National Environmental Research Institute
University of Aarhus · Denmark

NERI Technical Report No. 705, 2009

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State and trends 1998-2003

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Data sheet

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Abstract: This report presents the results of hazardous substances and heavy metals monitoring in the aquatic environment under the Danish National Monitoring Programme of the Aquatic Environment during the period 1998-2003 (NOVA-2003). The report describes the sources and occurrences of the substances in groundwater, watercourses, lakes and marine waters, and estimates the significance of these occurrences. The report is based on the annual reports prepared for each subprogramme by the Topic Centres. The latter reports are based on data collected and submitted by the Danish county authorities.

Keywords: Hazardous substances, pesticides, heavy metals, inorganic trace elements, imposex, environmental state, groundwater, watercourses, lakes, marine waters, terrestrial environment, areas of conservation, atmospheric deposition, wastewater, liquid manure

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Supplementary notes: This report is a translation of a part of NERI Technical Report No. 585. This report gives a general introduction to the Danish monitoring of hazardous substances and heavy metals and summarizes the monitoring results from 1998-2003. In addition, the Danish version of the report goes into details in each group of substances.

Contents

Preface

Hazardous substances and heavy metals have remained high on the environmental policy agenda for many years, the reason being that these substances do not only affect nature and the environment, but are also detrimental to human health.

The Danish Nationwide Monitoring Programme was implemented in 1988 in order to monitor the effects of the Action Plan on the Aquatic Environment I. Consequently, the focus was initially mainly on nutrients and organic substances. When the monitoring programme was revised in 1998 monitoring of hazardous substances and heavy metals was accorded greater priority. The present report summarizes these monitoring results comprised by the NOVA-2003 programme 1998-2003.

Monitoring of hazardous substances and heavy metals encompasses measurements made at the source of the substances in the aquatic environment, including wastewater treatment plants, leaching from cultivated areas and atmospheric deposition. The state and trend of the occurrence of these substances in the aquatic environment comprise monitoring results of groundwater, watercourses, lakes and marine waters. Chapters 1-5 of this report provide an introduction to the organisation and strategy of the monitoring of hazardous substances and heavy metals. Chapter 6 includes a summary and evaluation of the monitoring results. Chapters 7-19, which are only available in the Danish version of this report, provide a detailed description of the results of each individual group of substances. These chapters describe the occurrence, use and environmental characteristics of the substances, and a presentation and assessment of the monitoring results. The monitoring results are evaluated in relation to the established limit values to the extent that these are available.

Summary

The Danish Nationwide Monitoring and Assessment Programme for the Aquatic Environment (NOVA-2003) run over the period 1998-2003. The programme monitored the air, wastewater treatment plants and other point sources, agricultural catchments, the groundwater, watercourses, lakes and marine waters. The Danish monitoring programme was established in 1987 in order to follow development in the aquatic environment and record the effects of the reduction in discharges. In 1998 monitoring of hazardous substances and heavy metals was added to the programme. The monitoring results were reported yearly and the present report gives an overview of the results for the entire period.

The overall picture shows that hazardous substances and heavy metals are generally found in low concentrations and with no environmental significance. The monitoring results have further documented that some areas are impacted or the Danish permitted limits or quality criteria have been exceeded.

Pesticides found most frequently in groundwater and fresh surface water

Pesticides were found in groundwater and fresh surface water while other organic hazardous substances were hardly present. Pesticides were found more frequently in lakes than in watercourses, but concentrations were generally lower in the lakes than in the watercourses.

Accumulation in the marine environment

Among the organic hazardous substances only pesticides occurred in a few cases in watercourses and lakes. Even the substances which were found most often in outlets from wastewater treatment plants or substances discharged in large amounts were found in only a few samples from watercourses and lakes. This applies, for example, to the softener DEHP.

A number of substances were found in sediment, mussels and fish from marine areas. It was typically slowly degradable and accumulative substances such as DEHP, which were found widely in sediment in fjords and coastal areas. Some of the substances which occurred in sediment, mussels and fish are not used in Denmark any longer. DDT is one example. Some of the substances occurred in such high concentration that they may pose an environmental risk.

Heavy metals in the aquatic environment

The amount of heavy metals transported to inland areas and surface waters has been decreasing since the 1990s. In 2003 heavy metal concentrations in fresh surface waters were low compared to the Danish quality criteria, while most metals in marine areas were found in concentrations exceeding the level described by the OSPAR quality criteria as a good environmental state. Concentrations of a few naturally occurring metal

in groundwater were so high that they could pose health problems, if the water is used for drinking water.

Sexual changes in snails

The effect of TBT (tributyltin) in marine waters has been investigated in supplement to the measurements of concentrations of hazardous substances. TBT has been used as antifouling agent in ship painting. The substance acts specifically on snails by causing hormone disturbances and sexual changes. The sexual changes were widespread with the highest frequencies close to harbours, where there was the largest TBT impact.

Sammenfatning

Overvågningen har vist, at helt overordnet er koncentrationen af miljøfremmede stoffer og tungmetaller i de fleste tilfælde lave og uden miljømæssig betydning. Overvågningen har imidlertid også dokumenteret, at der er områder, hvor der er miljømæssige påvirkninger, eller hvor danske grænseværdier og kvalitetskrav er overskredet.

Pesticider mest udbredt i grundvand og fersk overfladevand

I grundvand og fersk overfladevand blev der fundet pesticider, mens de øvrige organiske miljøfremmede stoffer stort set ikke blev fundet. Pesticiderne blev fundet oftere i søer end i vandløb, men koncentrationerne var generelt lavere i søerne end i vandløbene.

Ophobning i det marine miljø

Der blev kun i enkelte tilfælde fundet andre organiske miljøfremmede stoffer end pesticider i vandløb og søer. Selv de stoffer, der er fundet hyppigst i udløb fra renseanlæg eller er udledt i størst mængde, er kun fundet i få prøver fra vandløb og søer. Det gælder eksempelvis blødgøreren DEHP. Til gengæld blev en række af stofferne fundet i sediment, muslinger og fisk i det marine miljø. Der var typisk tale om vanskeligt nedbrydelige og akkumulerbare stoffer, bl.a. DEHP som blev fundet udbredt i sediment i kyst- og fjordområder. Nogle af de stoffer, der blev fundet i sediment, muslinger og fisk, bliver ikke længere anvendt i Danmark. Det gælder bl.a. DDT. I nogle tilfælde optrådte stofferne i så høje koncentrationer, at de kan udgøre en miljømæssig risiko.

Tungmetaller i vandmiljøet

Tilførslen af tungmetaller via nedbør og luft til land- og vandområder har været faldende siden 1990erne. I fersk overfladevand var koncentrationerne i 2003 lave i forhold til danske kvalitetskrav, mens de fleste metaller i det marine miljø blev fundet i koncentrationer, der var højere end de niveauer, der karakteriserer god tilstand i henhold til OSPAR's kvalitetskriterier. I grundvand var koncentrationen af enkelte naturligt forekommende metaller så høj, at de kan udgøre et sundhedsmæssigt problem, hvis vandet anvendes til drikkevand.

Kønsforandringer hos snegle

Som supplement til målingerne af koncentrationerne af de miljøfremmede stoffer er effekten af TBT (tributyltin) i det marine miljø undersøgt. TBT har bl.a. været anvendt i bundmaling til skibe. Stoffet virker specifikt på snegle ved at kunne føre til hormonforstyrrelser og kønsforandringer. Kønsforandringerne blev fundet meget udbredt med størst hyppighed i nærheden af havne, hvor belastningen med TBT har været størst.

Perspektivering

Resultaterne fra overvågningen af miljøfremmede stoffer og tungmetaller har, ud over anvendelsen til at beskrive udviklingen og tilstanden i vandmiljøet, haft en konkret anvendelse i forbindelse med Vandrammedirektivets basisanalyse. Endvidere har resultaterne dannet grundlag for en optimering af overvågningen af miljøfremmede stoffer og tungmetaller i forbindelse med revisionen af NOVA-2003, idet de stoffer der ikke eller kun i ubetydeligt omfang var blevet fundet, blev taget ud af programmet.

Vandrammedirektivet forpligter medlemslandene til at overvåge en række prioriterede miljøfremmede stoffer og tungmetaller. De fleste af stofferne er med i det danske overvågningsprogram. De stoffer der endnu ikke er med i overvågningen, vil først blive inddraget efter at det ved en screeningsundersøgelse er dokumenteret at det er relevant at inddrage dem. Denne strategi anvendes for alle ”nye” stoffer.

Overvågningen af miljøfremmede stoffer og tungmetaller omfatter primært måling af stofkoncentrationer i forskellige medier. Koncentrationsmålingerne var i det marine program i NOVA-2003 suppleret med undersøgelse af effekt i form af kønsforandringer hos snegle. Ved revisionen af NOVA-2003 blev der derudover implementeret enkelte andre effektundersøgelser i det marine program.

1 Introduction and objective

This report provides a summary of the results of six years' monitoring of organic pollutants, heavy metals and other inorganic trace elements comprised by the Danish National Monitoring Programme of the Aquatic Environment during the period 1998-2003, also known as NOVA-2003.

