPA-PAH <sub>16</sub> [kg/a]	$\Sigma$ EPA-PAH <sub>16</sub> [kg/a]
MWWTP	1,269	860	1.480
Industry	443	443	443
Inland navigation	1,600	1,600	1.600
Atm. deposition	4,165	2,233	5.165
Erosion	1,248	958	1.398
Surface runoff	3,556	1,906	4.409
Drainage	93	93	93
Groundwater inflow	1,256	1,256	1.256
Sewer systems	5,794	3,763	6.844
<b>Sum</b>	<b>19,423</b>	<b>13,112</b>	<b>22.688</b>
<b>Reduction [%]</b>		<b>- 32</b>	<b>+ 17</b>
<b>Reduction [kg]</b>		<b>- 6,312</b>	<b>+ 3,265</b>

In contrast the scenario „no amendment“ assumes the  $\Sigma$  EPA-PAH<sub>16</sub> emissions from domestic fires will increase by 30 % compared to 2005 by the year 2025. This results in a calculated increase of PAH total emissions into waterbodies of 3.3 t annually which would correspond to an increase of 16.8 % compared to 2005. Assuming that the reduction measures are only implemented for domestic fires each leads to changed percentages of the observed pathways of  $\Sigma$  EPA-PAH<sub>16</sub> emissions into waterbodies. Prognoses on the basis of available data show that enforcing or desisting measures for domestic fires are elementary for the further development of PAH emissions into waterbodies (comp. Figure 91). Even though further reductions for all pathways are to be strived for, reductions for domestic fires should be given preferential consideration.

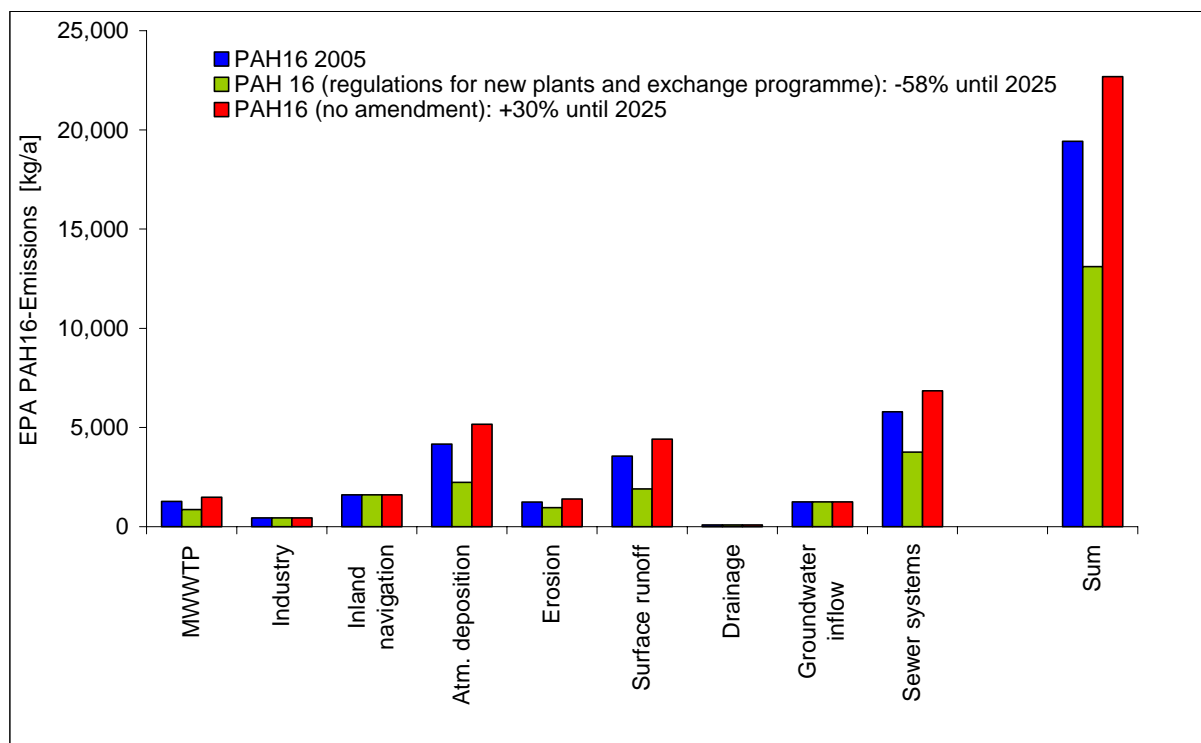


Figure 91: The influence of PAH<sub>16</sub> emissions from domestic fires on emissions into waterbody by 2025

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## Annex

Annex 1: Population development 1980-2005 [1,000 inh]

Federal state / Country	1980	1985	1990	1995	2000	2005
Baden-Wurttemberg	9,190	9,241	9,619	10,272	10,476	10,736
Bavaria	10,871	10,958	11,221	11,922	12,155	12,469
Berlin	3,055	3,064	3,400	3,472	3,387	3,395
Brandenburg	2,659	2,660	2,578	2,542	2,602	2,559
Bremen	695	666	674	680	663	663
Hamburg	1,653	1,592	1,626	1,706	1,705	1,744
Hesse	5,576	5,535	5,661	5,981	6,052	6,092
Mecklenburg-W. Pomerania	2,101	2,114	1,964	1,832	1,789	1,707
Lower Saxony	7,234	7,216	7,284	7,715	7,899	7,994
North Rhine-Westphalia	17,017	16,704	17,104	17,816	18,000	18,058
Rhineland-Palatinate	3,633	3,624	3,702	3,952	4,031	4,059
Saarland	1,069	1,051	1,065	1,084	1,072	1,050
Saxony	5,149	5,030	4,764	4,567	4,426	4,250
Saxony-Anhalt	3,060	3,021	2,874	2,759	2,649	2,470
Schleswig-Holstein	2,599	2,614	2,595	2,708	2,777	2,833
Thuringia	2,727	2,726	2,684	2,518	2,449	2,335
Germany	78,287	77,815	78,813	81,526	82,130	82,414
Belgium	9,855	9,858	9,948	10,131	10,239	10,446
Czech Republic	10,326	10,326	10,326	10,333	10,278	10,221
Denmark	5,122	5,111	5,135	5,216	5,330	5,411
France	53,714	55,173	56,577	58,567	60,538	62,638
Italy	56,389	56,602	56,694	56,846	56,929	58,462
Luxembourg	364	366	379	406	434	455
Netherlands	14,090	14,454	14,893	15,424	15,864	16,306
Austria	7,584	7,574	7,645	7,943	8,002	8,207
Poland	38,073	38,073	38,073	38,284	38,254	38,157
Liechtenstein	28	28	28	31	32	35
Switzerland	6,751	6,751	6,751	7,019	7,164	7,415

Sources: EUROSTAT (2007a), *Statistical State Offices of Brandenburg, Saxony, Saxony-Anhalt, Thuringia* and STATISTICAL YEARBOOK OF THE GDR (1986)

Annex 2: Connection rate of the population to public sewer systems [%]

<b>Federal state / Country</b>	<b>1983</b>	<b>1987</b>	<b>1991</b>	<b>1995</b>	<b>1998</b>	<b>2001</b>	<b>2004</b>
Baden-Wurttemberg	97.2	98.0	98.1	98.2	98.5	98.8	99.0
Bavaria	85.2	88.0	89.8	92.2	93.2	94.4	95.5
Berlin	96.9	96.9	96.9	98.0	98.4	98.5	98.4
Brandenburg	55.0	55.0	55.0	61.9	68.6	76.7	82.6
Bremen	99.1	99.9	100.0	100.0	99.9	99.4	99.8
Hamburg	94.4	95.2	98.7	96.7	98.5	100.0	98.9
Hesse	97.8	98.5	98.1	99.3	99.3	99.4	99.4
Mecklenburg-W. Pomerania	65.2	65.2	65.2	75.3	77.9	81.8	83.9
Lower Saxony	82.9	85.9	88.3	90.5	92.3	93.3	93.8
North Rhine-Westphalia	91.1	92.4	93.9	95.5	96.2	96.7	97.2
Rhineland-Palatinate	92.3	94.2	95.9	97.0	97.7	98.5	98.9
Saarland	98.5	98.5	98.7	98.8	98.8	99.2	99.1
Saxony	78.4	78.4	78.4	78.9	82.3	85.4	87.5
Saxony-Anhalt	73.4	73.4	73.4	79.4	79.3	84.3	88.3
Schleswig-Holstein	81.4	85.9	88.7	90.5	92.4	93.5	94.1
Thuringia	87.5	87.5	87.5	89.4	88.0	90.6	91.5
Germany	87.6	89.0	90.2	92.1	93.2	94.6	95.5
Belgium	81.0	81.0	81.0	81.0	82.0	82.0	82.0
Czech Republic	69.0	72.0	72.0	73.0	74.0	75.0	78.0
Denmark	88.0	88.0	87.0	87.0	89.0	89.0	89.0
France	81.0	81.0	81.0	81.0	79.0	82.0	82.0
Italy	61.0	61.0	61.4	63.0	67.5	69.0	69.0
Luxembourg	90.0	90.0	90.0	88.0	91.8	94.0	95.0
Netherlands	92.0	94.0	96.0	97.0	98.0	98.0	99.0
Austria	65.0	72.0	73.3	76.0	82.0	86.0	89.0
Poland	53.0	53.0	53.0	53.0	55.0	57.0	59.0
Liechtenstein	91.0	91.0	91.0	94.0	96.0	96.2	96.8
Switzerland	91.0	91.0	91.0	94.0	96.0	96.2	96.8

Sources: EUROSTAT (2007b), STATISTISCHES BUNDESAMT (1983, 1987, 1991, 1995, 1998, 2001, 2004)



Annex 3: Connection rate of the population to MWWTPs [%]

<b>Federal state/Country</b>	<b>1983</b>	<b>1987</b>	<b>1991</b>	<b>1995</b>	<b>1998</b>	<b>2001</b>	<b>2004</b>
Baden-Wurttemberg	93.9	96.5	97.3	97.6	98.2	99.0	98.9
Bavaria	80.5	84.6	87.5	90.5	92.0	94.0	94.9
Berlin	96.9	96.9	96.9	98.0	98.4	99.0	98.4
Brandenburg	53.7	53.7	53.7	61.1	68.5	77.0	82.5
Bremen	99.1	99.9	100.0	100.0	99.9	100.0	99.8
Hamburg	93.8	94.9	98.6	96.7	98.5	100.0	98.9
Hesse	86.6	91.6	95.8	97.2	98.5	99.0	99.0
Mecklenburg-W. Pomerania	63.0	63.0	63.0	70.7	76.9	82.0	83.7
Lower Saxony	81.7	84.9	87.3	90.3	92.2	93.0	93.8
North Rhine-Westphalia	89.8	91.9	93.6	95.3	96.1	97.0	97.2
Rhineland-Palatinate	80.3	85.8	89.9	94.1	96.3	98.0	98.7
Saarland	61.5	66.3	69.0	76.8	85.1	90.0	91.7
Saxony	59.5	59.5	59.5	64.2	73.2	78.0	81.9
Saxony-Anhalt	56.6	56.6	56.6	63.5	74.1	81.0	85.0
Schleswig-Holstein	79.8	84.3	87.6	89.9	92.2	93.0	93.9
Thuringia	49.0	49.0	49.0	53.6	57.6	61.0	65.0
Germany	81.1	83.5	85.7	88.6	91.0	92.8	94.1
Belgium	29.0	29.0	29.0	29.0	38.0	38.0	38.0
Czech Republic	46.0	49.0	51.0	56.0	62.0	65.0	71.0
Denmark	80.0	81.0	86.0	87.0	89.0	89.0	89.0
France	50.0	51.0	70.6	77.0	77.0	79.0	79.0
Italy	61.0	61.0	61.4	63.0	67.5	69.0	69.0
Luxembourg	83.0	86.5	90.0	88.0	91.8	94.0	95.0
Netherlands	72.0	89.0	94.0	97.0	98.0	98.0	99.0
Austria	65.0	72.0	72.0	75.0	81.0	86.0	89.0
Poland	34.0	34.0	34.0	42.0	49.2	55.0	59.0
Liechtenstein	87.0	91.0	91.0	94.0	96.0	96.2	96.8
Switzerland	87.0	91.0	91.0	94.0	96.0	96.2	96.8

Sources: EUROSTAT (2007c), STATISTISCHES BUNDESAMT (1983, 1987, 1991, 1995, 1998, 2001, 2004)

Annex 4: Share of combined sewer system in Germany [%]

<b>Federal state</b>	<b>1983</b>	<b>1987</b>	<b>1991</b>	<b>1995</b>	<b>1998</b>	<b>2001</b>	<b>2004</b>
Baden-Wurttemberg	89.3	88.6	88.2	87.4	85.8	83.8	82.7
Bavaria	84.7	83.0	81.9	77.8	74.9	72.4	69.6
Berlin	36.3	36.3	34.5	34.1	33.7	32.5	31.4
Brandenburg	26.8	26.8	26.8	28.5	8.3	6.5	5.1
Bremen	49.2	48.0	42.2	44.9	43.7	42.4	41.4
Hamburg	42.0	39.1	36.9	35.2	34.7	34.1	33.8
Hesse	89.3	88.4	91.0	88.8	89.2	88.3	87.8
Mecklenburg-W. Pomerania	38.6	38.6	38.6	24.2	14.4	11.1	9.4
Lower Saxony	14.3	13.2	11.5	10.3	8.9	8.6	7.6
North Rhine-Westphalia	71.7	70.1	70.1	68.8	67.5	66.1	64.1
Rhineland-Palatinate	90.5	90.5	90.5	88.8	86.3	83.6	81.1
Saarland	93.7	92.6	92.8	92.9	92.3	93.6	92.7
Saxony	77.8	77.8	77.8	67.8	58.1	55.6	52.2
Saxony-Anhalt	80.5	80.5	80.5	58.4	51.9	35.9	30.1
Schleswig-Holstein	18.7	19.8	16.3	14.8	13.8	12.7	12.0
Thuringia	89.5	89.5	89.5	85.7	82.0	80.7	81.6

Annex 5: Specific storage volume of the stormwater overflow tanks [m³/ha]

<b>Federal State</b>	<b>1983</b>	<b>1987</b>	<b>1991</b>	<b>1995</b>	<b>1998</b>	<b>2001</b>	<b>2004</b>
Baden-Wurttemberg	11.4	11.4	18.6	20.9	23.9	28.0	29.6
Bavaria	3.2	3.2	9.0	12.5	20.6	21.7	23.5
Berlin	1.7	1.7	1.7	2.2	2.3	2.7	5.2
Brandenburg	0.6	0.6	0.6	1.2	2.1	3.0	28.2
Bremen	10.6	10.6	18.7	22.0	28.6	23.5	24.1
Hamburg	0.0	0.0	3.8	8.3	4.4	5.3	17.1
Hesse	2.7	2.7	10.3	13.2	17.4	20.7	21.6
Mecklenburg-W. Pomerania	0.4	0.4	0.4	1.5	2.1	13.8	50.9
Lower Saxony	1.5	1.5	7.1	8.7	17.4	22.0	29.7
North Rhine-Westphalia	3.8	3.8	14.0	18.9	25.0	26.7	25.1
Rhineland-Palatinate	3.6	3.6	8.3	11.7	14.1	16.3	18.7
Saarland	0.3	0.3	4.8	8.3	11.2	15.4	17.4
Saxony	0.4	0.4	0.4	2.3	3.9	6.9	7.8
Saxony-Anhalt	0.1	0.1	0.1	1.7	2.5	6.4	9.2
Schleswig-Holstein	3.2	3.2	6.9	16.1	17.2	20.4	12.9
Thuringia	0.3	0.3	0.3	1.8	5.2	5.8	9.3

Sources: STATISTISCHES BUNDESAMT (1983, 1987, 1991, 1995, 1998, 2001, 2004)

Annex 6: Nitrogen emissions from MWWTPs [t/a]

<b>Federal State</b>	<b>2005/2004<sup>1</sup></b>	<b>2001<sup>2</sup></b>	<b>1998<sup>2</sup></b>	<b>1995<sup>3</sup></b>	<b>1985<sup>3</sup></b>
Baden-Wurttemberg	16,173	16,218	19,061	29,090	41,212
Bavaria	21,708	22,228	24,045	32,612	48,411
Berlin	830	1,422	2,362	2,854	7,050
Brandenburg	2,762	2,130	3,417	6,295	11,507
Bremen	529	645	766	3,307	4,121
Hamburg	2,041	2,296	2,245	2,780	5,867
Hesse	6,794	9,637	13,770	17,945	20,749
Mecklenburg-W. Pomerania	996	1,103	1,430	3,296	7,221
Lower Saxony	3,356	4,603	7,274	15,740	30,764
North Rhine-Westphalia	19,196	24,459	30,137	49,362	65,792
Rhineland-Palatinate	3670	4,767	5,841	9,347	14,045
Saarland	1,494	1,874	2,163	2,259	2,592
Saxony	3,827	5,554	6,668	13,513	16,001
Saxony-Anhalt	1,209	1,433	3,337	5,709	9,913
Schleswig-Holstein	1,772	3,485	5,430	6,289	10,311
Thuringia	1,297	1,414	2,443	4,460	6,695