The report was prepared by the National Environmental Research Institute (NERI) in cooperation with the Geological Survey of Denmark and Greenland (GEUS) and the Agency for Spatial and Environmental Planning (former Danish Environmental Protection Agency) based on data gathered by the Danish county authorities and the municipalities of Copenhagen and Frederiksberg. NERI has been responsible for data collection on the atmosphere and open marine waters.

Organic pollutants may be released into the environment through human activities but they can also occur naturally. PAH compounds illustrate this well. PAHs are formed by incomplete combustion of organic material, both by natural combustion and by man's combustion of oil products. PAHs occur naturally, but they occur also in concentrations exceeding the natural level due to man's activities.

The same applies to heavy metals and a number of other inorganic trace elements that are discussed in connection with the heavy metals in this report. Heavy metals and trace elements are part of the minerals of the Earth and are consequently naturally occurring substances. A number of metals and trace elements occur in the environment in elevated concentrations in consequence of man's activities.

Naturally occurring substances – organic as well as inorganic – were encompassed by the monitoring programme, if it was considered that the detected concentrations posed a threat to the environment.

The aim of this report is to provide an overall outline of the occurrence of heavy metals and hazardous substances in the aquatic environment and to give an account of the prospective effects of these substances on the environment, for example by comparing the findings with the determined quality objectives.

2 Measures to combat pollution by heavy metals and hazardous substances

2.1 International measures

At the beginning of the 1990s it was acknowledged that restrictions needed to encompass more substances than had presently been in focus. The Esbjerg Declaration therefore reiterated the objective of reducing discharges, emissions and losses of hazardous substances. Part 17 of the Esbjerg Declaration expresses this as follows:

The prevention of the pollution of the North Sea is to be achieved by continuously reducing discharges, emissions and losses of hazardous substances thereby moving towards the target of their cessation within one generation (25 years) with the ultimate aim of concentrations in the environment near background values for naturally occurring substances and close to zero concentrations for man-made synthetic substances.

At ministerial meetings in 1998, corresponding objectives and more specific strategies for the North Sea and Baltic Sea regions were adopted to follow up the objectives of the Esbjerg Declaration. This was achieved by political consent to the Oslo-Paris Convention (OSPAR) and Helsingfors Commission (HELCOM).

With the adoption of the UNEP's Global Action Plan for the Sea in Washington in 1995 the initiative has been taken for global regulation of the persistent organic pollutants (POPs).

The international marine agreements are explained in more detail in Dahllöff & Andersen (2009).

2.2 Danish initiatives

Parallel with international efforts Denmark has taken additional initiatives to reduce the use of pesticides. The objective of the first Pesticide Action Plan in 1986 was to reduce the total pesticide consumption and pesticide application frequency by 50% before 1 January 1997. Furthermore, the consumption was to be steered towards the use of less harmful pesticides. In a status for the Pesticide Action Plan in 1997, it was concluded that the total sales of active substances had decreased by 36%, but that pesticide application frequency remained almost unchanged. In 1999, total sales of active substances had decreased 59% relative to sales in 1986. The objective of Pesticide Action Plan II from 2000 was to reduce loading even further by establishing, for instance, pesticide-free border zones along watercourses and lakes for which a quality objective has been set and to areas identified as being particularly susceptible to pesticides (Miljø- og Energiministeriet og Ministeriet for Fødevarer, Landbrug og Fiskeri 2000). The latest Pesticide Action Plan 2004-2009 focuses on a continuous reduction in pesticide consumption and pesticide treatment frequency.

In 1998, the Danish Environmental Protection Agency published the first national list of undesirable substances. The list serves as a signal and guideline to manufacturers, product developers, purchasing agents, and other parties within the chemical industry, indicating substances whose use should be limited or eventually phased out. The list was subsequently updated in 2000 and 2003 (Miljøstyrelsen 2004a). A number of undesirable substances have been included in the NOVA-2003 monitoring programme.

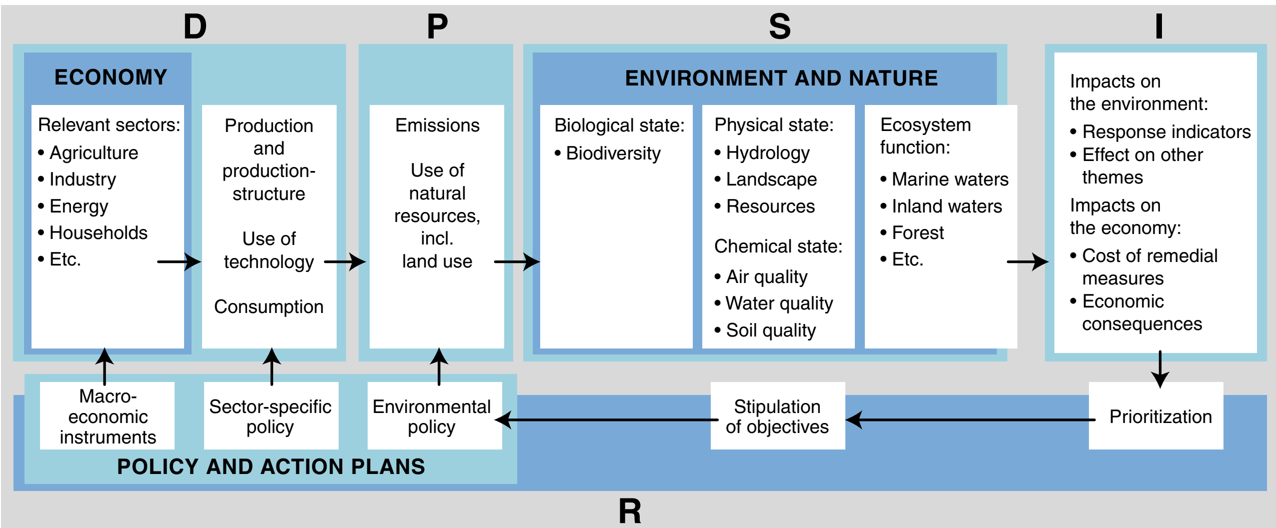
3 Monitoring of the aquatic environment

The purpose of including hazardous substances and heavy metals in the monitoring programme was to assess the occurrence and trend of hazardous substances and heavy metals in groundwater, inland waters and marine waters. The monitoring encompassed sources such as, for example, point sources and air as transport pathway.

Monitoring was implemented in order to comply with Danish obligations in relation to international directives and conventions, and the need to monitor the effect of the implemented Danish action plans.

3.1 Requirement-controlled monitoring

The monitoring programme was established on the basis of the so-called DPSIR concept (figure 3.1). The idea behind the concept is to describe quantitatively the relations between the various activities of society that involve use of the substances, emissions and discharge of substances to the surroundings, and finally the effect of the measures implemented to avoid adverse environmental impact.



Figur 3.1. DPSIR diagram describing the principles of the environmental correlations and decision-making process with a knowledge-based framework. DPSIR stands for driving forces, pressures, state, impact and responses.

The results of the NOVA-2003 monitoring of hazardous substances and heavy metals are encompassed in the documentation of the achieved effect on the environment of the implemented action plans and measures. Around one third of the approximately 260 monitored substances are related to the international conferences on the protection of the North Sea, OSPAR or HELCOM, and just under one third of the substances are related to the Drinking Water Directive. Additional substances were monitored because of stipulated quality criteria on surface water. Finally, a number of substances were included in order to determine the background level in groundwater or to gather knowledge about the occurrence of substances that may pose a threat to the environment. The NOVA-2003 Programme Description, annex 1 (Miljøstyrelsen 2000), describes the selection criteria for each individual substance. A total list of the current obligations to monitor and report the substances in relation to Danish legislation or international agreements can be seen on NERI's website (www.dmu.dk) (Miljøstyrelsen 2001a).

Monitoring of hazardous substances and heavy metals in NOVA-2003 mainly focuses on pressure (P) and state (S) and less extensively on impact (I), cf. figure 3.1.

3.2 Overview of the monitoring programme 1998-2003

The heavy metals lead, cadmium, copper, mercury, nickel and zinc were encompassed by all the individual parts of the monitoring programme, while inorganic trace elements were only monitored in groundwater.

The comprehensiveness of organic hazardous substances monitoring was mainly determined by the physical-chemical properties of the substances. Substances with lipophilic[1] characteristics were primarily monitored in sewage, sediment and biota, while substances with hydrophilic[2] characteristics were monitored in wastewater, surface water and groundwater.

The extent of monitoring has varied according to the various types of matrices (table 3.1). Sampling took place more frequently in matrices with great temporal variation, for example watercourses, where measurements were made 6-12 times yearly. Other areas of the environment with less pronounced variation, for instance marine sediment, were sampled twice during the 6-year programme period.

3.3 Wastewater and sewage

Monitoring of hazardous substances and heavy metals in wastewater has been planned primarily with a view to producing an overview of the inputs to water bodies from various types of point sources and to complying with international commitments. Monitoring encompassed measurements at wastewater treatment plants, enterprises with individual discharges, and stormwater outfalls from combined and separate sewerage systems. Monitoring also encompassed data from freshwater fish farms and saltwater fish farms and discharges from sparely built-up areas.

The measured contents of heavy metals and hazardous substances in wastewater and sewage are presented in Annex 1 as means of the concentrations measured at the monitored wastewater treatment plants.

[1] Lipophilic: fat loving. Lipophilic substances are soluble in fat and most organic solvents. The substances are to a large extent bound to particles.
[2] Hydrophilic: water loving. Hydrophilic substances are water soluble.

Table 3.1. Overview of the NOVA-2003 Monitoring Programme for hazardous substances, heavy metals and other inorganic trace elements for the period 1998-2003

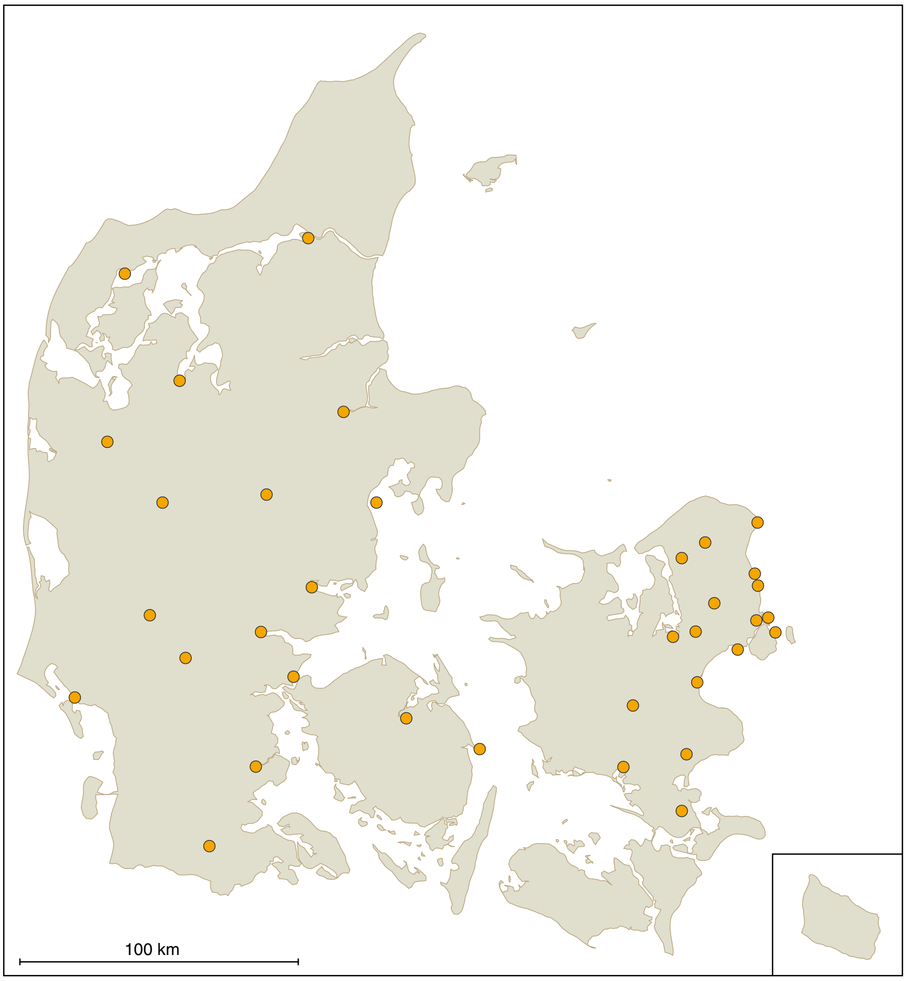
	Point sources			Organic fertilizer	Atmos-phere	Ground-water	Water-courses	Lakes	Marine waters		
	Waste water	Sewage	Stormwater outfalls						Seawater	Sedi-ment	Biota
Heavy metals	X	X	X	X	X	X	X	X	X	X	X
Inorganic trace elements						X					
Pesticides	X	X	X	X		X	X	X	X		
Phenols and chlorophenols	X	X	X	X		X	X	X	X	X	X
Alcylphenols	X	X	X	X		X	X	X		X	
Plasticizers	X	X	X	X		X	X	X		X	X
Anionic detergents	X	X				X	X	X	X		
Solvents	X	X	X			X	X		X		
Ether (MTBE)	X	X	X			X	X	X			
PAH	X	X	X	X			X	X		X	X
P triesters	X	X		X							
Aliphatic amines	X	X									
PCB	X	X								X	X
Chlorinated pesticides	X	X				X	X			X	X
Dioxines and furans		X									
Organotin compounds										X	X
Medicines				X							

About one third of the substances monitored at the 38 wastewater treatment plants are prohibited, partly prohibited, or encompassed by voluntary agreements or the POP[3] convention. Some of these regulatory measures have been implemented quite recently and have therefore not yet had an effect on the monitoring results. About a quarter of the substances were regulated by legislation introduced before the beginning of the NOVA-2003 period, i.e. before 1998.

As a general rule there were two monitoring rounds at each of the investigated treatment plants: the first round during the period 1998-2000 and the second round during the period 2001-2003. One monitoring round included analyses of four weekly composite samples each consisting of five 24-hour samples. The substance content was determined from analyses of the weekly mixed sample. Consequently, the results reflect the mean discharges over time with no details of the maximum concentrations.

[3] POP Convention (Persistent Organic Pollutant). International convention whose purpose it is to prohibit production and consumption of 12 persistent organic substances.

Figure 3.2. Location of the 38 wastewater treatment plants encompassed by the monitoring programme for hazardous substances and heavy metals.



Data treatment

Total mean values were calculated for the concentration of substances in outflow and sludge from all treatment plants. If a substance was detected at five or fewer plants, it was not included in the data presentation. If more than 10% of the analysis results for a substance were below the detection limit the mean value was not calculated.

The total national mean value of the outflow concentration was calculated as a mean value from the individual treatment plant and subsequently as a national mean value based on all these treatment plants. The mean values have not been weighted in relation to the size of the treatment plant, as it has been shown not to affect the level at the national scale. The 95% percentile for concentrations in outflow and sludge has been calculated on the basis of the deviation in the mean values for the individual plants.

Analyses results below the detection limit have been included on the basis of the following two criteria:

- if more than 50% of the analyses are above the detection limit, the results below the detection limit have been included as ½ * the detection limit
- if less than 50% of the analyses are above the detection limit, results under the detection limit have been included as zero.

Where the analysis result has been included as zero when calculating the mean value it will most probably result in a slight underestimation.

Very high mean concentrations of a few substances have been detected at a few plants. As a result, there is a large deviation in the calculated national mean value, and the mean value is very high compared to the median value. When the calculated mean value exceeds the median by a factor 10, the mean value for the substance in question is indicated with a star in the data presentation in Annex 1.

The total annual discharge of substances from wastewater treatment plants has been calculated by multiplying the average of the national mean values for the substances for the period 1998-2003 by the mean total annual discharge of wastewater. The calculations are subject to considerable uncertainties.

Enterprises with separate industrial discharges

Monitoring of enterprises with separate industrial discharges to the aquatic environment encompassed discharges from 14 enterprises, including discharges from landfill sites and preventive arrangements in connection with soil pollution incidents. The discharges were to the widest possible extent selected so as to give a representative estimate.

Two monitoring rounds were performed at each enterprise. Each monitoring round comprised analyses of 4 1-week composite samples, each consisting of 5 diurnal samples. In addition, county supervision data and enterprise in-house control data were incorporated in the calculation.

The methodology applied to calculate the level of substances discharged from the 14 enterprises was the same used in connection with the wastewater treatment plants as an average for the period 2001-2003. The calculations are subject to considerable uncertainties as enterprises with separate industrial discharges are not included in the monitoring programme every year.

Stormwater outfalls

Stormwater outfalls have been monitored with the purpose of providing an overall assessment of the impact of these outfalls on the loading of the water bodies. During the period 1998-2000 outfalls from a catchment with a combined sewerage system were monitored in Nordjylland County (Frejlev), and outfalls from a catchment with a separate sewerage system in Nordjylland County (Sulsted) and a catchment with a combined sewerage system in the Copenhagen Municipality (Toftøjevej) were monitored from 2001 to 2003.

The estimated discharged substances from stormwater outfalls from areas with combined sewerage systems are based on measurements taken in the catchment with a combined sewerage system Nordjylland County multiplied by the total annual discharged outfalls from all catchments with combined sewerage systems.

Sparsely built-up areas

The total discharge of heavy metals from sparsely built-up areas has been estimated on the basis of information of the number of properties, the anticipated PE (person equivalents) per property, treatment measures at the individual property and the expected degrees of treatment. The number of heavy metals per PE has been estimated on the basis of measurements from nine wastewater treatment plants receiving only domestic sewerage. The scientific basis for making similar estimates for hazardous substances is still insufficient.

Freshwater fish farms

Information of the total consumption of medicine and auxiliary substances was collected from the freshwater fish farms, while only the total consumption of medicines was available from sea-based fish farms.

3.4 Air

Monitoring of the air in NOVA-2003 comprised monitoring of heavy metals in wet deposition and the atmospheric content of particle-bound heavy metals. Monitoring has to the possible extent been performed in accordance with the common European guidelines (EMEP[4]).