Annex 7: Phosphorus emissions from MWWTPs [t/a]

<b>Federal State</b>	<b>2005/2004<sup>1</sup></b>	<b>2001<sup>2</sup></b>	<b>1998<sup>2</sup></b>	<b>1995<sup>3</sup></b>	<b>1985<sup>3</sup></b>
Baden-Wurttemberg	1,158	1,437	1,388	1,511	7,475
Bavaria	1,729	2,148	1,906	1,864	8,149
Berlin	25	41	69	58	286
Brandenburg	182	167	215	497	2,185
Bremen	23	24	26	54	590
Hamburg	90	91	102	110	1,391
Hesse	826	973	1,024	872	4,304
Mecklenburg-W. Pomerania	126	114	114	186	1,608
Lower Saxony	414	538	688	1,030	5,566
North Rhine-Westphalia	1,475	1,659	1,944	2,088	11,503
Rhineland-Palatinate	403	589	607	680	2,961
Saarland	140	196	201	227	607
Saxony	359	455	574	1,060	3,850
Saxony-Anhalt	132	141	241	531	2,669
Schleswig-Holstein	176	187	206	212	1,685
Thuringia	199	235	221	370	1,811

Sources: <sup>1</sup>FEDERAL ENVIRONMENT AGENCY, <sup>2</sup>FDZ (2007), <sup>3</sup>BEHRENDT ET AL. (1999)

Annex 8: Aggregation of the land use classes of the CORINE map to the land use classes used in MONERIS

<b>MONERIS-landuse</b>	<b>CLC-ID</b>	<b>Description</b>
Urban areas	110	
Urban areas	111	Continuous urban fabric
Urban areas	112	Discontinuous urban fabric
Urban areas	121	Industrial or commercial units
Urban areas	122	Road and rail networks and associated land
Urban areas	123	Port areas
Urban areas	124	Airports
Open pit	131	Mineral extraction sites
Open pit	132	Dump sites
Open pit	133	Construction sites
Urban areas	141	Green urban areas
Urban areas	142	Sport and leisure facilities
Arable land	211	Non-irrigated arable land
Arable land	212	Permanently irrigated land
Arable land	213	Rice fields
Arable land	221	Vineyards
Arable land	222	Fruit trees and berry plantations
Arable land	223	Olive groves
Grassland/Pastures	231	Pastures
Arable land	241	Annual crops associated with permanent crops
Arable land	242	Complex cultivation patterns
Arable land	243	Land principally occupied by agriculture, with significant areas of natural vegetation
Arable land	244	Agro-forestry areas
Natural covered areas	311	Broad-leaved forest
Natural covered areas	312	Coniferous forest
Natural covered areas	313	Mixed forest
Natural covered areas	321	Natural grasslands
Natural covered areas	322	Moors and heathland
Natural covered areas	323	Sclerophyllous vegetation
Natural covered areas	324	Transitional woodland-shrub
Not covered areas	331	Beaches, dunes, sands
Not covered areas	332	Bare rocks
Not covered areas	333	Sparsely vegetated areas
Not covered areas	334	Burnt areas
Not covered areas	335	Glaciers and perpetual snow
Wetlands	411	Inland marshes
Wetlands	412	Peat bogs
Wetlands	421	Salt marshes
Wetlands	422	Salines

Wetlands	423	Intertidal flats
Water surfaces	511	Water courses
Water surfaces	512	Water bodies
Water surfaces	521	Coastal lagoons
Water surfaces	522	Estuaries
Water surfaces	523	Sea and ocean
Other areas	999	
Other areas	950	
Other areas	951	
Other areas	952	

#### Annex 9: Overview of time-constant input data (MONERIS Basic-Info)

Variable	Description	Unit	Source
ID	Identifier of the analytical unit		IGB
BI_ID_GIS	ID_GIS		IGB
BI_Country	Country		-
BI_State	Federal state		-
BI_WA	Working area		-
BI_SEA	Sea		-
BI_des	Description		-
BI_AU	Name of the analytical unit		-
BI_SU	Subunit		-
BI_SB	Subbasin		-
BI_RB	River basin		-
BI_RBD	River basin district		-
BI_MS	Quality monitoring station		Chap. 3.2
BI_MONIcatch_A	Official size of the quality monitoring station	km <sup>2</sup>	Chap. 3.2
BI_AU_A	Size of the analytical unit	km <sup>2</sup>	IGB
BI_FNE_nosplit	Flow net equation without splitting		Chap. 2.2
BI_FNE_split	Flow net equation with splitting		Chap. 2.2
BI_FNE_upst	Flow net equation upstream		Chap. 2.2
BI_AD_nhxlt	Atmospheric deposition, NH <sub>4</sub> , long term	mg/m <sup>2</sup>	EMEP (2006)
BI_AD_noxlt	Atmospheric deposition, NO <sub>x</sub> , long term	mg/m <sup>2</sup>	EMEP (2006)
BI_PREC_yrIt	Yearly precipitation, long term	mm/a	GPCC (2006)
BI_PREC_slIt	Precipitation summer, long term	mm/a	GPCC (2006)
BI_EVAP0_lIt	Evapotranspiration, long term	mm/a	GPCC (2006)
BI_LU_urb	Urban areas	km <sup>2</sup>	EEA (2005)
BI_AL_A1	Arable land <1 %	km <sup>2</sup>	EEA (2005); USGS (1996)
BI_AL_1_2	Arable land 1-2 %	km <sup>2</sup>	EEA (2005); USGS (1996)

BI_AL_2_4	Arable land 2-4 %	km <sup>2</sup>	EEA (2005); USGS (1996)
BI_AL_4_8	Arable land 4-8 %	km <sup>2</sup>	EEA (2005); USGS (1996)
BI_AL_8	Arable land >8 %	km <sup>2</sup>	EEA (2005); USGS (1996)
BI_GL	Grassland / Pastures	km <sup>2</sup>	EEA (2005)
BI_NATCOV	Natural covered areas	km <sup>2</sup>	EEA (2005)
BI_WSA	Water surfaces (CORINE)	km <sup>2</sup>	EEA (2005)
BI_OPM	Open pit mine	km <sup>2</sup>	EEA (2005)
BI_OA	Not covered areas	km <sup>2</sup>	EEA (2005)
BI_WL	Wetlands	km <sup>2</sup>	EEA (2005)
BI_REM	Other areas	km <sup>2</sup>	EEA (2005)
BI_POTERO	Areas with erosion potential	km <sup>2</sup>	EEA (2005)
BI_TD	Tile drained areas	%	BEHRENDT ET AL. (2003a)
BI_ELEVA	Average elevation of the analytical unit	m	USGS (1996)
BI_SLOPE_1000	Average slope (1000m) in the analytical unit	%	USGS (1996)
BI_SLOPE_100	Average slope (100m) in the analytical unit	%	CGIAR (2004)
BI_SO_S	Sand dominated soils	km <sup>2</sup>	BGR (1998); FAO (2007)
BI_SO_C	Clay dominated soils	km <sup>2</sup>	BGR (1998); FAO (2007)
BI_SO_L	Loam dominated soils	km <sup>2</sup>	BGR (1998); FAO (2007)
BI_SO_F	Fen	km <sup>2</sup>	BGR (1998); FAO (2007)
BI_SO_B	Bog	km <sup>2</sup>	BGR (1998); FAO (2007)
BI_SO_SI	Silt dominated soils	km <sup>2</sup>	BGR (1998); FAO (2007)
BI_SO_Ncont	Nitrogen content in topsoil	%	BGR (1998); FAO (2007)
BI_SO_Ccont	Clay content in topsoil	%	BGR (1998); FAO (2007)
BI_SL_AL1	Soil erosion on arable land with a slope < 1%	t/(ha·a)	EEA (2005); EUROPEAN SOIL BUREAU (2007); NASA- SRTM (2005)
BI_SL_AL1_2	Soil erosion on arable land with a slope 1-2 %	t/(ha·a)	EEA (2005); EUROPEAN SOIL BUREAU (2007); NASA- SRTM (2005)



BI_SL_AL2_4	Soil erosion on arable land with a slope 2-4 %	t/(ha·a)	EEA (2005); EUROPEAN SOIL BUREAU (2007); NASA-SRTM (2005)
BI_SL_AL4_8	Soil erosion on arable land with a slope 4-8 %	t/(ha·a)	EEA (2005); EUROPEAN SOIL BUREAU (2007); NASA-SRTM (2005)
BI_SL_AL8	Soil erosion on arable land with a slope >8 %	t/(ha·a)	EEA (2005); EUROPEAN SOIL BUREAU (2007); NASA-SRTM (2005)
BI_SL_GL	Soil erosion on grassland / pastures	t/(ha·a)	EEA (2005); EUROPEAN SOIL BUREAU (2007); NASA-SRTM (2005)
BI_SL_NATCOV	Soil erosion on natural covered areas	t/(ha·a)	EEA (2005); EUROPEAN SOIL BUREAU (2007); NASA-SRTM (2005)
BI_SL_mean	Mean soil erosion on all areas	t/(ha·a)	EEA (2005); EUROPEAN SOIL BUREAU (2007); NASA-SRTM (2005)
BI_C	C-Factor (ABAG)	-	Chap. 4.1.2.3
BI_Pacc	Accumulation of phosphorus (reference year)	kg/(ha·a)	BEHRENDT ET AL. (2003a); WERNER ET AL. (1994)
N_surpl	Nitrogen surplus (reference year)	kg/(ha·a)	AUERSWALD et al. (1986); BACH & SKITSCHAK (2007)
BI_HYG_uncs	Unconsolidated rocks, shallow groundwater	km <sup>2</sup>	USGS (1996)
BI_HYG_uncd	Unconsolidated rocks, deep groundwater	km <sup>2</sup>	USGS (1996)
BI_HYG_conhp	Consolidated rocks, highly permeable	km <sup>2</sup>	USGS (1996)
BI_HYG_conimp	Consolidated rocks, low permeable	km <sup>2</sup>	USGS (1996)
BI_GW_rest	Mean residence time in groundwater	a	KUNKEL ET AL. (2007)
BI_Lakes_mrA	Area of lakes in the main river	km <sup>2</sup>	Chap. 2.4
BI_Lakes_tribA	Area of lakes in the tributary river	km <sup>2</sup>	Chap. 2.4
BI_mrA	Water surface of flowing waters in main river	km <sup>2</sup>	Chap. 2.4
BI_tribA	Water surface of flowing waters in tributary river	km <sup>2</sup>	Chap. 2.4

BI_WSA_mrtrib	Total water surface	km <sup>2</sup>	Chap. 2.4
BI_WSA_mrol	Area of lakes in the main river at the outlet of the analytical unit	km <sup>2</sup>	Chap. 2.4

Annex 10: Overview of the temporally varying input data (MONERIS Periodical-Data)

Short name	Description	Unit	Source
PD_AD_nh	Atmospheric deposition, NHx	mg/m <sup>2</sup>	EMEP (2006)
PD_AD_no	Atmospheric deposition, NOx	mg/m <sup>2</sup>	EMEP (2006)
PD_AD_tp	Atmospheric deposition, TP	mg/m <sup>2</sup>	BEHRENDT ET AL. (2003a)
PD_PREC_yr	Yearly precipitation	mm/a	EEA (2007)
PD_PREC_s	Precipitation summer	mm/a	EEA (2007)
PD_SPL_fact	Splitting-factor	-	Chap. 2.2, 2.3
PD_Q_calc_net	Modelled discharge of the analytical unit	m <sup>3</sup> /s	Chap. 2.3
PD_Qobs_Aucatch	Observed discharge	m <sup>3</sup> /s	Chap. 3.2
PD_Lobs_DIN	Observed DIN river load	t/a	Chap. 3.2
PD_Lobs_TN	Observed TN river load	t/a	Chap. 3.2
PD_Lobs_TP	Observed TP river load	t/a	Chap. 3.2
PD_W_temp	Water temperature	°C	Chap. 3.2
PD_INH_tot	Total inhabitants	-	Chap. 3.3.7
PD_INH_con	Inhabitants connected	-	Chap. 3.3.8
PD_INH_conSW	Inhabitants connected to public sewer systems and MWWTP	-	Chap. 3.3.8
PD_W_TPhist	Factor for considering the change in WWTP discharges relating to a reference year, TP	-	Chap. 3.3.8
PD_W_TNhist	Factor for considering the change in WWTP discharges relating to a reference year, TN	-	Chap. 3.3.8
PD_InD_TP	Emissions from industrial direct dischargers, TP	kg/a	Chap. 3.3.8
PDInD_TN	Emissions from industrial direct dischargers, TN	kg/a	Chap. 3.3.8
PD_CSS_share	Share of the length of combined sewers in the total sewer system	%	Chap. 3.3.8
PD_CSO_storage	Storage volume in combined sewer system	%	Chap. 3.3.8
remaining WWTP_TN	Remaining loads from MWWTP, recorded as sum, TN	kg/a	Chap. 3.3.8
remaining WWTP_TP	Remaining loads from MWWTP, recorded as sum, TP	kg/a	Chap. 3.3.8
W_SC_PCI	Transfer of the scenario-settings to each WWTP: share of population connected to public sewer systems and WWTP	-	Chap. 3.3.8
W_Catch_ID	ID of the analytical unit into which a WWTP discharges	-	Chap. 3.3.8

W_name	Name of the WWTP / Location	-	Chap. 3.3.8
W_size_class	Size class of the WWTP	-	Chap. 3.3.8
W_TS_1	Primary treatment available? Y/N, Indicator for distinction between industrial direct dischargers and WWTP	Y/N	Chap. 3.3.8
W_LTN	TN-load of the MWWTP	kg/a	Chap. 3.3.8
W_LTP	TP-load of the MWWTP	kg/a	Chap. 3.3.8
W_Q	Discharge of the MWWTP	m <sup>3</sup> /s	Chap. 3.3.8
W_TNconc	Average concentration of the MWWTP-effluent, TN	mg/l	Chap. 3.3.8
W_TPconc	Average concentration of the MWWTP-effluent, TP	mg/l	Chap. 3.3.8
NOW_LN	TN-load of the direct industrial discharger	t/a	Chap. 3.3.8
NOW_LP	TP- load of the direct industrial discharger	t/a	Chap. 3.3.8
Dis_NOW	Discharge of the direct industrial discharger	m <sup>3</sup> /s	Chap. 3.3.8
NOW_concN	Average concentration in the effluent of the direct industrial discharger, TN	mg/l	Chap. 3.3.8
NOW_concP	Average concentration in the effluent of the direct industrial discharger, TP	mg/l	Chap. 3.3.8

## Annex 11: Overview of used variables

Short name	Description	Unit
<b>General precalculations</b>		
IM_QcalcAUCatch	Calculated discharge of the analytical unit	m <sup>3</sup> /s
IM_Qobscorr	Corrected discharge of the analytical unit	m <sup>3</sup> /s
IM_qcalc	Specific runoff in the analytical unit	l/s/km <sup>2</sup>
IM_qcalcAUCatch	Average specific runoff in the analytical unit	l/s/km <sup>2</sup>
IM_AL1_A	Arable land < 1% under consideration of scenarios on land use change	km <sup>2</sup>
IM_AL1-2_A	Arable land 1-2% under consideration of scenarios on land use change	km <sup>2</sup>
IM_AL2-4_A	Arable land 2-4% under consideration of scenarios on land use change	km <sup>2</sup>
IM_AL4-8_A	Arable land 4-8% under consideration of scenarios on land use change	km <sup>2</sup>
IM_AL8_A	Arable land > 8% under consideration of scenarios on land use change	km <sup>2</sup>
IM_AL_Atot	Total arable land	km <sup>2</sup>
IM_AL_and_AU	Share of arable land in the analytical unit	%
IM_GL_A	Area of grassland in the analytical unit	km <sup>2</sup>
IM_AL_A	Agricultural area in the analytical unit	km <sup>2</sup>
IM_NONIMP_A	Unsealed area in the analytical unit	km <sup>2</sup>
IM_SNOW_A	Snow- und ice-covered area in the analytical unit	km <sup>2</sup>
IM_TD_and_AU	Share of drained areas in the analytical unit	%
IM_USG_and_AU	Unconsolidated rocks, shallow groundwater	%
IM_UDG_and_AU	Unconsolidated rocks, deep groundwater	%
IM_CHP_and_AU	Consolidated rocks, highly permeable	%
IM_CI_and_AU	Consolidated rocks, low permeable	%
IM_SCNHX	NHx deposition, reduced at NAC	mgN/m <sup>2</sup>
IM_SCNOX	NOx deposition, reduced at NAC	mgN/m <sup>2</sup>
IM_Nsurp	N surplus	kg/(ha·a)
IM_TPacc	P accumulation	kg/ha·a
IM_CSS_and_TSS	Share of combined sewer system	%
IM_SSS_and_TSS	Share of separate sewer system	%
IM_IUA	Impervious area	km <sup>2</sup>
IM_AUMONcorr	Correction factor on catchment-area between discharge and quality monitoring station	-
IM_SCNHXlt	NHx deposition long-term average, reduced at NAC	mgN/m <sup>2</sup>
IM_SCNOXlt	NOx deposition long-term average, reduced at NAC	mgN/m <sup>2</sup>
IM_Lobs_DIN	Observed DIN river load	t/a
IM_Lobs_TN	Observed TN river load	t/a
IM_LOBS_TP	Observed TP river load	t/a
IM_WSAtot	Total water surface	km <sup>2</sup>
<b>Country data</b>		