Six main stations were monitored and the wet deposition was measured at an additional three stations (figure 3.3). The monitoring stations were located so as to provide a reasonable geographic coverage. Based on the time series, the overall trends, and consequently also the effect of implemented measures to reduce emissions, are assessed. It is presumed that the mean deposition on the monitoring stations is representative for that of the Danish landmass and aquatic environment. The total deposition is calculated as the sum of the wet and dry deposition.

Wet deposition of heavy metals

Wet deposition was measured by collecting the total precipitation for one month and analyse it for heavy metal content. The annual wet deposition was calculated as the sum of the monthly depositions.

Dry deposition of heavy metals

The content in the air of particle-bound heavy metals was collected with filterpack collectors at 6 monitoring stations, where the atmospheric content of particles was collected for 24 hours by sucking air through particle filters. The diurnal samples were collected throughout the year.

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[4] Co-operative programme for Monitoring and Evaluation of the Long-range Transmissions of Air Pollutants in Europe (see e.g., Ellerman et al. 2004)

Dry deposition of the particle-bound heavy metals is calculated on the basis of the monitored amounts of heavy metals on the filters and the calculated rate of deposition of atmospheric particles, assuming an average particle diameter of 0.8 µm (the dry deposition module of the ACEDP model is described by Ellerman et al. 2005).

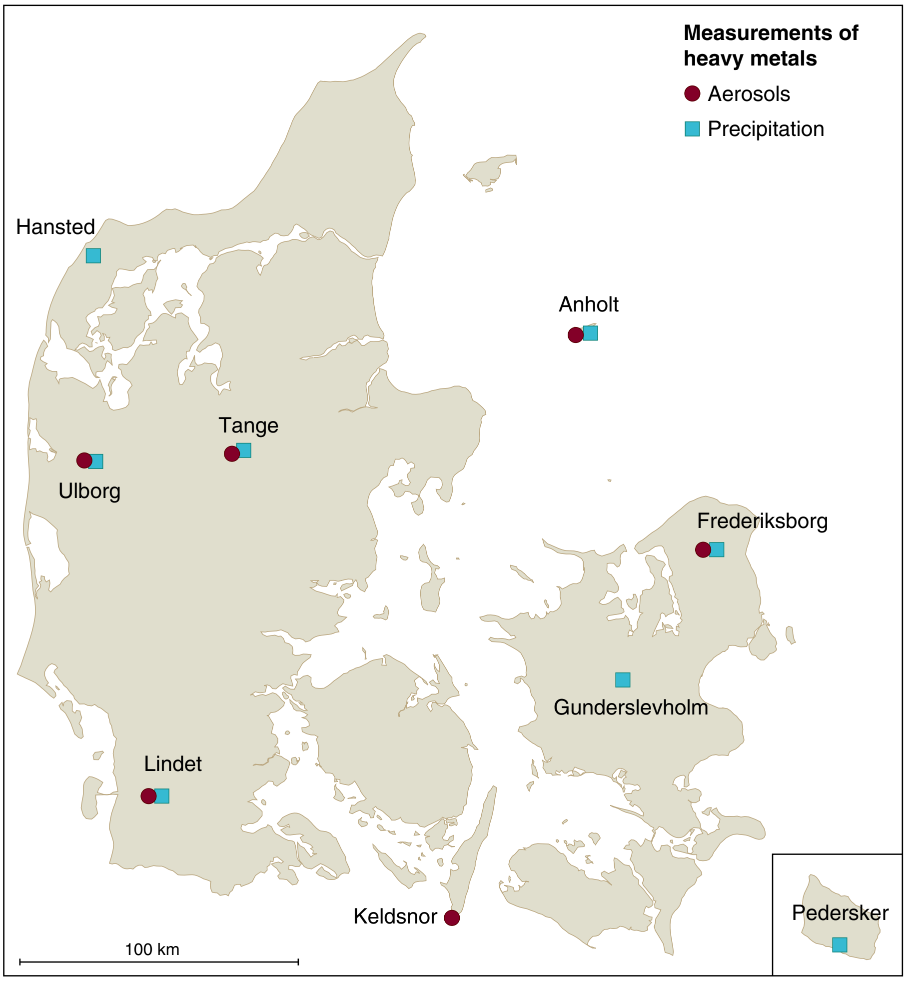


Figure 3.3. Monitoring stations for atmospheric content of heavy metals in particles (aerosols) and in precipitation (wet deposition) (Ellermann et al. 2004).

The atmospheric annual mean concentrations are determined by calculating the average of the diurnal results. For chrome and cadmium, where concentrations frequently were below the detection limit, the annual mean concentrations are, however, calculated after the data have been transformed to log-normal distribution of the measurements above the detection limit (typically 40-160 measurements per monitoring station).

3.5 Agricultural catchments

Monitoring of the agricultural catchments (LOOP) comprised monitoring of pesticides in soil water, drainage water, groundwater and watercourses. Monitoring of heavy metals and organic pollutants in groundwater and measurements in liquid manure were also included.

Figur 3.4. Groundwater monitoring sites (GRUMO) and agricultural catchments (LOOP) in NOVA-2003.

LOOP monitoring took place in five small agriculturally dominated watercourse catchments of 5-15 km^2 each. The catchments were selected so as to represent the national average as far as possible with regard to soil type, climate, types of farms, number and distribution of livestock and crop mix. The catchments were situated in the counties of Nordjylland, Århus, Sønderjylland, Fyn and Storstrøm (figure 3.4).

Monitoring programme

The strategy for monitoring hazardous substances and heavy metals in LOOP has been to monitor the destiny of the substances from the time they were applied until they appeared in the various stages of the hydrological cycle.

Information has been obtained from interviews of approx. 160 farms within LOOP during the period 1993-2004 regarding application of pesticides on fields corresponding to 5000 ha agricultural land. Additional information about the consumption of the previous year was gathered for a number of fields where samples of the groundwater below the field have been taken.

Pesticide monitoring in watercourses and drainage water was carried out with the majority of the samples being taken during the spraying season when the pesticide concentrations are presumably highest. Monitoring of the other substances in drainage water and watercourses (heavy metals and other hazardous substances) took place on a monthly basis. These results have been combined with data from the rest of the monitoring programme for watercourses, cf. Section 3.7.

Limited investigations of liquid manure were carried out in 2001 followed by a more comprehensive investigation in 2002. In 2002, 45 samples of liquid manure from four different stock types in the 5 LOOP catchments were examined: 17 samples from conventional cattle holdings, 8 samples from organic cattle holdings, 18 samples from conventional pig farms and 2 samples from a mixture of conventional farms. The results have been assessed on the basis of the detection frequency and the mean concentration in samples with concentrations above the detection limit.

Monitoring of groundwater in LOOP catchments comprised subsurface groundwater at a depth of 1-5 m.

3.6 Groundwater

Monitoring of groundwater comprised 70 groundwater monitoring sites (GRUMO) with more than 1100 intakes, and approx. 120 intakes in five agricultural catchments, cf. section 3.5. The areas are distributed across the country and widely represent the different geological conditions in Denmark (figure 3.4). The results from the well controls at the approx. 10,000 waterworks wells have also been included (Miljøministeriet 2001).

The purpose of groundwater monitoring in Denmark is to ensure a continuous built-up of knowledge of the groundwater quality and its suitability for drinking water purposes, and knowledge of how much the groundwater quality contributes to the pollution of the water bodies and wetlands to which the groundwater flows. The occurrence of hazardous substances is particularly undesirable in drinking water, and for some substances the limit values are identical with the detection limits that were achievable at the time when the limit values were set. For some of the heavy metals and inorganic trace elements that occur naturally in groundwater aquifers, the limit values for drinking water are typically determined on the basis of health considerations.

The objective of monitoring of heavy metals and inorganic trace elements is to describe the general state, and to demonstrate the influence of the different geological conditions and the land use on the groundwater quality. Finally, monitoring should document occurrence, concentration and trend of non-naturally conditioned high concentrations in groundwater. All the substances included in the monitoring programme have been were widely used in Denmark or occur naturally.

The groundwater monitoring strategy was based on the classification of groundwater as young or old, based on tritium or CFC dating[5]. “Old” groundwater, i.e. groundwater formed before 1950, has typically only been tested for organic environmental hazardous substances once during the period 1998-2003, as these substances were assumed not to have been

[5] Dating of age based on the isotope composition in CFC gasses.

used at the time when the groundwater was formed. Old groundwater has been tested for naturally occurring substances twice during the same period.

"Young" groundwater may be affected by the activities of last 50 years and has typically been investigated at least twice during the period 1998-2003. Certain substances, such as e.g. pesticides, have been investigated yearly or several times a year. In the agricultural catchments with near-surface groundwater intakes samples were collected up to four times per year.

The waterworks wells have been subject to routine controls in the individual well every third to fifth year, depending on the volume of the water abstraction.

Result assessment

The results from the groundwater monitoring programme in GRUMO, LOOP, and the waterworks' well controls have been described on the basis of the detection frequency of the individual substances in the investigated filters, and the median/maximum concentrations for the period 1998-2003. The detection percentage is calculated as percentage findings above and under the limit value in drinking water to the extent that limit values have been set.

Annual assessments have been made of the detection frequency of filters or wells with one or more pesticides above or below the limit value for pesticides in drinking water.

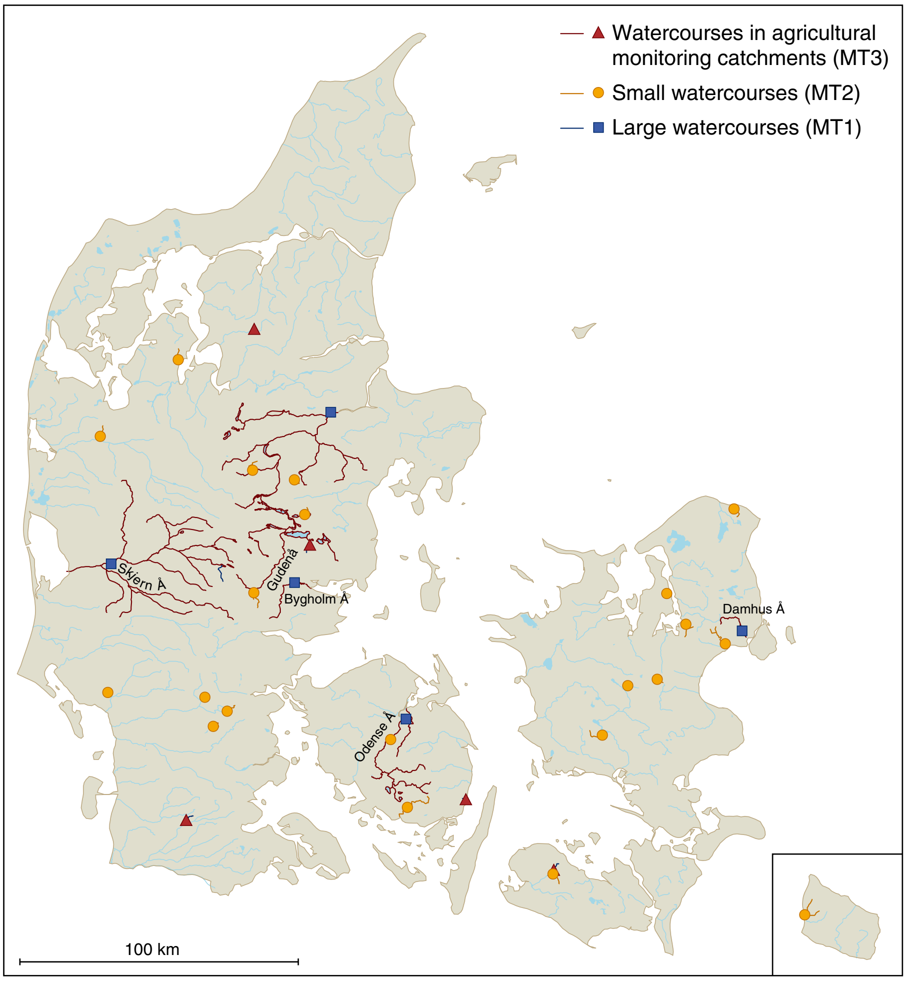
3.7 Watercourses

The purpose of the monitoring strategy for hazardous substances and heavy metals in watercourses is to

- document the general background level of hazardous substances and heavy metals
- document the level of hazardous substances in watercourses in connection with the land use in the catchment, e.g. during the pesticide spraying season where losses to the watercourses may occur as a result of wind drift and inadequate cleaning of spraying equipment
- determine the occurrence of hazardous substances and heavy metals in connection with natural incidental events in the catchment, such as heavy precipitation or snowmelt
- quantify the transport and subsequently the loss of hazardous substances and heavy metals from the entire catchment.

The monitoring programme for watercourses comprised three different types of watercourses evenly distributed throughout Denmark: five selected "large" watercourses (MT1), 21 small watercourses in areas with different soil conditions (MT2), and finally watercourses in the agricultural catchments (LOOP) (MT3), cf. section 3.5 (figure 3.5).

Figure 3.5. Watercourses where the concentration of hazardous substances and heavy metals have been monitored.



The catchments of the five large watercourses constitute 11% of the total Danish catchment. Monitoring of these watercourses comprised heavy metals, pesticides and other organic hazardous substances, and the results of the monthly measurements were used to quantify the transport of the substances to marine waters.

Monitoring of the small watercourses focused on the cultivation related loss of pesticides. The objective of the sampling strategy was to optimize the detection of pesticide discharges to watercourses. In watercourses in areas with different soil conditions, six annual samples distributed within the spraying periods in spring and autumn were taken. In LOOP watercourses, 16 annual samples were taken: six samples during the spraying season and the remaining as monthly samples, with the exception of July and August.

Result assessment

The assessment of the results of pesticide monitoring in watercourses is based on the detection frequency of the individual substances and on the maximum concentrations detected. The assessment of other hazardous substances and heavy metals are based on median values of the measured concentrations. The detected concentrations were furthermore compared with the quality objectives for the substances in surface water to the extent that quality objectives have been set.

The occurrence of heavy metals in watercourses has been described solely on the basis of monitoring from 2001 as a new method was introduced in 2000.

3.8 Lakes

The objective of the monitoring of hazardous substances and heavy metals in lakes is to describe the general occurrence of the substances. It was not expected to detect considerable temporal variation in the occurrence of the substances in the individual lakes and consequently the number of samples from lakes was substantially lower than the number of samples from watercourses. The input of hazardous substances and heavy metals to a lake can be detected over a long period of time; how long depends on the hydraulic retention time in the lake, and the turnover and sedimentation.

Eight lakes were analysed for hazardous substances in six water samples from each lake in 2001. Two samples were collected in June and July and one sample in August and September. Monitoring encompassed eight selected lakes as indicated in figure 3.6.

Figure 3.6. Lakes analysed for hazardous substances and heavy metals.

Result assessment

The principles used for assessing the lake monitoring results are the same as those used for watercourse monitoring.

3.9 Marine waters

The description of the geographic distribution and temporal variation of the occurrence of hazardous substances and heavy metal in the marine environment is based on measurements of the detected substances in mussels, fish and sediment. The water phase was investigated for a few individual water soluble hazardous substances that do not, or at least only to a limited extent, accumulate in living organisms (biota) or sediment. The concentration of heavy metals and hazardous substances in the water phase is analysed at intervals of two or three years. A total of 19 stations were encompassed by the monitoring programme for hazardous substances and heavy metals in the marine environment (figure 3.7). The extent of the monitoring was planned in deference to the local conditions.

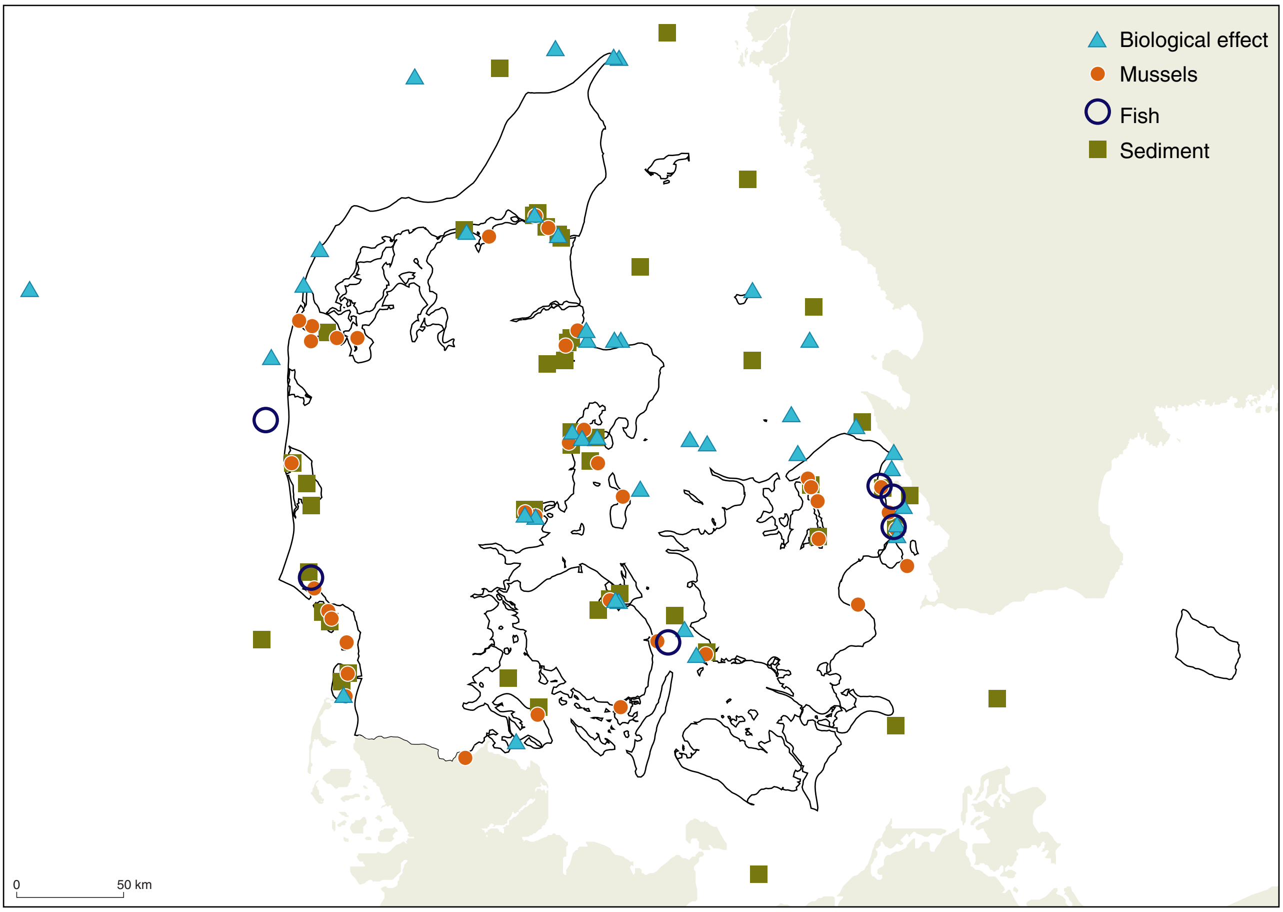


Figure 3.7. Marine monitoring stations with measurements of hazardous substances and heavy metals.