CD_Nsurp_cur	N surplus corrected by country data in the calculation year	kg/(ha·a)
CD_Nsurp_gwres	Average N surplus corrected by country data during the groundwater retention time	kg/(ha·a)
CD_Nsurp_ratio	N surplus determined by country data in the calculation year	-
CD_Admin_index	Sort key federal state	-
CD_Admin_row	Sort key country data	-
CD_GW_resclass	Reclassified groundwater retention time	a
CD_Pcont_basis	Phosphorus content, basic value	mg/kg
CD_Pcont	Phosphorus content, corrected	mg/kg
CD_Pacc_coun	Phosphorus accumulation	kg/(ha·a)
CD_Cfact_coun	C-factor for erosion according to ABAG	-
CD_Pinh_coun	TP-inhabitant specific disposal per day according to country data	mg/(E·d)
CD_Pdet_coun	TP-inhabitant specific disposal per day from detergents according to country data	mg/(E·d)
CD_CSOV_coun	Extension degree of CSO according to country data	%
<b><u>Atmospheric deposition</u></b>		
AD_TP	Total deposition onto water surfaces TP	t/a
AD_TN	Total deposition onto water surfaces TN	t/a
AD_DIRPREC_Q	Water balance (N-V) over water surfaces	m³/s
<b><u>Surface runoff</u></b>		
SR_TP	Total emissions via surface runoff TP	t/a
SR_TN	Total emissions via surface runoff TN	t/a
SR_Qtot	Total surface runoff (absolut)	m³/s
SR_q	Total surface runoff (specific)	mm
SR_Q_WSV	Surface runoff from natural areas with vegetation	m³/s
SR_AA_Q	Surface runoff from agricultural areas	m³/s
SR_SNOW_Qpl	Surface runoff from snow-covered areas (interim result)	m³/s
SR_SNOW_Q	Surface runoff from snow-covered areas (final result)	m³/s
SR_WSV_TPC	TP-concentration in surface runoff from areas covered by vegetation	mg/l
SR_WSV_TNC	TN-concentration in surface runoff from areas covered by vegetation	mg/l
SR_WSV_TP	TP in surface runoff from natural areas with vegetation	t/a
SR_WSV_TN	TN in surface runoff from natural areas with vegetation	t/a
SR_TPaccratio	Ratio of P accumulation in the analytical unit to the average P-accumulation in Germany	-
SR_AI_TPC	TP-concentration in surface runoff from arable area	mg/l
SR_pasture_TPC	TP-concentration in surface runoff from grassland/pastures	mg/l
SR_mean_TPC	Average TP-concentration in surface runoff	mg/l
SR_AL_TNC	TN-concentration in surface runoff from arable area	mg/l
SR_AL_TP	TP-emission from surface runoff from agricultural area	t/a
SR_AL_TN	TN-emissions from surface runoff from agricultural area	t/a
SR_SNOW_TP	TP-emission from surface runoff from snow-covered area	t/a

SR_SNOW_TN	TN-emission from surface runoff from snow-covered area	t/a
<b><u>Erosion</u></b>		
ER_TP	Total emissions via erosion, TP	t/a
ER_TN	Total emissions via erosion, TN	t/a
ER_AL_pl	Arable area, without soil conservation measures	km²
ER_AL	Arable area, mit bodenkonservierenden Maßnahmen	km²
ER_SL_AL	Soil loss from arable area, specific	t/(ha·a)
ER_SL	Soil loss from arable area, absolute	t/a
ER_SLcorr	Soil loss from arable area, corrected	t/a
ER_SLnat	Soil loss from naturally covered areas	t/a
ER_SDR	Ratio of soil loss to sediment input into surface waters	%
ER_SEDin	Sediment input, absolute	t/a
ER_SEDspec	Sediment input, specific	t/(km²·a)
ER_ENR	Enrichment ratio	-
ER_TS_TPcont	P-content in topsoil	mg/kg
ER_TS_TNcont	N-content in topsoil	mg/kg
ER_PRECcorr	Correction factor on precipitation	-
ER_RF	R-factor according to ABAG, current calculation year	N/m²
ER_RFlt	R-factor according to ABAG, long-term	N/m²
ER_SEDtot	Total sediment input	t/a
<b><u>Drainage</u></b>		
TD_TP	Total emissions via drainage, TP	t/a
TD_TN	Total emissions via drainage, TN	t/a
TD_Nsurp_tot	TN-input from topsoil into drained areas	t/a
TD_A	Drained areas	km²
TD_q	Tile drainage rate	mm/a
TD_Q	Discharge from drained areas	m³/s
TD_TPC	TP-concentration from drained areas	mg/l
TD_TNC	TN-concentration from drained areas	mg/l
<b><u>Groundwater</u></b>		
GW_TP	Total emissions via groundwater, TP	t/a
GW_TN	Total emissions via groundwater, TN	t/a
GW_TNin	TN-input from topsoil into groundwater	t/a
GW_R_A1	Areas contributing to groundwater recharge (part 1)	km²
GW_R_A2	Areas contributing to groundwater recharge (Teil 2)	km²
GW_Q	Groundwater discharge, absolute	m³/s
GR_q	Groundwater discharge, specific, interim result, current calculation year	mm/a
GW_qcorr	Groundwater discharge, specific, final result, current calculation year	mm/m²
GW_qltcorr	Groundwater discharge, specific, final result, long-term	mm/a
GW_LW_q	Seepage water amount, spezifisch	mm/a
GW_RT	Retention time of groundwater	a
GW_TNsurp_RT	average N-surplus during retention time	kg/(ha·a)



GW_TNsurp_tot	Total considered N-surplus	kg
GW_ADcorr	corrected atmospheric deposition	kg
GW_TNsurp_spec	Total N-surplus	kg/(ha-a)
GW_TNC_LW	N-concentration in seepage water	mg/l
GW_TNCSUBS	N-concentration in subsoil	mg/l
GW_RET_TNS%LW	N-retention during soil percolation	%
GW_RET_TNGW%SUBS	N-retention in groundwater	%
GW_RET_TNS+GW	Total retention during soil percolation and in groundwater	%
GW_TNC	N-concentration in groundwater	mg/l
GW_TPC	P-concentration, area-weighted (according to soil type)	mg/l
GW_TPCcorr	corrected P-concentration (considering anoxic conditions)	mg/l
GW_CR_TN	Concentration ratio groundwater/seepage water	-
GW_CR_TNlt	Concentration ratio groundwater/seepage water (long-term)	-
GW_BG_TNC	TN-concentration in groundwater (natural background)	mg/l
GW_BG_TNL	TN-emission groundwater (natural background)	t/a
GW_BG_Nsurp	N-surplus on all agricultural areas (natural background)	kg
GW_BG_NsurpAD	N-surplus and atmospheric deposition on all agricultural areas (natural background)	kg
GW_BG_TPC	TP-concentration in groundwater (natural background)	mg/l
GW_BG_TP	TP-emission Groundwater (natural background)	t/a
GW_TS_TNC	TN-concentration in topsoil	mg/l
GW_RZ_TNC	TN-concentration root zone	mg/l
GW_TS_TN	TN-emissions from topsoil	t/a
GW_RZ_TN	TN-emissions from root zone	t/a
GW_RZ_Q	Seepage water after percolating the root zone	m³/s
GW_ON	Input of dissolved organic N (DON) from groundwater	t/a
<b><u>Natural background</u></b>		
BG_TP	Total emissions under natural conditions, TP	t/a
BG_TN	Total emissions under natural conditions, TN	t/a
BG_AD_TP	TP via atmospheric deposition	t/a
BG_AD_TN	TN via atmospheric deposition	t/a
BG_GWR_A	Areas contributing to groundwater recharge	km²
BG_LW_q	Seepage rate	mm
BG_LW_TNC	TN-concentration in seepage water	mg/l
BG_GW_TNC	TN-concentration in groundwater	mg/l
BG_GW_TNCcorr	Corrected TN-concentration in groundwater	mg/l
BG_GW_TP	TP-concentration in groundwater	t/a
BG_GW_TN	TN-concentration in groundwater	t/a
BG_SR_TNC	TP-concentration in surface runoff	mg/l
BG_SR_TP	TP in surface runoff	t/a
BG_SR_TN	TN in surface runoff	t/a
BG_ER_TP	TP-emissions via erosion (natural background)	t/a
BG_ER_TN	TN-emissions via erosion (natural background)	t/a

BG_SNOW_Q	Runoff from snow-covered areas	m³/s
BG_SNOW_TP	TP-emissions from snow-covered areas (natural background)	t/a
BG_SNOW_TN	TN-emissions from snow-covered areas (natural background)	t/a
BG_SDR	Ratio of erosion and sediment input in surface waters	%
<b><u>Sewer systems</u></b>		
US_TP	Total emissions via sewer systems, TP	t/a
US_TN	Total emissions via sewer systems, ZN	t/a
US_POPdens	Inhabitants per ha	E/ha
US_POP	Inhabitants, total, calculated from population density	E/1000
US_INH	Inhabitants, total	E/1000
US_INH_conn	connected inhabitants, total	E/1000
US_INH_connW+SS	connected inhabitants, MWWTP and sewer systems	E/1000
US_INH_connSS	connected inhabitants, to sewer systems but not to MWWTP	E/1000
US_INH_notconn	Not connected inhabitants	E/1000
US_IUA_and_AU	Share of impervious areas of urban areas	%
US_IUA_tot	Impervious areas, absolute	km²
US_A_SS	impervious areas, connected to separate sewer system	km²
US_A_CS	impervious areas, connected to combined sewer system	km²
US_A_onlySS	impervious areas, connected to sewer systems but not to MWWTP	km²
US_A_notconn	impervious areas, not connected	km²
US_SWEff	Rainfall event causing CSO	-
US_SWE	Rainfall event	-
US_CS_SV	Storage volume of CSO (bezogen auf 23,3 m³/ha = 100 %)	%
US_CS_Qr	Share of discharged wastewater amount without storage volume	%
US_CS_Q	Share of discharged wastewater amount with storage volume	%
US_impA_Qratio	Specific runoff from impervious areas	-
US_SS_Q	Water amount via separate sewer system	m³/a
US_CS_QPREC	Water amount via precipitation	m³/a
US_CSO_QPREC	Water amount from CSO	m³/a
US_CSO_Q_INH	Water amount from inhabitants	m³/a
US_CSO_Q_SV	Water amount during rainfall event from CSO	m³/a
US_CSO_Q_tot	Water amount during an overflow	m³/a
US_Q_SSnotW	Water amount, discharge via sewer systems without connection to MWWTP	m³/a
US_Q_noSS	Water amount, not connected households and areas	m³/a
US_Qurb	Total water amount from sewer systems	m³/s
US_SSRW_TNC	TN-concentration in storm sewer of separate sewer system	mg/l
US_SSRW_TPC	TP-concentration in storm sewer of separate sewer system	mg/l
US_CSO_TNC	N-concentration in the combined sewer system	mg/l

US_CSO_TPC	P-concentration in the combined sewer system	mg/l
US_RET_noSS_TN	TN-retention in soil and groundwater	%
US_RET_noSS_TP	TP- retention in soil and groundwater	%
US_SS_TN	TN-emissions via separate sewer system	t/a
US_SS_TP	TP-emissions via separate sewer system	t/a
US_CS_TN	TN-emissions via combined sewer system	t/a
US_CS_TP	TP-emissions via combined sewer system	t/a
US_INH_TN	TN-emissions from inhabitants only connected to sewer systems	t/a
US_INH_TP	TP-emissions from inhabitants only connected to sewer systems	t/a
US_IUA_TN	TN-emissions from areas only connected to sewer systems	t/a
US_IUA_TP	TP-emissions from areas only connected to sewer systems	t/a
US_onlySS_TN	TN-emissions from inhabitants and areas only connected to sewer systems	t/a
US_onlySS_TP	TP-emissions from inhabitants and areas only connected to sewer systems	t/a
US_noSS_TN	TN-emissions from not connected areas and inhabitants	t/a
US_noSS_TP	TP-emissions from not connected areas and inhabitants	t/a
US_Pfree_ratio	Share of detergents of the total inhabitant-specific P-emissions	-
US_noW_TN	N-emissions from impervious areas and inhabitants, connected to sewer systems but not to MWWTP	t/a
US_noW_TP	P-emissions from impervious areas and inhabitants, connected to sewer systems but not to MWWTP	t/a
US_noW_Q	Water amount via direct dischargers, calculated from detailed MWWTP-inventory	m³/a
US_notconn_TN	TN-emissions via direct dischargers, calculated from detailed MWWTP-inventory	t/a
US_notconn_TP	TP-emissions via direct dischargers, calculated from detailed MWWTP-inventory	t/a
<b>Point sources</b>		
PS_TP_sum	TN-emissions from point sources	t/a
PS_TN_sum	TP-emissions from point sources	t/a
PS_Q_sum	Discharges from point sources	m³/s
W_Name	Name of the MWWTP	-
PS_SC_P2	Assumed maximum P-concentration, for MWWTP of 2 <sup>nd</sup> size class	mg/l
PS_SC_P3	Assumed maximum P-concentration, for MWWTP of 3 <sup>rd</sup> size class	mg/l
PS_SC_P4	Assumed maximum P-concentration, for MWWTP of 4 <sup>th</sup> size class	mg/l
PS_SC_P5	Assumed maximum P-concentration, for MWWTP of 5 <sup>th</sup> size class	mg/l
PS_SC_P6	Assumed maximum P-concentration, for MWWTP of 6 <sup>th</sup> size class	mg/l
PS_SC_N2	Assumed maximum N-concentration, for MWWTP of 2 <sup>nd</sup> size class	mg/l

PS_SC_N3	Assumed maximum N-concentration, for MWWTP of 3 <sup>rd</sup> size class	mg/l
PS_SC_N4	Assumed maximum N-concentration, for MWWTP of 4 <sup>th</sup> size class	mg/l
PS_SC_N5	Assumed maximum N-concentration, for MWWTP of 5 <sup>th</sup> size class	mg/l
PS_SC_N6	Assumed maximum N-concentration, for MWWTP of 6 <sup>th</sup> size class	mg/l
PS_W_SC	Size class of each MWWTP	-
PS_Nconcmax	maximum effluent concentration of the MWWTP, TN	mg/l
PS_Pconcmax	maximum effluent concentration of the MWWTP, TP	mg/l
PS_Nconc	corrected effluent concentration of the MWWTP, TN	mg/l
PS_Pconc	corrected effluent concentration of the MWWTP, TP	mg/l
PS_Qcur	Water amount of the MWWTP	m <sup>3</sup> /s
PS_W_LTN	TN-emission of each MWWTP	t/a
PS_W_LTP	TP-emission of each MWWTP	t/a
<b><u>Retention</u></b>		
RE_Lcalc_DIN	Modelled river load for comparing with observed river load, DIN	t/a
RE_Lcalc_TN	Modelled river load for comparing with observed river load, TN	t/a
RE_Lcalc_TP	Modelled river load for comparing with observed river load, TP	t/a
RE_rett_tribDIN	Retention in tributary, DIN	t/a
RE_rett_mrDIN	Retention in main river, DIN	t/a
RE_rett_tribTN	Retention in tributary, TN	t/a
RE_rett_mrTN	Retention in main river, TN	t/a
RE_rett_tribTP	Retention in tributary, TP	t/a
RE_rett_mrTP	Retention in main river, TP	t/a
RE_TOT_TN	TN, total emissions from analytical unit, net	t/a
RE_TOT_TP	TP, total emissions from analytical unit, net	t/a
RE_DIRMR_TN	TN, direct emissions in the main river, net	t/a
RE_DIRMR_TP	TP, direct emissions in the main river, net	t/a
RE_GW_ON	Dissolved organic nitrogen emissions from groundwater, net	t/a
RE_GW_AUcatchON	Dissolved organic nitrogen emissions from groundwater, tot	t/a
RE_GW_upsON	Dissolved organic nitrogen emissions upstream, DON	t/a
RE_HL_trib	Average hydraulic load in tributary (HL)	m/a
RE_HL_mr	Average hydraulic load in main river (HL)	m/a
RE_HL_mrol	Average hydraulic load in surface water bodies at the outlet of an analytical unit (HL)	m/a
RE_HL_tribTP	Retention factor (HL-approach for tributaries), TP	-
RE_q_tribTP	Retention factor (specific runoff for tributaries), TP	-
RE_mean_tribTP	Average retention factor (specific runoff and HL-approach for tributaries), TP	-