Common mussel, flounder and plaice are suitable for describing the level of heavy metals and hazardous substances in estuarine fjords and coastal waters, as they are stationery and widely distributed. Common mussels furthermore filter large quantities of water and are therefore able to take up and accumulate any pollutants in the water. Measurement of the concentration in the water phase only reflects the immediate concentration whereas measurement of the concentration in biota reflects the concentration over a longer period.

Monitoring encompassed flounder and plaice at a few stations where these types of fish have previously been monitored. Samples were collected once per year, all samples were collected in October-November.

In 2000 and 2003 sediment was investigated at three stations along a gradient from a point source. The sediment samples were mainly collected in areas with a sedimentation rate of 1-2 mm per year. A sample of the upper cm will therefore contain sediment from the previous 5-10 years. The analyses were performed on the fraction of the sediment that was below 2 mm in diameter. A few samples were also analysed on a fraction of < 63 µm.

Where possible, the samples of mussels and sediment were collected from stations situated sufficiently close to each other geographically to presume that the samples from the two stations were affected by the same sources.

Organotin compounds (antifouling agents) from ship paint can result in changes in the sexual characteristics in snails. These changes are the only biological impact examined in NOVA 2003. Samples of sediment analysed for organotin compounds and snails monitored for hormonal disturbances (imposex) were collected from the same stations or stations situated as close to each other as possible.

Mussels and fish

Data from 14 stations with mussels and 3 stations with fish (however, only 1 with organochlorines) are available.

The time series for metals in fish comprise data for up to 25 years. Data for common mussel (*Mytilus edulis*) or soft-shelled clam (*Mya arenaria*) include three independent subsamples per station per year. Each subsample consists of 25-50 mussels. Data for flounder (*Platichthys flesus*) or plaice (*Pleuronectes platessa*) consist of 25 individuals per station per year for metals and 10 individuals per station per year for organochlorines.

Concentrations of organic and inorganic hazardous substances in the mussels are indicated per kg dry matter. The concentration of organochlorines in fish is indicated per kg fat content and for the metals per kg dry matter. The only exception is mercury, which is indicated per kg wet weight in order to be able to compare with the pre-1998 data.

The trend in the concentration of hazardous substances and heavy metals has been estimated on the basis of a statistical analysis recommended by ICES (International Council for the Exploration of the Sea). The analysis includes stations with no less than 5 years' measurements in either mussels or fish.

Sediment

1-2 samples per station with two samplings for a period of six years were analysed.

In 1985 and 1990/91 an international investigation analysed the concentrations of heavy metals in the North Sea, the Skagerrak and the Kattegat (Pedersen, 1996). Five of the stations from this investigation are identical to the stations encompassed by NOVA-2003 and the measurements could therefore be used to assess a possible trend.

4 Quality criteria for water bodies

The measured concentrations of heavy metals and hazardous substances have been assessed in relation to already set or suggested quality objectives, quality criteria and limit values.

4.1 Surface water

Quality criteria for heavy metals and hazardous substances are described in Statutory Order No. 921 of 8 October 1996 on quality criteria for the aquatic environment and discharge criteria for specific hazardous substances to watercourses, lakes or the sea (Miljø- og Energiministeriet 1996). In addition, the Danish Environmental Protection Agency has recommended certain water quality standards in relation to a number of substances to help the municipal authorities to finally establish the water quality criteria.

Normally the discharge from a wastewater treatment plant is diluted at least 10-fold at the outfall. However, there will be some exceptions, such as when the discharge is to watercourses with a low summer flow level. In contrast, discharges to marine waters will often have an enhanced dilution. When assessing the general degree of compliance with the specified quality objectives for the aquatic environment, the assessments in this report are based on a 10-fold dilution.

4.2 Groundwater and drinking water

Statutory Order No. 871 of 21 September 2001 on water quality criteria and supervision with water supply plants stipulates limit values that apply to drinking water (Miljø- og Energiministeriet 2001). There are currently no specified limit values for groundwater, and the groundwater was therefore assessed according to the limit values for drinking water.

4.3 Marine biota and sediment

There are in Denmark currently no specified limit values for heavy metals or organopollutants in sediment or biota in the marine environment.

The monitoring results of marine biota have primarily been assessed in relation to the ecotoxicological assessment criteria "Ecotoxicological Assessment Criteria" (EACs) drawn up by the OSPAR commission (OSPAR 1998). The EAC value is expressed as a concentration interval.

The upper limit (EAC_{High}) is set such as to indicate that exceedance of this limit entails the risk that long-term exposure could have effects on the most sensitive species in the ecosystem. If the concentration lies within the interval the possibility of effects cannot be excluded. If the concentration is below the lower limit (EAC_{Low}) it is probable that harmful effects on the environment will not occur.

Metal concentrations in mussels have been assessed on the basis of the guiding Norwegian classification system elaborated by the Norwegian Pollution Control Authority (Statens Fourensningstilsyn (SFT) 1997). The system grades the degree of pollution (the environmental state) on a scale from I to V:

I. Slightly to moderately polluted (good state)

II. Moderately polluted (fair state)

III. Markedly polluted (poor state)

IV. Extremely polluted (bad state)

V. Severely polluted (very bad state)

4.4 Sludge

Statutory order No. 623 of 30 June 2003 on application of waste products for agricultural purposes (Miljøministeriet 2003) stipulates limit values for certain heavy metals and hazardous substances. The sludge cannot be applied to agricultural land unless the concentration of the substances referred to is below the limit value.

5 Analysis quality assurance

In order to assure the analysis quality, the laboratories must to the widest possible extent be accredited for the analyses comprised by the monitoring programme. However, at the time when NOVA-2003 was introduced, some analyses of heavy metals and hazardous substances could not be made as accredited analyses. Consequently, it was decided that the Danish Environmental Protection Agency was to appoint laboratories for the individual analysis in the individual types of samples. The Danish Environmental Protection Agency has appointed the laboratories on the basis of performance testing results. Where performance testing has not taken place, the appointment was made on the basis of comparable documentation. The list of laboratories appointed by the Environmental Protection Agency is updated once a year.

Several of the substances comprised by NOVA-2003 had not previously been analysed in the matrices where they were to be analysed. It was therefore necessary to develop analysis methods before laboratories could be appointed. The problems were solved during the programme period, and in recent years all substances have been analysed by appointed laboratories. For a few substances it appeared that the costs of solving the analysis problem would exceed the utility value of the analysis result. This applies to, for example, fluoroacetic acid in wastewater and silver in groundwater. These substances were therefore excluded from NOVA-2003.

For some substances it has proved impossible to meet the detection limit requirements. Throughout the monitoring period considerations have been made whether to raise the detection limit with the risk of the analysis result being insignificant or whether to postpone monitoring until it would be possible to meet with the detection limit requirements.

Sampling requirements in NOVA-2003 are described in detail in technical instructions (Kronvang et al. 1998; Kaas & Markager 1999; Geological Survey of Denmark and Greenland 1999; Danish Environmental Protection Agency, 1999c). These mandatory requirements are set to ensure correct and uniform sampling methods.

6 Summary of monitoring results for the individual groups of substances

Discharges from wastewater treatment plants are one of the main reasons for the presence of heavy metals and organic micropollutants in the aquatic environment. Seen as a whole, the group of metals accounts for the greatest proportion of discharge for the period 1998-2003 (table 6.1). Among all the organic hazardous substances the aliphatic amine dimethylamine accounts for the greatest proportion of discharge.

Table 6.1. Total average annual discharge, detection frequency and mean concentration of metals and organic micropollutants from wastewater treatment plants for the period 1998-2003. The table shows the 20 substances with the greatest discharge.