RE_Lcalc_tribTP	Modelled load of tributaries, TP	t/a
RE_HL_mrTP	Retention factor (HL-approach for main river), TP	-
RE_HL_mrolTP	Retention factor (HL-approach for surface water bodies at the outlet of an analytical unit), TP	-
RE_Lcalc_upsTP	Modelled load upstream, TP	t/a
RE_Lcalc_mrTP	Modelled load in main river, TP	t/a
RE_Lcalc_TOTTP	Modelled river load for comparing with observed river load, TP	t/a
RE_THL_tribDIN	Retention factor (THL-approach for tributaries), DIN	-
RE_Lcalc_tribDIN	Modelled river load, tributaries, DIN	t/a
RE_THL_mrDIN	Retention factor (THL-approach for main river), DIN	-
RE_THL_mrolDIN	Retention factor (THL-approach for surface water bodies at the outlet of an analytical unit), DIN	-
RE_Lcalc_upsDIN	Modelled load upstream, DIN	t/a
RE_Lcalc_mrDIN	Modelled load in main river, DIN	t/a
RE_Lcalc_TOTDIN	Modelled river load for comparing with observed river load, DIN	t/a
RE_THL_tribTN	Retention factor (THL-approach for tributaries), TN	-
RE_Lcalc_tribTN	Modelled load of tributaries, TN	t/a
RE_THL_mrTN	Retention factor (THL-approach for main river), TN	-
RE_THL_mrolTN	Retention factor (THL-approach for surface water bodies at the outlet of an analytical unit), TN	-
RE_Lcalc_upsTN	Modelled load upstream, TN	t/a
RE_Lcalc_mrTN	Modelled load in main river, TN	t/a
RE_Lcalc_TOTTNwoDON	Modelled load without dissolved organic fraction, TN	t/a
RE_Lcalc_TOTTN	Modelled river load for comparing with observed river load, TN	t/a

#### Annex 12: Overview of the variables of the scenario manager

Short name	Description	Unit/value
SC_W_and_S	All inhabitants connected to sewer system are also connected to MWWTP	1/0
SC_PCI	Share of inhabitants connected to sewer system and MWWTP	%
SC_CSO	Increase of volume of treated wastewater CSO tanks	%
SC_SRSS	Increase of volume of treated stormwater in separate sewer system	%
SC_urb_pave	Transformation of impervious into unsealed areas	%
SC_P_det	Use of phosphate-free detergents	1/0
SC_W_P2	max. P-effluent concentration of a MWWTP, 2 <sup>nd</sup> size class	mg/l
SC_W_P3	max. P-effluent concentration of a MWWTP, 3 <sup>rd</sup> size class	mg/l
SC_W_P4	max. P-effluent concentration of a MWWTP, 4 <sup>th</sup> size class	mg/l
SC_W_P5	max. P-effluent concentration of a MWWTP, 5 <sup>th</sup> size class	mg/l
SC_W_P6	max. P-effluent concentration of a MWWTP, 6 <sup>th</sup> size class 6	mg/l

SC_W_N2	max. N-effluent concentration of a MWWTP, 2 <sup>nd</sup> size class	mg/l
SC_W_N3	max. P-effluent concentration of a MWWTP, 3 <sup>rd</sup> size class	mg/l
SC_W_N4	max. P-effluent concentration of a MWWTP, 4 <sup>th</sup> size class	mg/l
SC_W_N5	max. P-effluent concentration of a MWWTP, 5 <sup>th</sup> size class	mg/l
SC_W_N6	max. P-effluent concentration of a MWWTP, 6 <sup>th</sup> size class	mg/l
SC_arab_con	Connection of arable areas to surface waters	%
SC_TD	Change of share of drained areas	%
SC_N_surpl	Change of N-surplus	%
SC_AtG_1	Transformation of arable areas to grassland/pastures, areas < 1 %	1/0
SC_AtG_1_2	Transformation of arable areas to grassland/pastures, areas 1-2 %	1/0
SC_AtG_2_4	Transformation of arable areas to grassland/pastures, areas 2-4 %	1/0
SC_AtG_4_8	Transformation of arable areas to grassland/pastures, areas 4-8%	1/0
SC_AtG__8	Transformation of arable areas to grassland/pastures, areas >8 %	1/0
SC_contill_1	Conservation tillage < 1%	1/0
SC_contill_1_2	Conservation tillage 1-2%	1/0
SC_contill_2_4	Conservation tillage 2-4%	1/0
SC_contill_4_8	Conservation tillage 4-8%	1/0
SC_contill_8	Conservation tillage > 8%	1/0
SC_W_fixN	General reduction of discharges via MWWTP < 2.000 PT resp. Of the summed-up emissions via MWWTP from analytical units outside Germany, TN	%
SC_W_fixP	General reduction of discharges via MWWTP < 2.000 PT resp. Of the summed-up emissions via MWWTP from analytical units outside Germany, TP	%

#### Annex 13: Overview of the used model constants

Constant	Description	Value
<b><u>Surface runoff</u></b>		
CSR1	P-concentration via fertilizer / forest	0.035
CSR2	P-concentration via fertilizer / open land	0.035
CSR3	N-concentration via fertilizer / arable area	0.3
CSR4	N- concentration via fertilizer / grassland/pastures	0.0
CSR5	N- concentration via fertilizer / forest and open land	0.0
CSR6	P-saturation / arable land	90
CSR7	P-saturation / grassland/pastures	80
CSR8	P-accumulation Germany	1100
CSR9	P-water solubility factor 1	0.035
CSR10	P-water solubility factor 2	0.000000618
CSR11	P-water solubility factor 3	0.155
CSR12	P-concentration snow	0.01
CSR13	N-concentration snow	0.1
CSR14	Precipitation upper limit	800
CSR15	Runoff snow factor 1	4



CSR16	Runoff snow factor 2	1
CSR17	Criterion snowmelt	0.545637289
CSR18	Coefficient surface runoff	0.0426
CSR19	Exponent surface runoff	1.2461
<b><u>Erosion</u></b>		
CE1	Sediment delivery ratio, factor 1	0.006684
CE2	Sediment delivery ratio, factor 2	0.3
CE3	Sediment delivery ratio, factor 3	1.5
CE4	Sediment delivery ratio, factor 4	-0.25
CE5	If slope smaller	0.25
CE6	Then SDR =	0
CE7	Enrichment ratio factor 1, TP	18
CE8	Enrichment ratio factor 1, TN	7.7
CE9	Enrichment ratio factor 2	-0.47
CE10	if SEDspec smaller	1
CE11	then EnR =	18
CE12	Natural soil erosion	4
CE13	P-content forest	150
CE14	P-content arable area	250
CE15	Calculation factor for soil erosion	1.0
CE16	Reduction of soil erosion because of cultivation without plough	90
CE17	R-factor summer-precipitation factor 1	0.152
CE18	R-factor summer-precipitation factor 2	6.88
CE19	Sediment-delivery under no-agronomic conditions	20
<b><u>Drainage</u></b>		
CTD1	Runoff coefficient winter	0.5
CTD2	Runoff coefficient summer	0.1
CTD3	P-concentration sand	0.2
CTD4	P-concentration loam	0.06
CTD5	P-concentration fen	0.3
CTD6	P-concentration bog	2
CTD7	Exponent for describing denitrification in soil	0.85
<b><u>Groundwater</u></b>		
CGW1	Ratio TP/SRP aerobic conditions	1
CGW2	Ratio TP/SRP anaerobic conditions	2.5
CGW3	P-concentration / forest / current values	0.02
CGW4	P-concentration / sand / current values	0.1
CGW5	P-concentration / loam / current values	0.03
CGW6	P-concentration / fen / current values	0.1
CGW7	P-concentration / bog / current values	0.5
CGW8	P-concentration / forest / background conditions	0.02
CGW9	P-concentration / sand / background conditions	0.02

CGW10	P-concentration / loam / background conditions	0.02
CGW11	P-concentration / fen / background conditions	0.02
CGW12	P-concentration / bog / background conditions	0.035
CGW13	Unconsolidated rocks, shallow groundwater: factor 1	2752.22
CGW14	Unconsolidated rocks, shallow groundwater: factor 2	-1.54
CGW15	Unconsolidated rocks, deep groundwater: factor 1	68561.63
CGW16	Unconsolidated rocks, deep groundwater: factor 2	-1.96
CGW17	Consolidated rocks, high porosity: factor 1	60.23
CGW18	Consolidated rocks, high porosity: factor 2	-0.90
CGW19	Consolidated rocks, low permeable: factor 1	0.01
CGW20	Consolidated rocks, low permeable: factor 2	0.66
CGW21	Exponent for considering denitrification in the unsaturated zone	0.64
CGW22	Factor for the long-term change of the atmospheric deposition	1
CGW23	Sub-surface flow: factor 1	1.0990
CGW24	Sub-surface flow: factor 2	0.9497
CGW25	Interflow: factor 1	0.1463
CGW26	Interflow: factor 2	1.1247
CGW27	Baseflow: factor 1	1.1760
CGW28	Baseflow: factor 2	0.8535
CGW29	Surface runoff: factor 1	0.0426
CGW30	Surface runoff: factor 2	1.2461
CGW31	Threshold value of the redox-ratio	0.1
CGW32	Organic N / forest	0
CGW33	Organic N / wetlands	5
<b><u>Sewer systems</u></b>		
CUS1	Calculation of the share of impervious areas in the total urban area, factor 1	0.4047
CUS2	Calculation of the share of impervious areas in the total urban area, factor 2	9.6
CUS3	Calculation of the share of impervious areas in the total urban area, factor 3	0.573
CUS4	Calculation of the share of impervious areas in the total urban area, factor 4	0.0391
CUS5	Runoff ratio, factor 1	0.15
CUS6	Runoff ratio, factor 2	0.75
CUS7	Flood events, factor 1	0.0000012
CUS8	Flood events, factor 2	2.5
CUS9	P-concentration in commercial wastewater	0.5
CUS10	specific P-input from impervious areas	2.5
CUS11	Inhabitant-specific N-input	12
CUS12	N-concentration in commercial wastewater	2
CUS13	spezific N-input from impervious areas	4
CUS14	Inhabitant-specific water amount	130

CUS15	Wastewater from commercial areas	0.1
CUS16	Duration of wastewater discharge from commercial areas	10
CUS17	N-disposal from inhabitants connected only to sewer systems	9
CUS18	N-disposal from not connected inhabitants	9
CUS19	Share of P-disposal from not connected inhabitants	0.75
CUS20	N-retention in soil / consolidated rocks	50
CUS21	N-retention in soil / unconsolidated rocks	90
CUS22	P-retention in soil / consolidated rocks	50
CUS23	P-retention in soil / unconsolidated rocks	90
CUS24	Average rainfall runoff rate	1
CUS25	Combined sewer system, factor 1	0.01534
CUS26	Combined sewer system, factor 2	0.97541
CUS27	Combined sewer system, factor 3	196.66
CUS28	Share of the suspended solids transported from septic tanks to MWWTP	5
CUS29	Connection to separate sewer system: factor 1	0.125
CUS30	Connection to separate sewer system: factor 2	0.368
CUS31	Point sources correction factor	0.7
<b><u>Natural background</u></b>		
CBG1	P-concentration snow	0.01
CBG2	N-concentration snow	0.1
CBG3	Precipitation upper limit	850
CBG4	Runoff snow, factor 1	2
CBG5	Runoff snow, factor 2	0.45
CBG6	P-Back groundwater	0.02
CBG7	P-Back atmosphere	0.1
CBG8	P-Back surface runoff	0.035
CBG9	N-Back atmosphere	1
CBG10	N-Back surface runoff	1
CBG11	Enrichment ratio TP: factor 1	18
CBG12	Enrichment ratio TN: factor 1	7.7
CBG13	Enrichment ratio: factor 2	-0.47
CBG14	if SEDspec smaller	1
CBG15	then EnR =	18
CBG16	Natural soil erosion / slope	0
CBG17	Natural soil erosion	2
CBG18	Natural P-accumulation	150
CBG19	Minimum seepage water rate	20
CBG20	C-factor / background conditions	0.005
CBG21	Sediment-delivery for not-agronomic conditions	5
<b><u>Eingangsdaten, Grundlagen- und Szenarienberechnung</u></b>		
CSP1	Clay-P-model, factor 1	10.2
CSP2	Clay-P-model, factor 2	150

<b><u>Retention</u></b>		
CR1	q-approach, RT, factor 1	5.1
CR2	q-approach, RT, factor 2	-1
CR3	HL-approach, TP, factor 1	25.74
CR4	HL-approach, TP, factor 2	-1.00
CR5	THL-approach, DIN, factor 1	8.58
CR6	THL-approach, DIN, factor 2	0.07
CR7	THL-approach, DIN, factor 3	-1.00
CR8	THL-approach, TN, factor 1	4.74
CR9	THL-approach, TN, factor 2	0.07
CR10	THL-approach, TN, factor 3	-1.00
<b><u>Municipal WWTP</u></b>		
CW1	Average retention for SC-direct dischargers into the MWWTP, TN	0.7
CW1	Average retention for SC-direct dischargers into the MWWTP, TP	0.7
<b><u>Water surfaces</u></b>		
WSA1	River width tributaries, factor1	0.152
WSA2	River width tributaries, factor2	0.102
WSA3	River width tributaries, factor3	1.018
WSA4	River width tributaries, factor4	-0.25
WSA5	River width main river, factor1	0.35
WSA6	River width main river, factor2	0.468
WSA7	River width main river, factor3	0.36
WSA8	River width main river, factor4	-0.027
WSA9	Scale correction factor for the tributaries	1 - 8.4 (Table 1)
WSA10	Scale correction factor for the main river	1 – 1.18 (Table 1)

#### Annex 14: Overview of the results variables

<b>Net emissions / Total emissions</b>		
<b>Short name</b>	<b>Description</b>	<b>Unit</b>
EN_POP ET_POP	Number of inhabitants	inh.
EN_AU_A ET_AU_A	Area of analytical unit	km <sup>2</sup>
EN_WSA_A ET_WSA_A	Total water surface	km <sup>2</sup>
EN_AL_A ET_AL_A	Agricultural area	km <sup>2</sup>
EN_Q ET_Q	Total discharge, analytical unit	m <sup>3</sup> /s
EN_RZ_Q	Groundwater recharge (including drainage)	m <sup>3</sup> /s

ET_RZ_Q		
EN_SR_Q ET_SR_Q	Surface runoff from agricultural areas	m³/s
EN_AD_TN ET_AD_TN	Emissions via direct atmospheric deposition onto surface water bodies, TN	t/a
EN_SR_TN ET_SR_TN	Emissions via surface runoff (all areas except impervious areas), TN	t/a
EN_TD_TN ET_TD_TN	Emissions via drained areas, TN	t/a
EN_ER_TN ET_ER_TN	Emissions via erosion, TN	t/a
EN_GW_TN ET_GW_TN	Emissions via groundwater, TN	t/a
EN_W_TN ET_W_TN	Emissions via MWWTP including WWTP < 2,000 PT and not connected inhabitants, TN	t/a
EN_US_TN ET_US_TN	Emissions via sewer systems, TN	t/a
EN_TOT_TN ET_TOT_TN	Total TN-emissions	t/a
EN_BG_TN ET_BG_TN	Emissions via natural background, TN	t/a
EN_USO_TN ET_USO_TN	Emissions via urban sources (WWTP + sewer systems), TN	t/a
EN_ASO_TN ET_ASO_TN	Emissions via agriculture, TN	t/a
EN_OSO_TN ET_OSO_TN	Emissions via other sources, TN	t/a
EN_Nsurp_AA ET_Nsurp_AA	N-surplus on agricultural areas	t/a
EN_Nsurp_TOT ET_Nsurp_TOT	Total N-surplus	t/a
EN_RZ_TN ET_RZ_TN	N loss in the root zone	t/a
EN_SRAA_TN ET_SRAA_TN	Emissions via surface runoff from agricultural areas, TN	t/a
EN_GWRET_TN ET_GWRET_TN	N-retention in groundwater	t/a
EN_GW_ON ET_GW_ON	Emissions via persistent dissolved organic N-compounds	t/a
EN_DIRMR_TN ET_DIRMR_TN	Emissions via point sources and sewer systems direct in the main river of an analytical unit, TN	t/a
EN_AD_TP ET_AD_TP	Emissions via direct atmospheric deposition onto surface water bodies, TP	t/a
EN_SR_TP ET_SR_TP	Emissions via surface runoff (all areas except impervious areas), TP	t/a
EN_TD_TP ET_TD_TP	Emissions via drained areas, TP	t/a