Substance	Group of substances	Discharge from waste water treatment plants (kg/year)	Detection frequency (% of analysed samples)	Mean concentration (µg/l)
Zinc	Metal	69,147	100	91
Copper	Metal	5,066	88	6.7
Nickel	Metal	4,817	93	6.4
Dimethylamine	Aliphatic amine	2,695	92	3.6
TCPP	P triester	1,581	99	2.1
Lead	Metal	1,402	64	1.9
DEHP	Plasticizer	1,361	57	1.8
Arsenic	Metal	981	42	1.3
Phenol	Phenol	368	82	0.5
Toluene	Aromatic hydrocarbon*	341	44	0.5
MTBE	Ether	323	51	0.4
Tributhylphosphate	P triester	276	89	0.4
Nonylphenol	Alkylphenol	228	68	0.3
Diethylamine	Aliphatic amine	160	11	3.6
Diethylphthalate	Plasticizer	158	31	0.2
Bisphenol A	Phenol	155	45	0.2
Dibutylphthalate	Plasticizer	95	15	0.1
Xylene	Aromatic hydrocarbon*	76	13	0.1
Cadmium	Metal	72	34	0.09
Mercury	Metal	66	32	0.09

*described under solvents

The results of the investigated occurrence in the environment of the individual substances will be described group wise in the following sections.

6.1 Heavy metals

The atmospheric deposition has decreased correspondingly with emission of heavy metals to the air since 1990. The most significant decrease was for lead which is largely ascribed to the phasing out of lead from petrol.

By comparing the atmospheric deposition of metals to the landmass and inner Danish marine waters with an estimate of the wastewater discharges it appears that the atmospheric deposition of the individual metals was 3-20 times higher than that of the wastewater discharge in Denmark.

In outflows from treatment plants copper was the only metal occasionally found in concentrations exceeding the quality standard for copper in surface waters recommended by the Danish Environmental Protection Agency, although the concentrations did not exceed the existing water quality criteria. Copper was also the heavy metal that displayed the lowest decrease in atmospheric emission.

As regards groundwater, the highest concentrations of heavy metals and most frequent exceeding of the limit values for drinking water were found in the surface waters in the agricultural catchments. Arsenic differed in that it was found in the highest concentrations in deeper groundwater. The high arsenic concentrations are probably geologically dependent. Metals are retained somewhat by the water treatment in the waterworks, and may consequently not pose a problem in relation to drinking water. At a few locations, the high concentrations of nickel and arsenic do, however, pose a problem.

Concentrations of heavy metals in watercourses were low in relation to the quality criteria for surface water. The concentration of metals in lakes was generally lower than that in watercourses with the exception of mercury with concentrations apparently on a par with or sometimes exceeding the concentrations in watercourses. This is due to an accumulation in the lake sediment of mercury deriving from previous discharges and a subsequent slow release of mercury from the sediment.

The mean concentrations of most metals in mussels in the marine environment were at a level corresponding to "moderate status". Lead concentrations corresponded to "good status" whereas copper concentrations corresponded to "bad status". Most monitoring stations did not exhibit any significant changes in concentrations during the period 1998-2003, although a few stations had significantly increasing concentrations of copper, cadmium, lead, and mercury. Concentrations of nickel had decreased markedly at one monitoring station.

6.2 Pesticides

Pesticide pollution in groundwater is most frequently attributable to BAM and the group of triazines. BAM is the degradation product of dichlobenil (Prefix and Casoron G) and chlorthiamid (Casoron). The mother compounds of BAM have been used for eliminating weeds on, for example, uncultivated land along roads, paths, pavements and railway tracks/stations. Application of the two herbicides was prohibited in 1997. The group of triazines include the active substances atrazine, hexazinon, terbutylazine and simazine. Use of the two first pesticides has now been phased out while the latter two are still in use. Simazine is among the pesticides used most frequently in the agricultural catchments and in public areas. In addition to the mother compounds various degradation products of triazines were also detected.

The frequency with which pesticides were detected in groundwater during the period 2001-2003 remained at the same level at approx. 27%, and approx. 10% of intakes exceeded the limit value for drinking water at 0.1 µg/l. The proportion of waterworks wells exceeding the limit value for drinking water during the same period fell approx. 5%. The proportion of waterworks wells exceeding the limit value is lower than the proportion of the intakes encompassed by groundwater monitoring (GRUMO intakes) that exceed the limit value because waterworks wells with too high pesticide concentrations have been shut down.

The pesticides detected most frequently were to a large extent the same in both watercourses and lakes. These pesticides are to some extent the same pesticides found most frequently in groundwater (table 6.2).

Table 6.2. List of the most frequently detected pesticides and degradation products in watercourses and lakes from 1998 to 2003, and total detections in groundwater in GRUMO, LOOP (agricultural catchments) and waterworks wells for the period 1993-2000. The detection percentage is indicated in brackets. The detection percentages in watercourses and lakes are calculated in proportion to the number of analysed samples, and in groundwater in proportion to the number of analysed intakes/wells. Data from Bøgestrand (ed.) 2004; Jensen et al. 2004 and GEUS 2004.

Watercourses (168 samples)	Lakes (48 samples)	GRUMO (approx. 1,000 intakes)	LOOP (50-100 intakes)	Waterworks (220-5,500 wells)
AMPA (91%)	TCA (61%)	BAM (20%)	4-nitrophenol (39%)	BAM (21%)
BAM (83%	AMPA (54%)	DEI atrazine (9.0%)	DEI atrazine (30%)	4-nitrophenol (3.2%)
Glyphosate (82%)	BAM (54%)	DIP atrazine (7.4%)	DIP atrazine (23%)	4CPP (2.6%)
TCA (52%)	4-nitrophenol (50%)	DE atrazine (6.8%)	AMPA (23%)	Atrazine (2.6%)
MCPA (32%)	Glyphosate (38%)	4-nitrophenol (6.6%)	Bentazon (21%)	DE atrazine (2.6%)
Terbutylazine (32%)	DNOC (26%)	Atrazine (5.3%)	Glyphosate (16%)	Bentazon (1.9%)
Bentazone (32%)	MCPA (23%)	Dichlorprop (4.3%)	DE atrazine (15%)	Mechlorprop (1.9%)

Glyphosate and AMPA were among the most frequently detected pesticides in both drainage water and in watercourses and lakes. The substances were also detected frequently in groundwater although not as frequent as in surface water (figure 6.1). The detection frequency was higher in the younger near-surface groundwater in the agricultural catchments than in the deeper groundwater. The total sales volume of glyphosate was increasing throughout the 1990s. In 2003, glyphosate constituted 32% of the total sales volume of pesticides for agricultural purposes and 45% of the total sales volume of herbicides.

Pesticide concentrations in drainage water beneath the fields were in most cases lower than the limit values for pesticides in drinking water. Concentrations in surface water were generally low with lower median concentrations in lakes than in watercourses. The detection frequency was, however, higher in lakes than in watercourses. Maximum concentrations in lakes were on a par with or slightly below the maximum concentrations in the investigated large watercourses, and the investigated small watercourses exhibited concentrations exceeding those in large watercourses as well as large lakes.

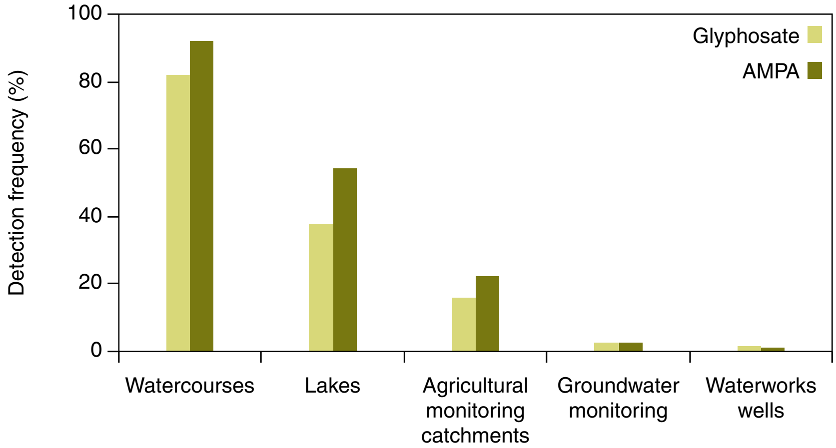


Figure 6.1. Detection frequency of glyphosate and AMPA in watercourses and lakes in 2003 and in groundwater in LOOP catchments, GRUMO sites and waterworks wells from 1998-2003. The detection frequency in watercourses and lakes is determined as % of analysed samples with detections and in groundwater as % analysed intakes with detections.

Danish quality criteria have been set for eight different pesticides in surface water. On a single occasion concentrations of four different pesticides in a watercourse were found to exceed the Danish quality criteria. A few substances (dinoseb, isoproturon and propinicol) with no set quality criteria were detected in concentrations so high that it cannot be the result of normal use, but was probably caused by a direct discharge to the watercourse, possibly in connection with rinsing of spraying equipment.

6.3 Phenols and chlorophenols

Phenols and chlorophenols were detected in small concentrations in the effluent from wastewater treatment plants. Pentachlorophenol was the only phenol included in the surface water monitoring. The substance was neither detected in seawater, nor in the four of the five watercourses. A few occurrences of pentachlorophenol were detected in a single watercourse in concentrations considerably higher than the mean level in outflow from point sources, but still below the quality criteria for surface water.