EN_ER_TP ET_ER_TP	Emissions via erosion, TP	t/a
EN_GW_TP ET_GW_TP	Emissions via groundwater, TP	t/a
EN_W_TP ET_W_TP	Emissions via MWWTP including WWTP < 2,000 PT and not connected inhabitants, TP	t/a
EN_US_TP ET_US_TP	Emissions via sewer systems, TP	t/a
EN_TOT_TP ET_TOT_TP	Total TP-emissions	t/a
EN_BG_TP ET_BG_TP	Emissions via natural background, TP	t/a
EN_USO_TP ET_USO_TP	Emissions via urban sources (WWTP + sewer systems), TP	t/a
EN_ASO_TP ET_ASO_TP	Emissions via agriculture, TP	t/a
EN_OSO_TP ET_OSO_TP	Emissions via other sources, TP	t/a
EN_RZ_TP ET_RZ_TP	P loss in the root zone	t/a
EN_SRAA_TP ET_SRAA_TP	Emissions via surface runoff from agricultural areas, TP	t/a
EN_DIRMR_TP ET_DIRMR	Emissions via point sources and sewer systems direct in the main river of an analytical unit, TP	t/a
<b>Scenario results</b>		
<b>Short name</b>	<b>Description</b>	<b>Unit</b>
SC_RACIt_DIN	Cumulative retention from the outlet of the analytical unit to the river basin outlet at the sea, TN	-
SC_RACIt_TN	Cumulative retention from the outlet of the analytical unit to the river basin outlet at the sea, DIN	-
SC_TOT_TNAU%RBD	Share of total emissions of an analytical unit in the total emissions of a river system, TN	%
SC_BG_TNAU%RBD	Share of the emissions via natural background of an analytical unit in the total emissions of a river system, TN	%
SC_USO_TNAU%RBD	Share of the emissions via urban sources of an analytical unit in the total emissions of a river system, TN	%
SC_ASO_TNAU%RBD	Share of the emissions via agricultural sources of an analytical unit in the total emissions of a river system, TN	%
SC_OSO_TNAU%RBD	Share of the emissions via other sources of an analytical unit in the total emissions of a river system, TN	%
SC_TOT_TPAU%RBD	Share of total emissions of an analytical unit in the total emissions of a river system, TP	%
SC_BG_TPAU%RBD	Share of the emissions via natural background of an analytical unit in the total emissions of a river system, TP	%
SC_USO_TPAU%RBD	Share of the emissions via urban sources of an analytical unit in the total emissions of a river system, TP	%



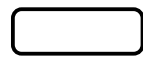
SC_ASO_TPAU%RBD	Share of the emissions via agricultural sources of an analytical unit in the total emissions of a river system, TP	%
SC_OSO_TPAU%RBD	Share of the emissions via other sources of an analytical unit in the total emissions of a river system, TP	%
SC_LBG_TNAU%OL	River load at the outlet of a river system caused by emissions via natural background of an analytical unit, TN	t/a
SC_LUSO_TNAU%OL	River load at the outlet of a river system caused by emissions via urban sources of an analytical unit, TN	t/a
SC_LASO_TNAU%OL	River load at the outlet of a river system caused by emissions via agricultural sources of an analytical unit, TN	t/a
SC_LOSO_TNAU%OL	River load at the outlet of a river system caused by emissions via other sources of an analytical unit, TN	t/a
SC_Load_TNAUatOL	River load at the outlet of a river system caused by the total emissions of an analytical unit, TN	t/a
SC_Load_TNAUcatchatOL	River load at the outlet of a river system caused by the total emissions of the catchment area of an analytical unit, TN	t/a
SC_Load_TNAUT	River load at the outlet of an analytical unit caused by the total emissions of an analytical unit, TN	t/a
SC_Load_TNAUcatch	River load at the outlet of an analytical unit caused by the total emissions of the catchment area of an analytical unit, TN	t/a
SC_LBG_TPAU%OL	River load at the outlet of a river system caused by emissions via natural background of an analytical unit, TP	t/a
SC_LUSO_TPAU%OL	River load at the outlet of a river system caused by emissions via urban sources of an analytical unit, TP	t/a
SC_LASO_TPAU%OL	River load at the outlet of a river system caused by emissions via agricultural sources of an analytical unit, TP	t/a
SC_LOSO_TPAU%OL	River load at the outlet of a river system caused by emissions via other sources of an analytical unit, TP	t/a
SC_Load_TPAUatOL	River load at the outlet of a river system caused by the total emissions of an analytical unit, TP	t/a
SC_Load_TPAUcatchatOL	River load at the outlet of a river system caused by the total emissions of the catchment area of an analytical unit, TP	t/a
SC_Load_TPAUT	River load at the outlet of an analytical unit caused by the total emissions of an analytical unit, TP	t/a
SC_Load_TPAUcatch	River load at the outlet of an analytical unit caused by the total emissions of the catchment area of an analytical unit, TP	t/a
<b>Target concentrations</b>		
<b>Short name</b>	<b>Description</b>	<b>Unit</b>
TC_AU_TNCmax	Assumed maximum concentration of the river loads resulting from the total emissions of an analytical unit, TN	mg/l
TC_OL_TNCresult	Resulting concentration of the river loads of a river system at the outlet of the river system, TN	mg/l

TC_AU_TPCmax	Assumed maximum concentration of the river loads resulting from the total emissions of an analytical unit, TP	mg/l
TC_OL_TPCresult	Resulting concentration of the river loads of a river system at the outlet of the river system, TP	mg/l
TC_Lref_TP	Resulting reference load of a river system at the outlet of the river system, TP	t/a
TC_Ltarget_TP	Resulting river load at compliance of the determined target-concentration of a river system at the outlet of the river system, TP	t/a
TC_Ldelta_tot_TP	Difference between reference load and load at target-concentration for a river system at the outlet of the river system, TP	t/a
TC_Ldelta_USO_TP	Difference between reference load and load at target-concentration resulting from emission changes due to scenarios on urban sources for a river system at the outlet of the river system, TP	t/a
TC_Ldelta_OS_TP	Difference between reference load and load at target-concentration resulting from assumed emission changes due to the compliance of the target-concentration for a river system at the outlet of the river system, TP	t/a
TC_Ldelta_tot_TP_pro	Difference between reference load and load at target-concentration for a river system at the outlet of the river system, TP	%
TC_Ldelta_USO_TP_pro	Difference between reference load and load at target-concentration resulting from emission changes due to scenarios on urban sources for a river system at the outlet of the river system, TP	%
TC_Ldelta_OS_TP_pro	Difference between reference load and load at target-concentration resulting from assumed emission changes due to the compliance of the target-concentration for a river system at the outlet of the river system, TP	%
TC_EMlref_TP	Total emissions of a river basin under reference conditions, TP	t/a
TC_EMltarget_TP	Total emissions of a river basin at compliance of the target-concentration, TP	t/a
TC_EMldelta_tot_TP	Difference of the emissions under reference conditions and at compliance of the target-concentration, TP	t/a
TC_EMldelta_USO_TP	Emission changes due to assumed scenarios on urban sources, TP	t/a
TC_EMldelta_OS_TP	Emission changes for compliance of the target-concentration, TP	t/a
TC_EMldelta_tot_TP_pro	Difference of the emissions under reference conditions and at compliance of the target-concentration, TP	%
TC_EMldelta_USO_TP_pro	Emission changes due to assumed scenarios on urban sources, TP	%
TC_EMldelta_OS_TP_pro	Emission changes for compliance of the target-concentration, TP	%
TC_EMlref_TN	Total emissions of a river basin under reference con-	t/a

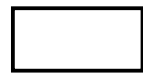
	ditions, TN	
TC_EMItarget_TN	Total emissions of a river basin at compliance of the target-concentration, TN	t/a
TC_EMIdelta_tot_TN	Difference of the emissions under reference conditions and at compliance of the target-concentration, TN	t/a
TC_EMIdelta_USO_TN	Emission changes due to assumed scenarios on urban sources, TN	t/a
TC_EMIdelta_OS_TN	Emission changes for compliance of the target-concentration, TN	t/a
TC_EMIdelta_tot_TN_pro	Difference of the emissions under reference conditions and at compliance of the target-concentration, TN	%
TC_EMIdelta_USO_TN_pro	Emission changes due to assumed scenarios on urban sources, TN	%
TC_EMIdelta_OS_TN_pro	Emission changes for compliance of the target-concentration, TN	%
TC_Lref_TN	Resulting reference load of a river system at the outlet of the river system, TN	t/a
TC_Ltarget_TN	Resulting river load at compliance of the determined target-concentration of a river system at the outlet of the river system, TN	t/a
TC_Ldelta_tot_TN	Difference between reference load and load at target-concentration for a river system at the outlet of the river system, TN	t/a
TC_Ldelta_USO_TN	Difference between reference load and load at target-concentration resulting from emission changes due to scenarios on urban sources for a river system at the outlet of the river system, TN	t/a
TC_Ldelta_OS_TN	Difference between reference load and load at target-concentration resulting from assumed emission changes due to the compliance of the target-concentration for a river system at the outlet of the river system, TN	t/a
TC_Ldelta_tot_TN_pro	Difference between reference load and load at target-concentration for a river system at the outlet of the river system, TN	%
TC_Ldelta_USO_TN_pro	Difference between reference load and load at target-concentration resulting from emission changes due to scenarios on urban sources for a river system at the outlet of the river system, TN	%
TC_Ldelta_OS_TN_pro	Difference between reference load and load at target-concentration resulting from assumed emission changes due to the compliance of the target-concentration for a river system at the outlet of the river system, TN	%

note: analytical unit (EN), sum according to FNE\_split (ET)

## Legend



input data



intermediate calculation



run-off related



area related



nutrient / substance related



non-specific relation



decision divide

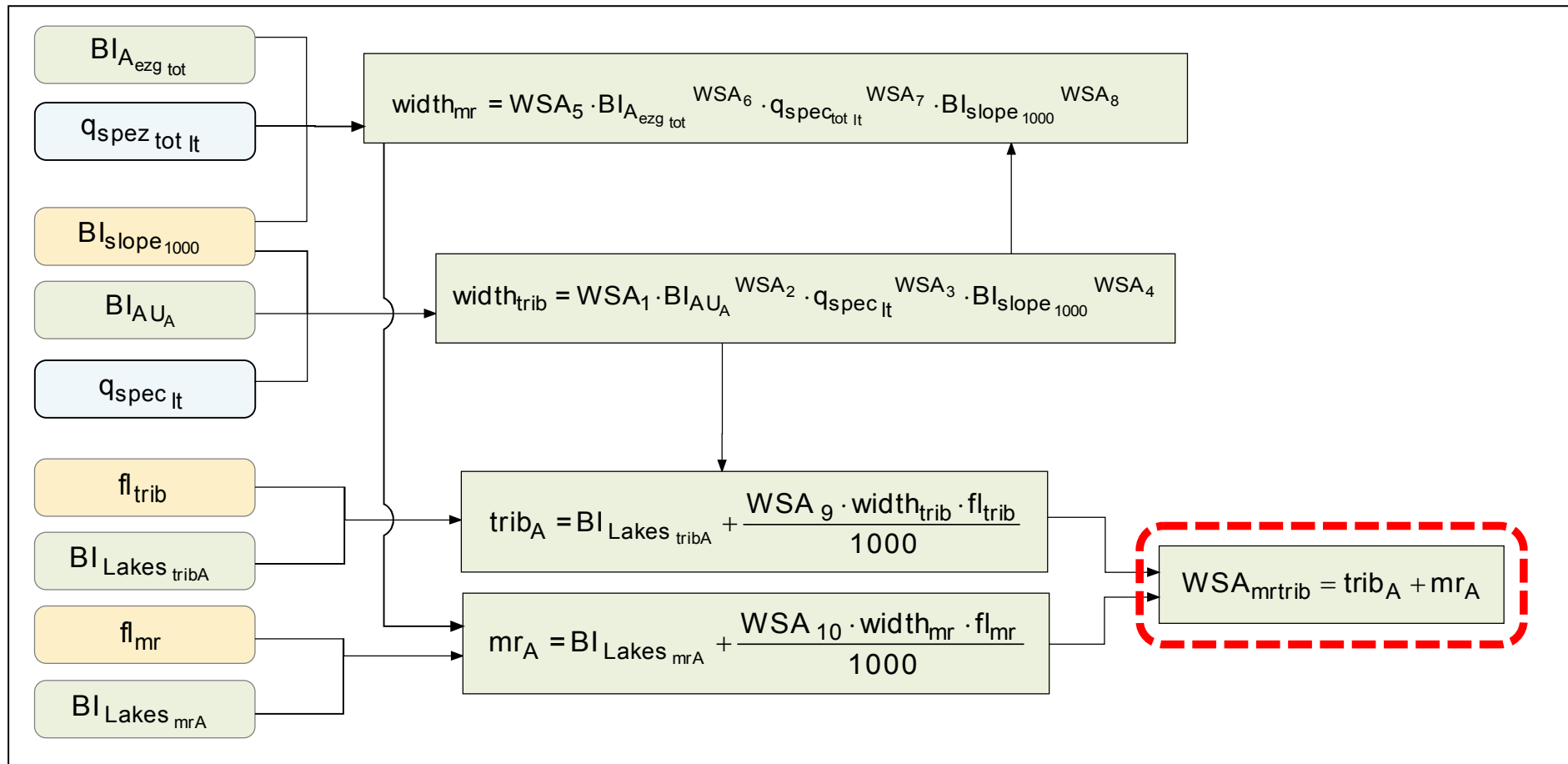


results from groundwater module

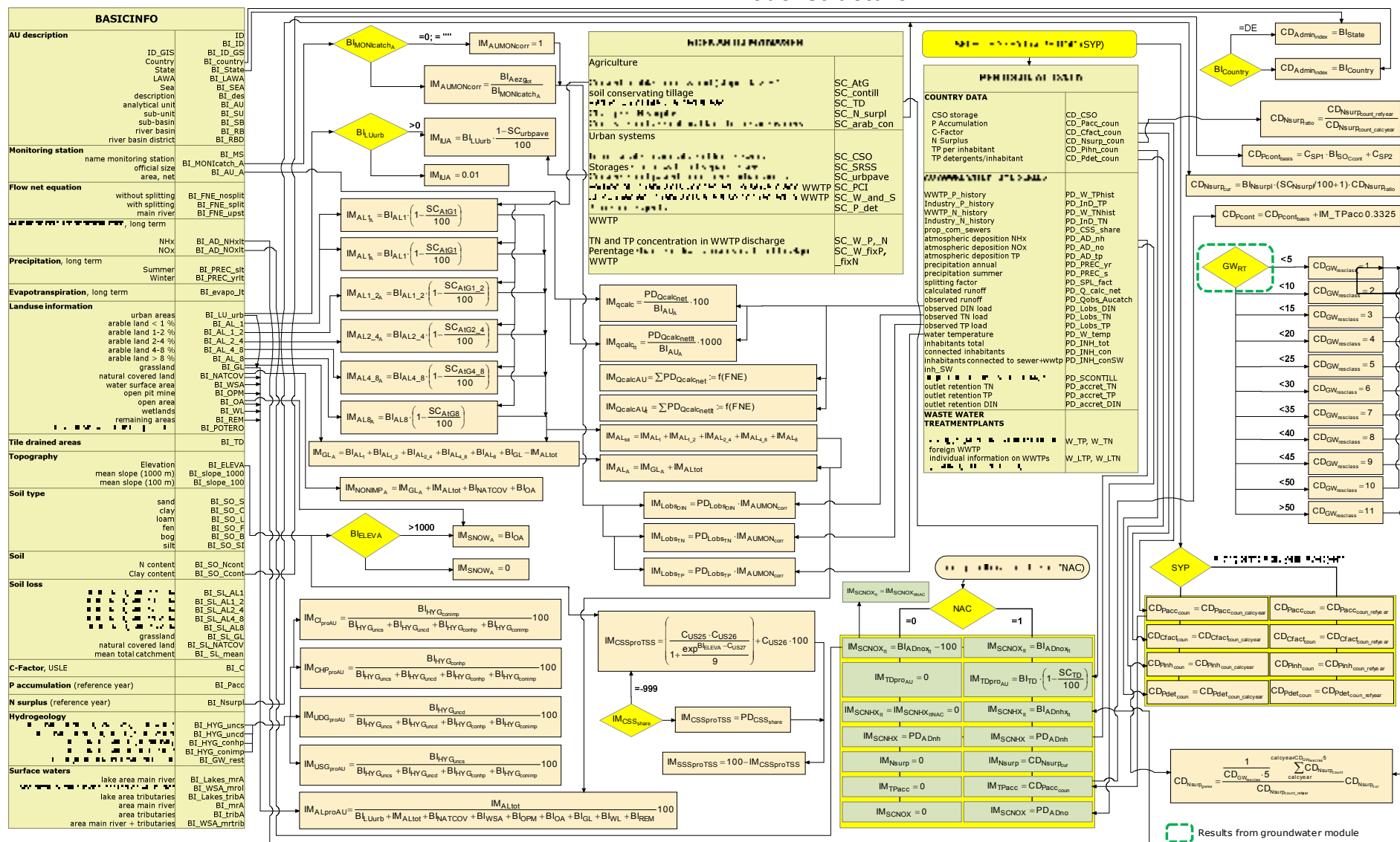


final results or results used by other modules

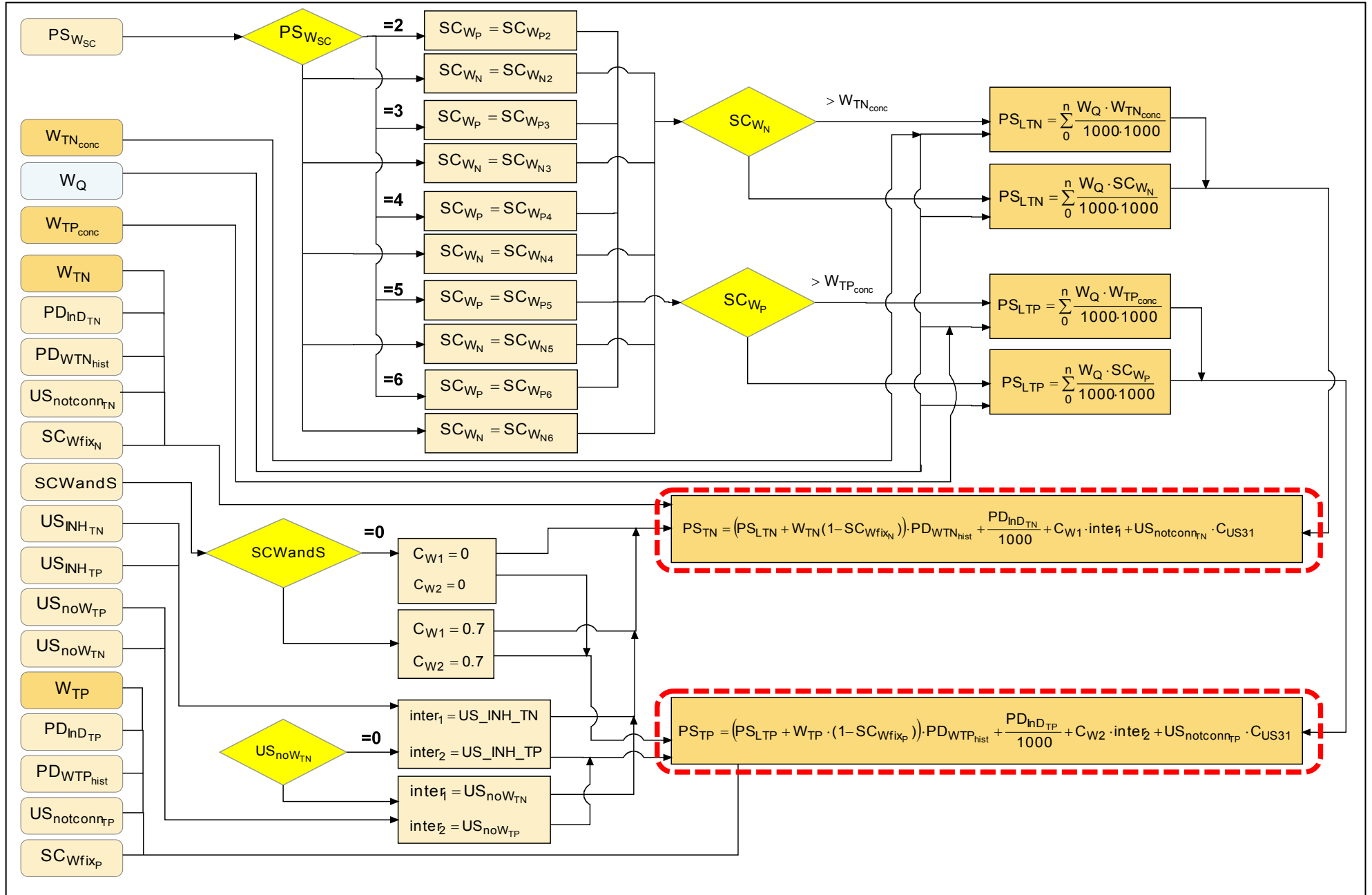
## Surface Water Areas



## Model structure

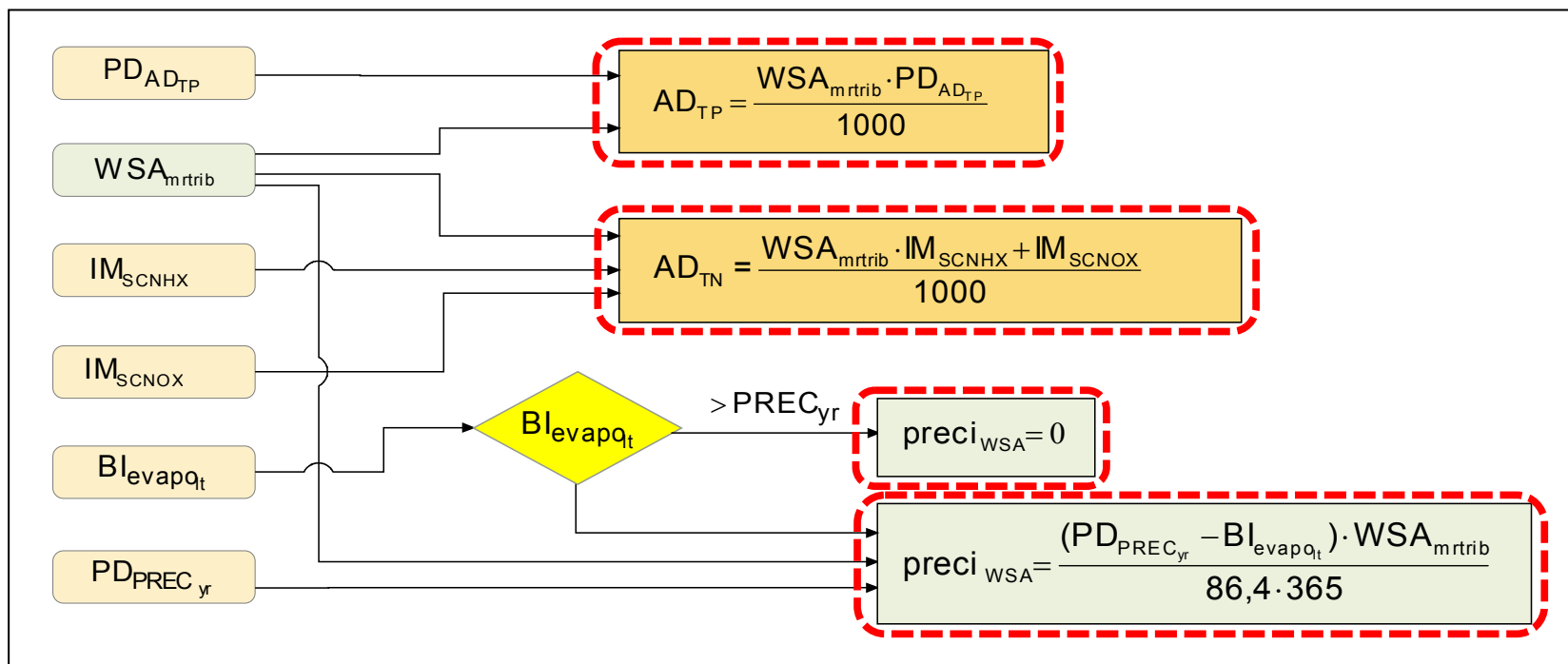


## Point Sources

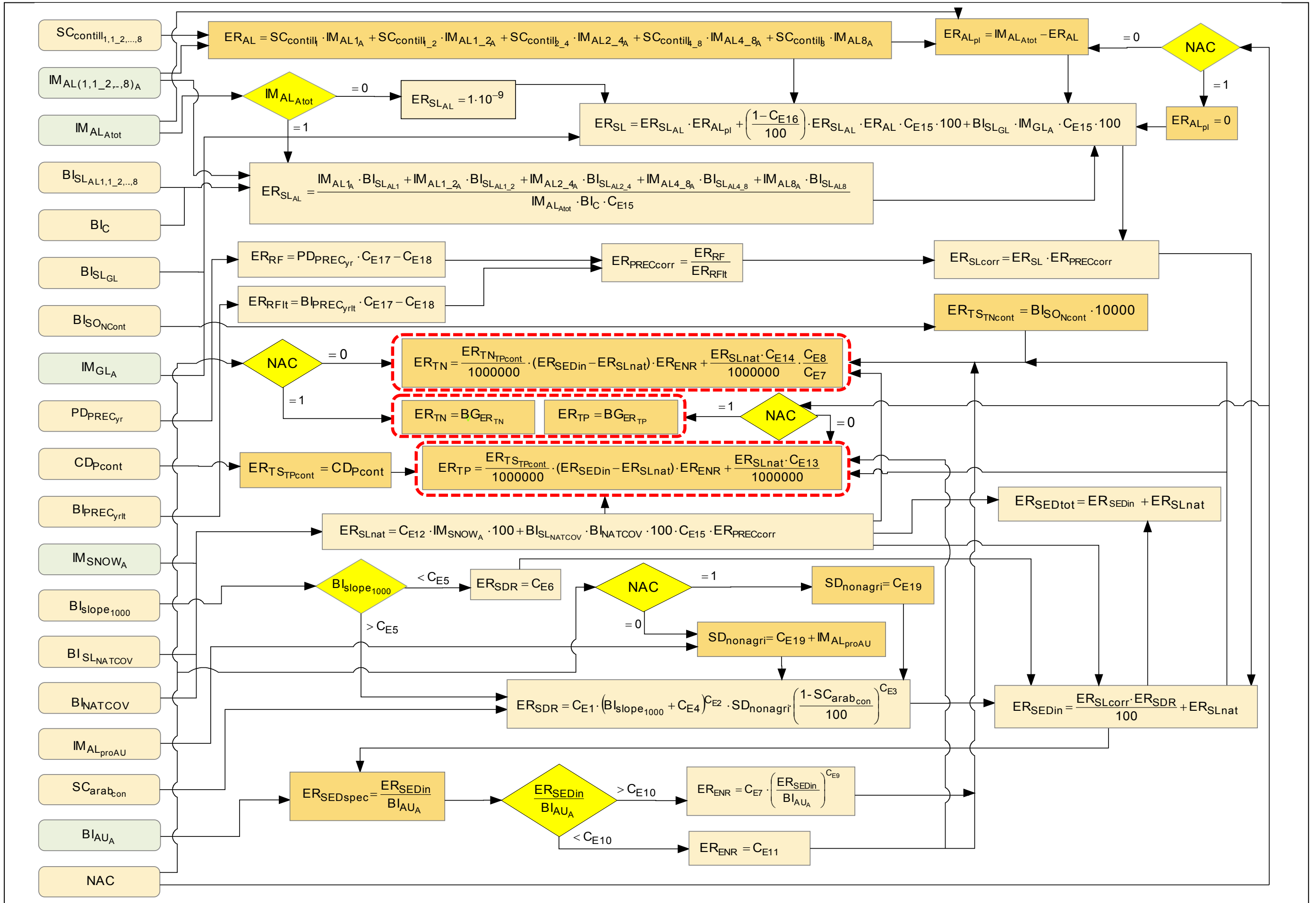




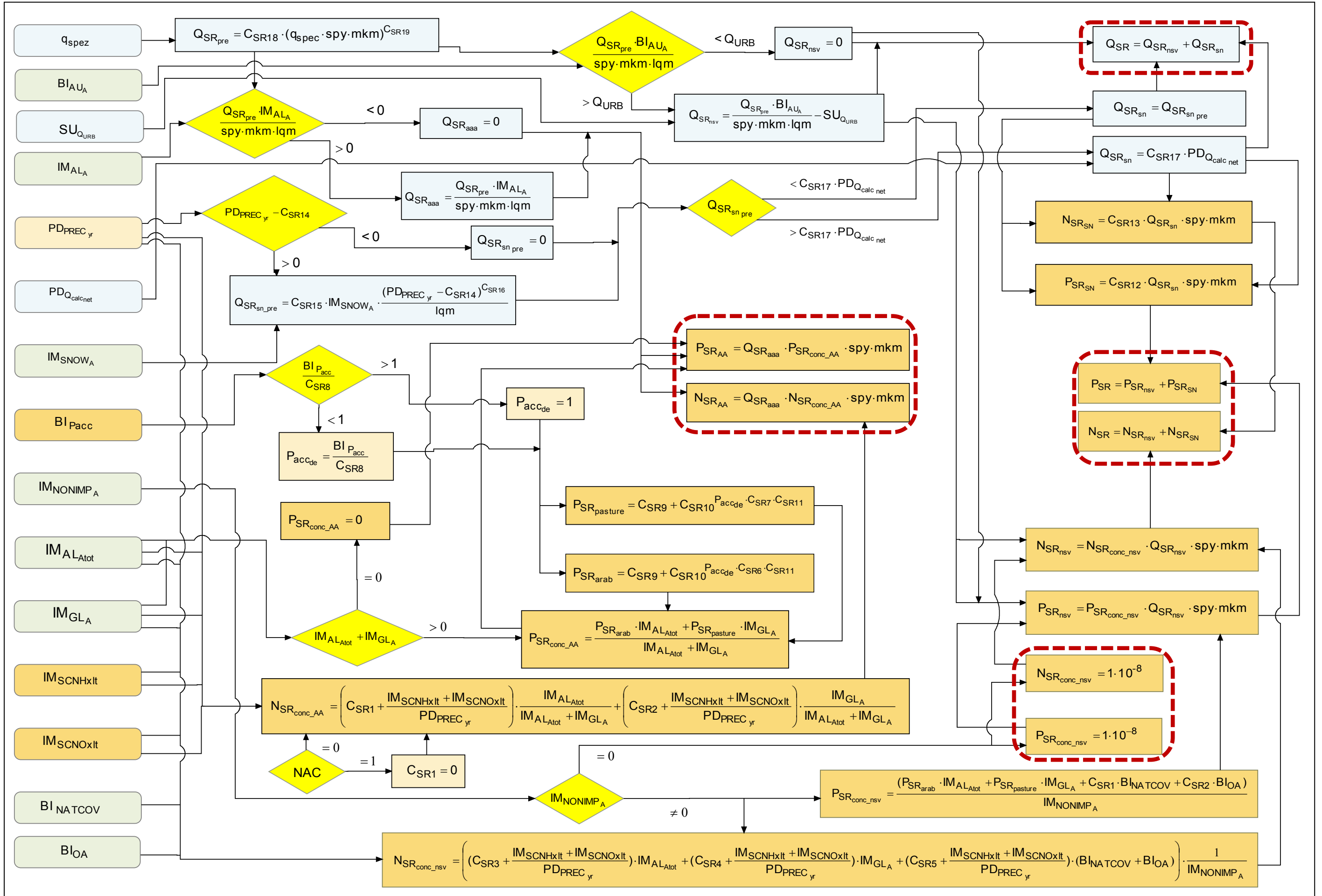
## Atmospheric Deposition rate



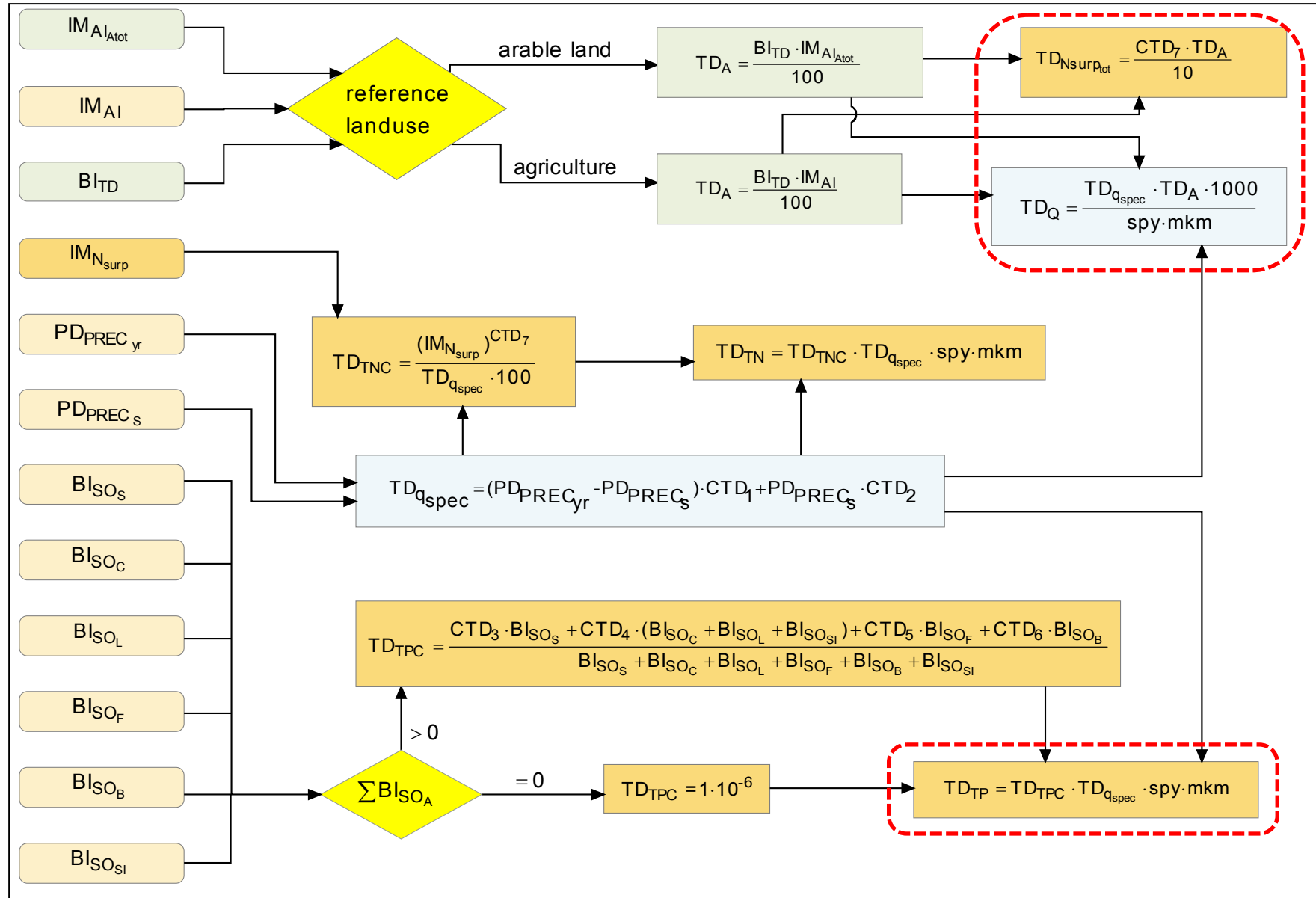
# Erosion



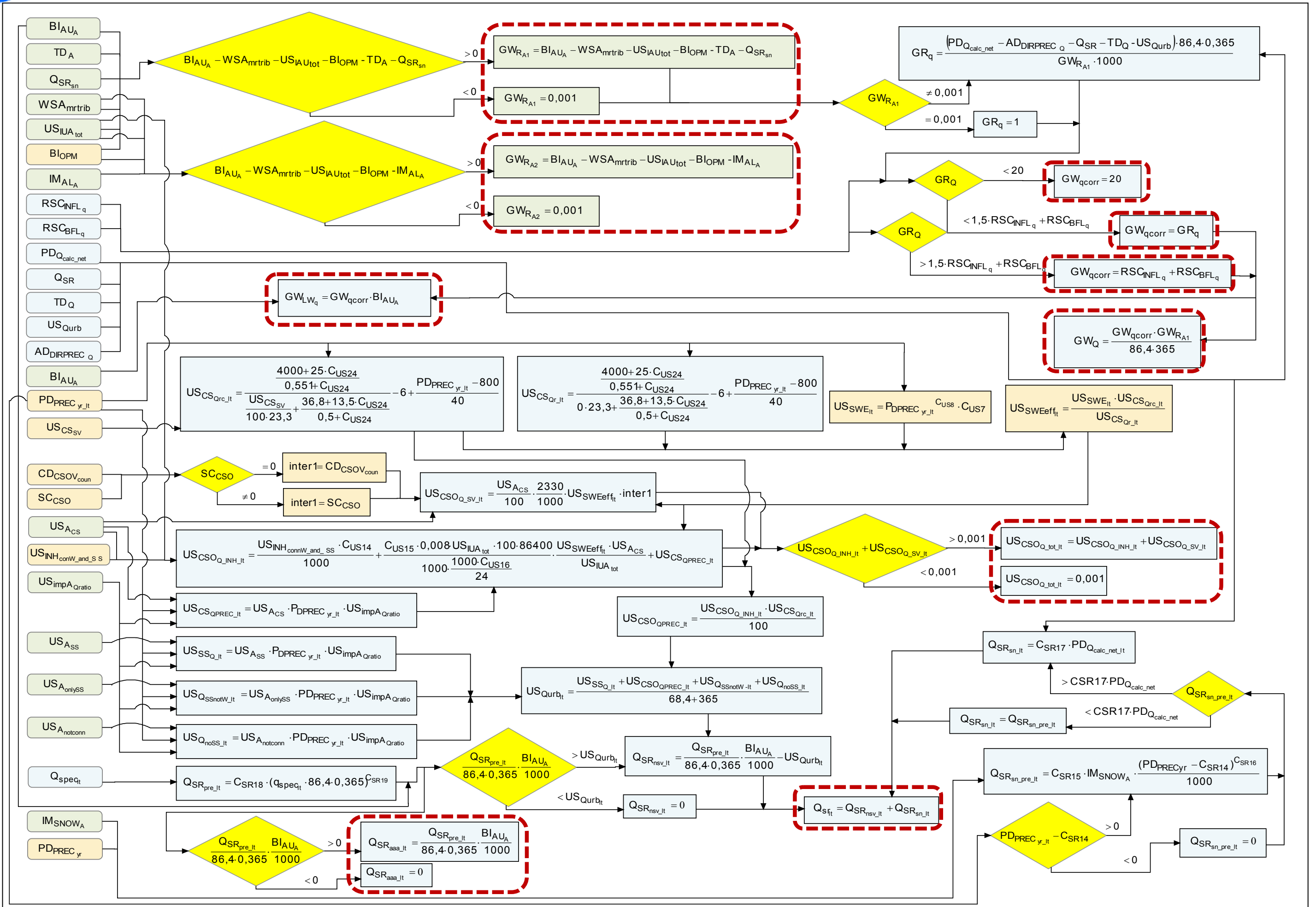
## Overland flow



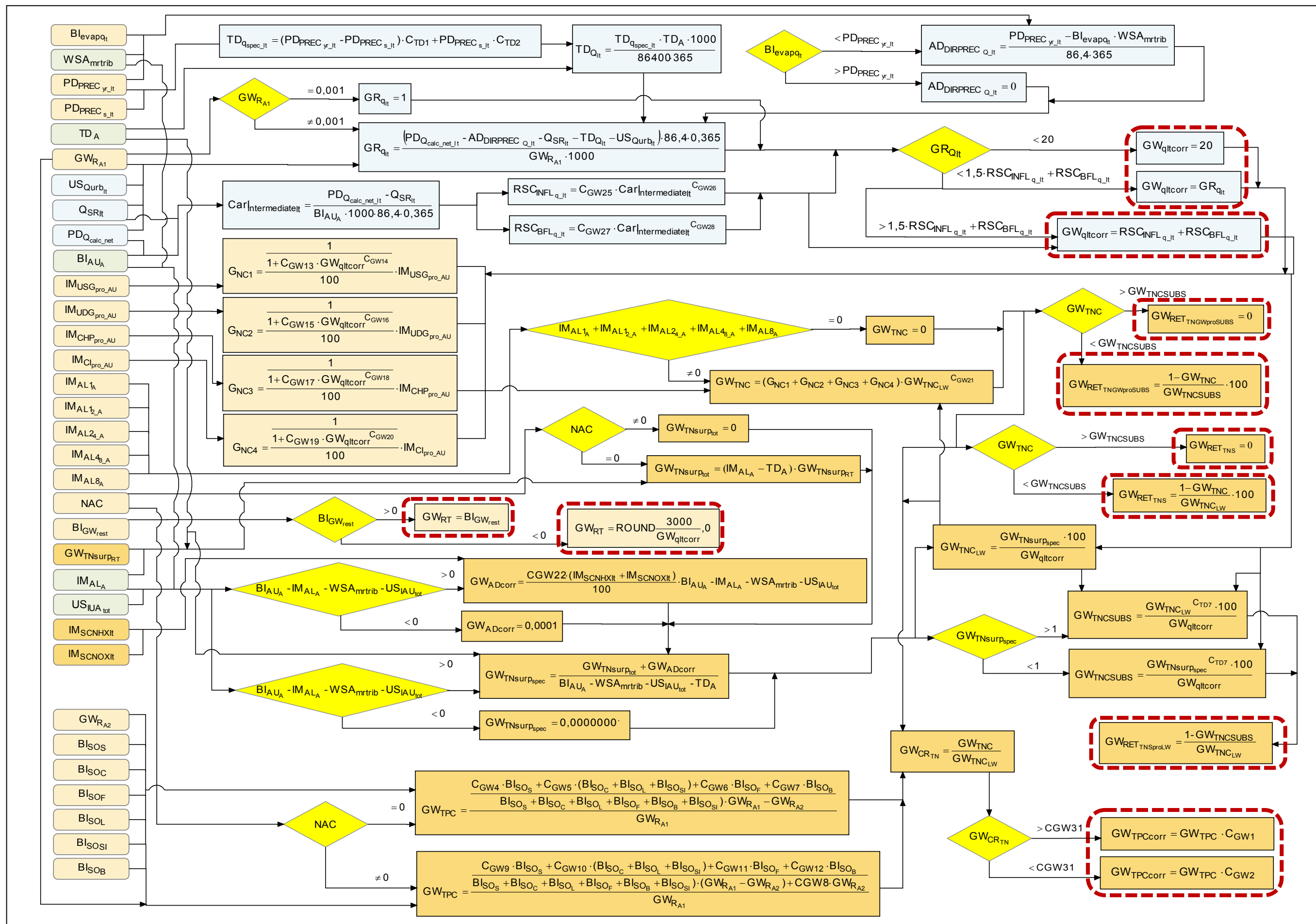
## Tile drainage



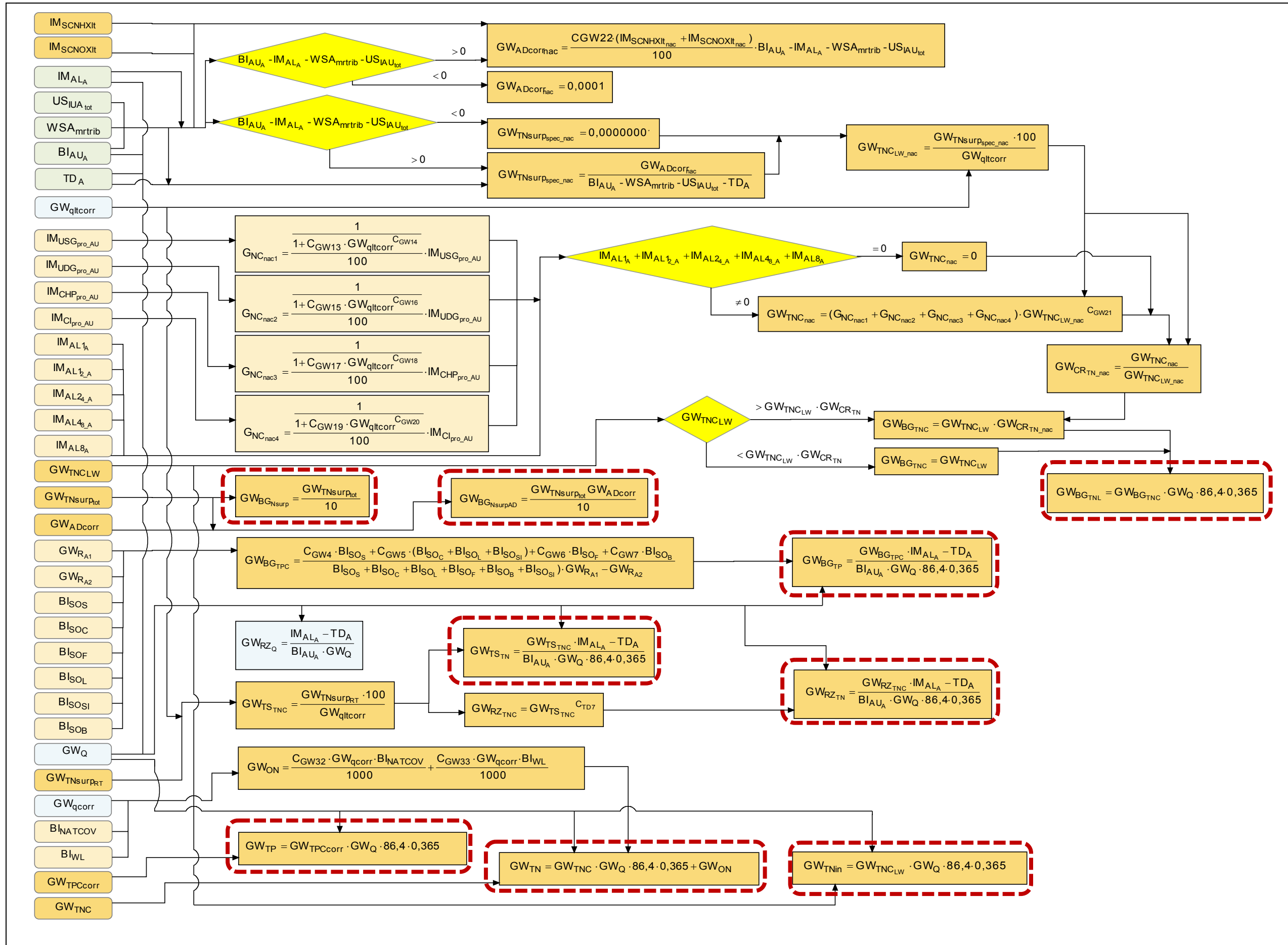
# Groundwater 1



## Groundwater 2

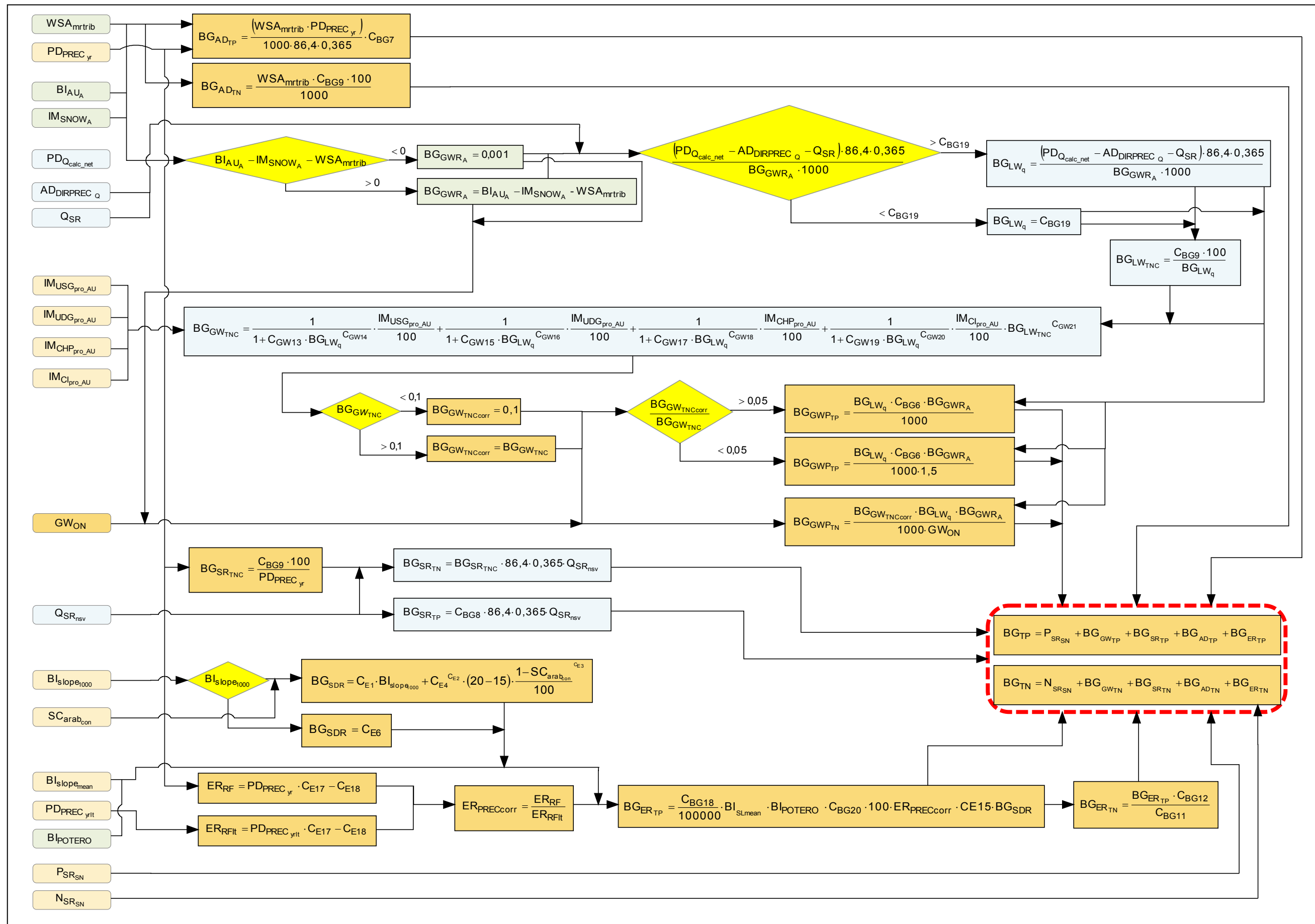


## Groundwater 3

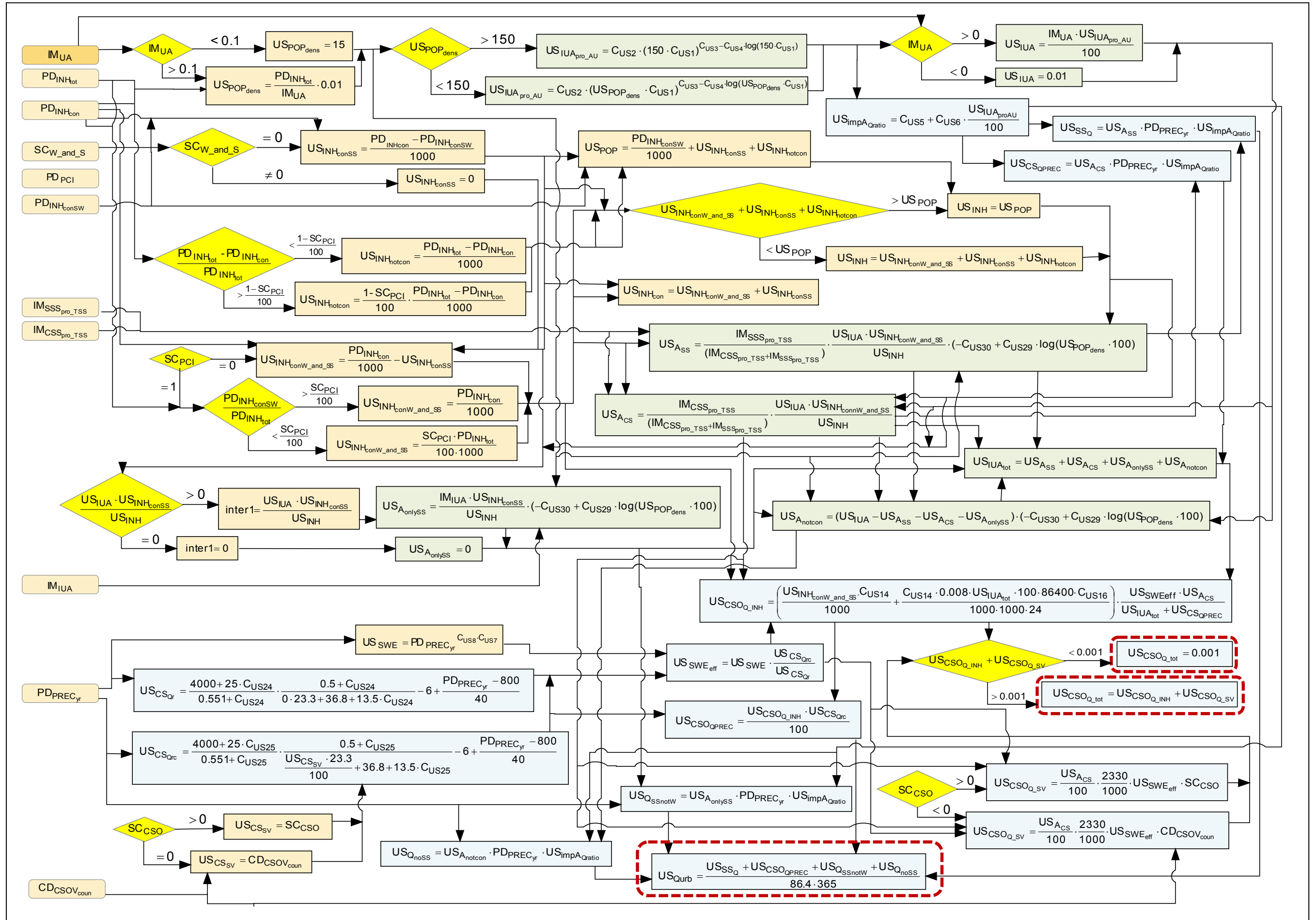


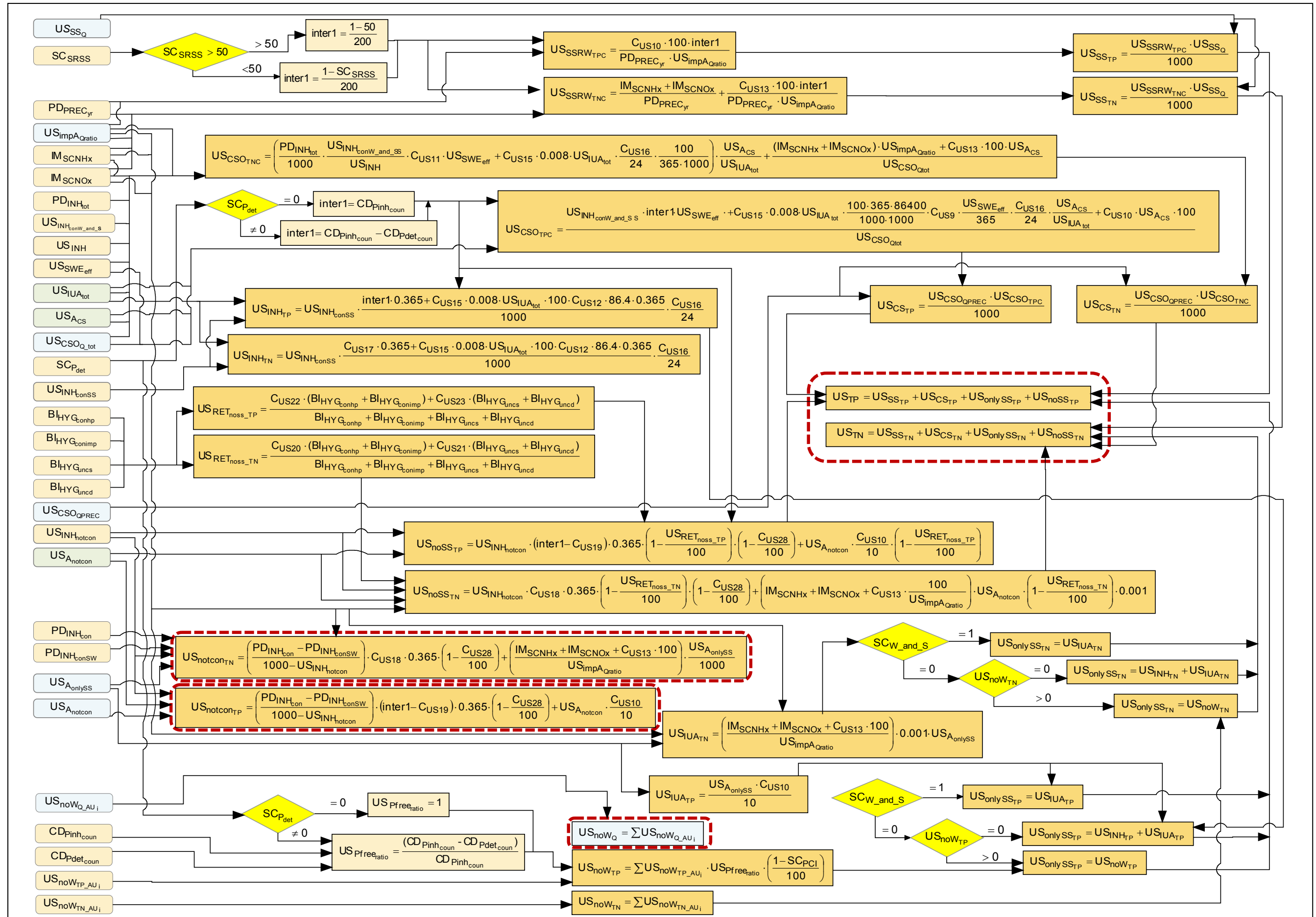


## Natural background condition

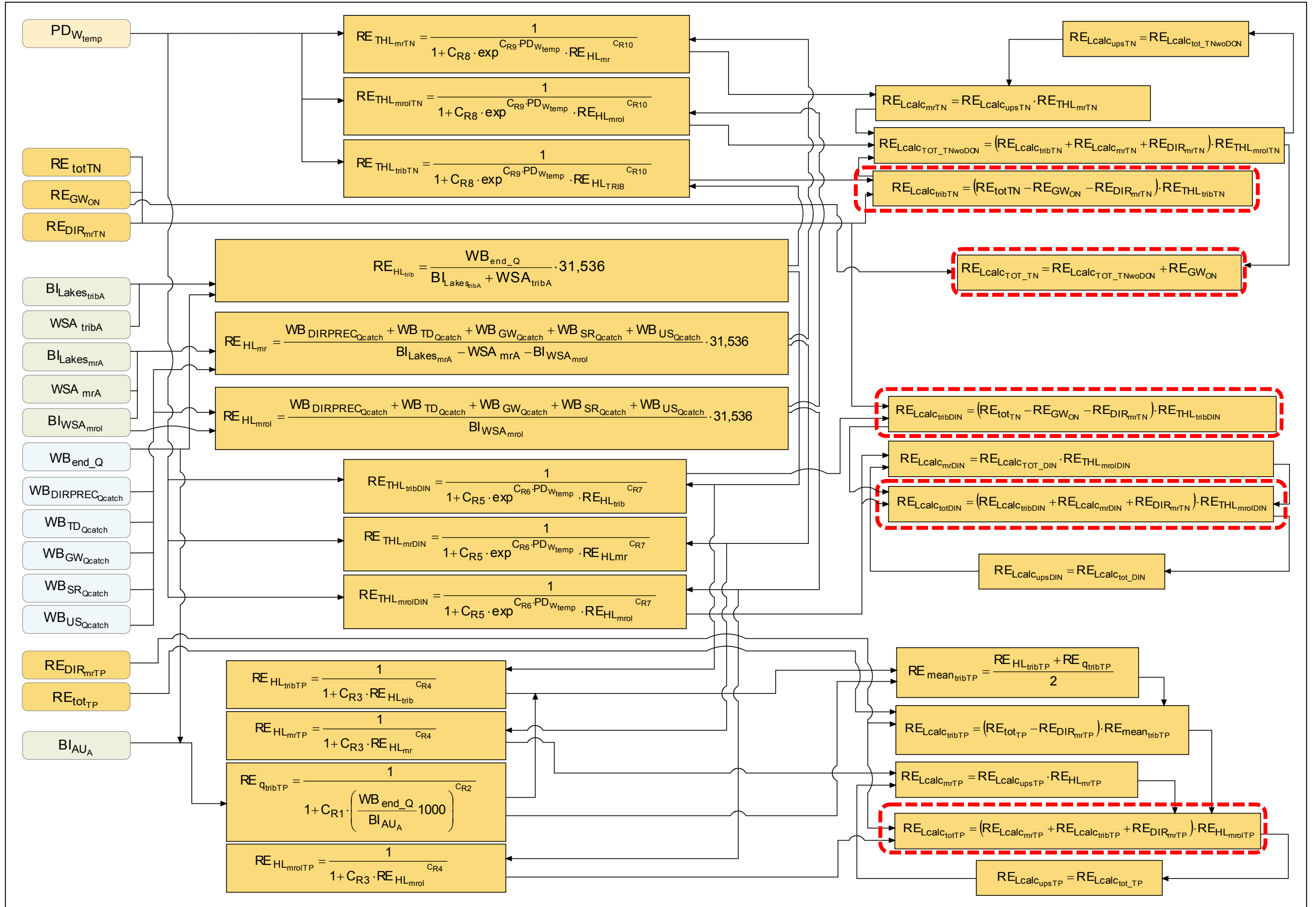


# Urban Systems 1 – areal and runoff calculations

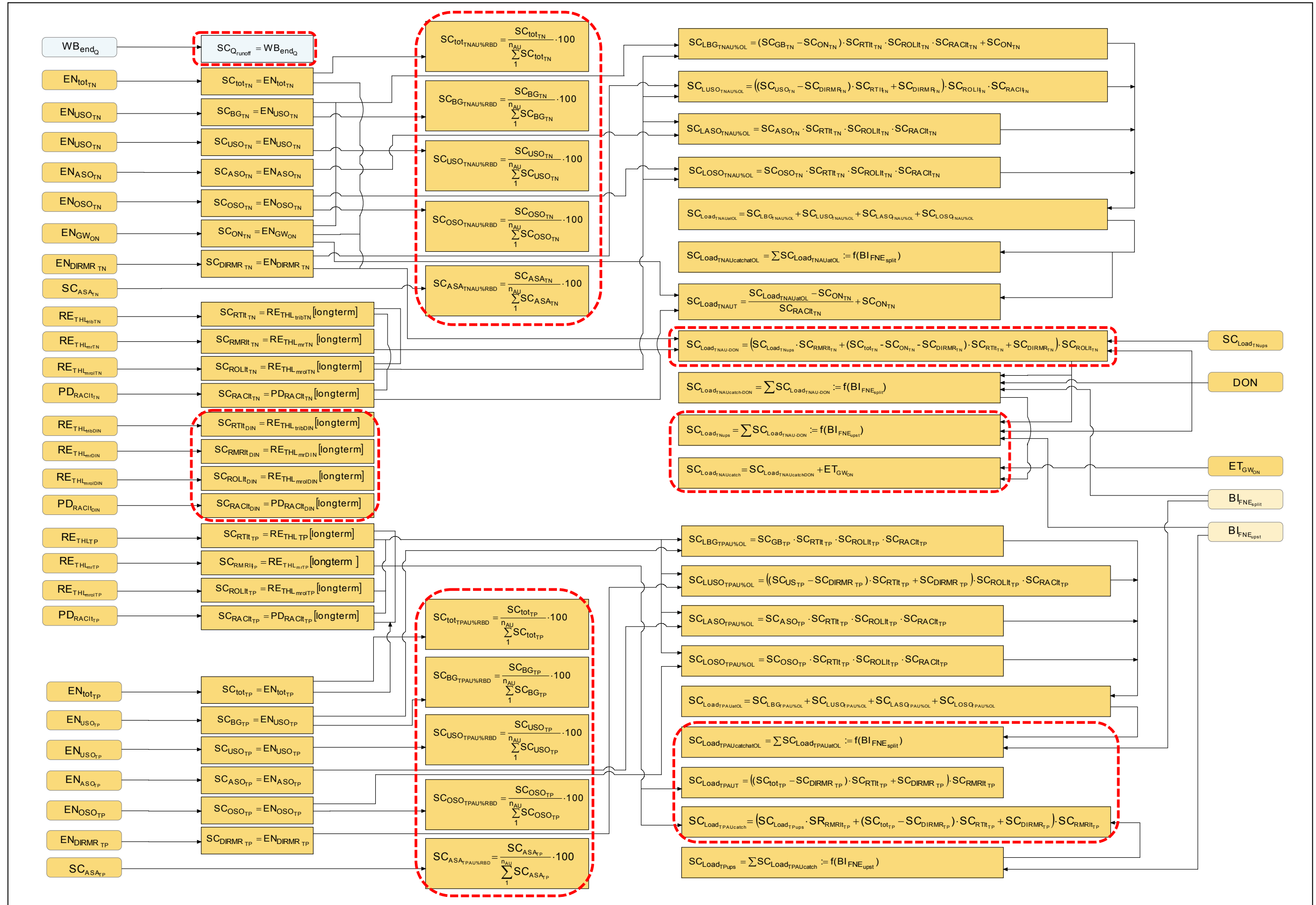




# Retention



## Scenario Results



## Target setting for concentration