Both phenol and chlorophenols were detected in groundwater. The occurrence was most frequent in the near-surface groundwater in the agricultural catchments. In a few incidents, the concentrations exceeded the limit value for the substances in drinking water.

In the group of phenols and chlorophenols, phenol was the substance most frequently detected in groundwater. This may be the result of phenol being formed naturally in the decomposition of organic matter. However, based on the detected high concentrations of phenol found in liquid manure it cannot be excluded that the frequent occurrence of phenol in near-surface groundwater may be referred to seepage following application of liquid manure on farmland. 2,4-dichlorophenol can be formed by decomposition of pesticides in the group of phenoxy acids which may be the cause of the occurrence of the substance in groundwater.

6.4 Alkylphenols

Alkylphenols, nonylphenols and nonylphenolethoxylates were detected in effluents and sludge from wastewater treatment plants, and in liquid manure. Nonylphenols were detected in most of the investigated sludge and were also among the substances detected most frequently in effluents from wastewater treatment plants.

Nonylphenols were detected in a few cases in watercourses, but not in any of the investigated lakes. There was no indication of any inputs of nonylphenols or nonylphenolethoxylates.

In groundwater, nonylphenols were detected most frequently in the near-surface groundwater, which may be a result of application of liquid manure or sludge, or spraying with pesticides. No exceedances of the limit value for nonylphenols or nonylphenolethoxylates in drinking water were detected.

Nonylphenols were detected frequently in sediment in fjords and coastal areas. No quality criteria or equivalent objectives have been set for nonylphenols in sediment and it is therefore not possible to assess the environmental risks of the detected concentrations.

6.5 Plasticizers – phthalates

Plasticizers were detected frequently in wastewater and sludge with di(2-ethylhexyl)phthalate (DEHP) as the predominant substance. DEHP was among the substances detected most frequently and in the greatest quantities in effluents from wastewater treatment plants. The quality criteria for DEHP in surface water of 0.1 µg/l recommended by the Danish Environmental Protection Agency has probably been exceeded occasionally, while the detected concentrations in sludge were considerably lower than the limit value for application of sludge on agricultural land as stipulated in the Statutory Order on the Application of Waste Products for Agricultural Purposes. For approximately two thirds of the wastewater treatment plants the input of DEHP to the treatment plants have decreased between 1998-2000 and 2001-2003.

In spite of the frequent occurrence of DEHP in effluent discharges from wastewater treatment plants, the detection of the substance in watercourses and lakes was very limited. The most frequent occurrence was in river Damhusåen. The detected concentrations in watercourses and lakes were close to the detection limit of 0.5 µg/l.

DEHP was detected frequently in the sediment of fjords and coastal areas. No quality criteria have been set for DEHP in marine sediment. It is therefore not possible to asses the environmental risk of the detected substances.

In groundwater, the phthalate with the highest water solubility, dibutylphthalate (DBP), was detected most frequently in the near-surface groundwater. DBP was also detected in liquid manure and sludge. The occurrence in the near-surface groundwater may be ascribed to leaching from applied liquid manure or sludge on agricultural fields. A few incidents of concentrations exceeding the limit values for drinking water were detected in both the near-surface groundwater and the deeper groundwater.

6.6 Anionic detergents

LAS was detected in 69% of the investigated sludge samples. The declining sales volume of LAS in recent years is reflected by a decrease in the mean concentration of LAS in intakes to wastewater treatment plants and in wastewater sludge. The concentrations of LAS in sludge decreased by approx. 62% during the period 1997-2002. Concentrations of LAS in sludge were significantly below the limit value for application on agricultural land.

LAS was only detected in a few samples of effluent from wastewater treatment plants and only in one of the investigated watercourses (river Damhusåen). The occurrence in river Damhusåen is not directly ascribable to discharge of wastewater from a wastewater treatment plant.

Concentrations of LAS in liquid manure were far below the limit value for LAS in sludge. It has been estimated that LAS concentrations in liquid manure will not entail exceedances of the current ecotoxological soil quality objectives within the next 50 years (Scott-Fordsmand et al. 1995).

6.7 Solvents

The group of solvents comprised aromatic hydrocarbons, halogenated aliphatic and aromatic hydrocarbons, and the ether MTBE. With a few exceptions the substances were detected to a small extent in effluent from wastewater treatment plants and in surface water. Chloroform and toluene were detected in 50% and 44%, respectively, of the analysed samples of effluent from wastewater treatment plants. The detected concentrations are not considered problematic in relation to the quality criteria for surface water.

The aromatic hydrocarbons were the most frequently detected solvents in sludge and also the group of substances detected in the highest concentrations. Seen as a whole, the aromatic hydrocarbons were among the substances detected most frequently in sludge.

In groundwater, toluene was the most frequently detected solvent in both the near-surface groundwater in the agricultural catchments and the deeper groundwater under the groundwater monitoring programme, and in the waterworks. Chloroform was detected most frequently in the deeper groundwater, but not in the near-surface groundwater. Investigations show that chloroform can be formed naturally under, for example, forest soils (Engvild 2000) and this may explain why chloroform is only present in the deeper groundwater. A few incidents of concentrations exceeding the limit value for drinking water were detected in the groundwater.

6.8 Polyaromatic hydrocarbons

The polyaromatic hydrocarbons (PAH) were detected in the majority of the sludge samples from wastewater treatment plants, but only in a small proportion of the samples of effluent from wastewater treatment plants. The concentration of PAH in sludge occasionally exceeded the level permitted in sludge used for application on agricultural fields.

Concentrations of PAH compounds in liquid manure were significantly lower than those in sludge. The light PAH compounds were detected most frequently, while the heavy compounds were only detected to a small extent or not at all.

In a few cases naphthalene was detected in groundwater in concentrations below the limit values for drinking water. A number of PAH compounds were detected in watercourses and lakes in concentrations close to the detection limit. The national quality criteria for PAH in surface water were generally determined on the basis of the most problematic PAHs. Consequently, the quality criteria was set very low (0.001 µg/l) and lower than the detection limit (0.01 µg/l).

In coastal marine waters and open marine waters PAH was detected frequently in both mussels and sediment, and in certain areas anthracene was detected in mussels in concentrations that entail the risk of effects. In sediment, both light and heavy PAH compounds were detected in concentrations that may entail the risk of effects.

6.9 Phosphortriesters

Trichloropropylphosphate (TCPP) was the predominant substance among the phosphortriesters (P triesters) in both wastewater and sludge. The substance was detected in the majority of the investigated samples and was among the substances found in discharges from wastewater treatment plants in the highhighest amounts. Knowledge about the effect of P triesters on the environment is very limited. A quality criteria for surface waters has been set for one of the P triesters, and the detected concentrations in wastewater were far below this criteria. P triesters were not included in other parts of the NOVA-2003.

6.10 Aliphatic amines

The group of aliphatic amines has not been in focus because these substances do no bioaccumulate and are not highly toxic. Diethylamin and dimethylamin, which were monitored at the wastewater treatment plants, were detected frequently in both sludge and wastewater, but the environmental impact was considered to be minimal. Mean concentrations of diethylamin and dimethylamin in outflow from wastewater treatment plants were below the quality criteria for the two substances in surface water.

6.11 PCB, dioxins and chlorinated pesticides

PCB, chlorinated pesticides, dioxins and furans degrade very slowly and will therefore occur in the environment long after cessation of application or discharges of the substances. PCB was frequently detected in sludge, indicating that although the use of PCB is now prohibited, PCB still occurs in non-treated wastewater. To the extent it was possible to detect a trend in PCB concentrations in marine sediment, concentrations were decreasing.

Dioxins and furans were detected in virtually all the investigated sludge. Most substances occurred in low mean concentrations. Four substances differed in that they were detected in significantly higher mean concentrations, this being mainly ascribed to a few occurrences of very high concentrations. Concentrations in sludge of three of these substances were found to be decreasing in most treatment plants during the period 1998-2003.

PCB and chlorinated pesticides were not detected in either outflow from wastewater treatment plants or in watercourses.

Chlorinated pesticides were detected in mussels in the marine environment, but not in sediment. The occurrence of chlorinated pesticides in the marine environment is attributable to inputs in earlier years.

6.12 Organotin compounds

Of the organotin compounds, trybutyltin (TBT) has been particularly highlighted because of its specific impact on the sexual organs of snails.

TBT was detected frequently in both sediment and in mussels in coastal areas with the highest concentrations detected in the vicinity of harbours. Correspondingly, imposex and the occurrence of sterile female snails were detected most frequently close to harbours.

TBT has also proved to be particularly toxic for the larval stage of, for example, mussels and snails, which is a factor that may inhibit the recruitment of new generations. As many species of snails do not have free-swimming larval stages the recruitment is additionally difficult. Reduced recruitment may result in a distortion of population structures, or lead to disappearance of the snails. The possible effect on populations further away from the harbours is connected with some uncertainty.

TBT and other organotin compounds are accumulated in all stages of the food chain in the sea, for example, molluscs, fish, birds and mammals, and even in humans (figure 6.2). The highest concentrations were detected in porpoises. TBT is mainly taken up through the food. The ability of the various species to degrade and excrete the substances is of great importance for the level of accumulation.

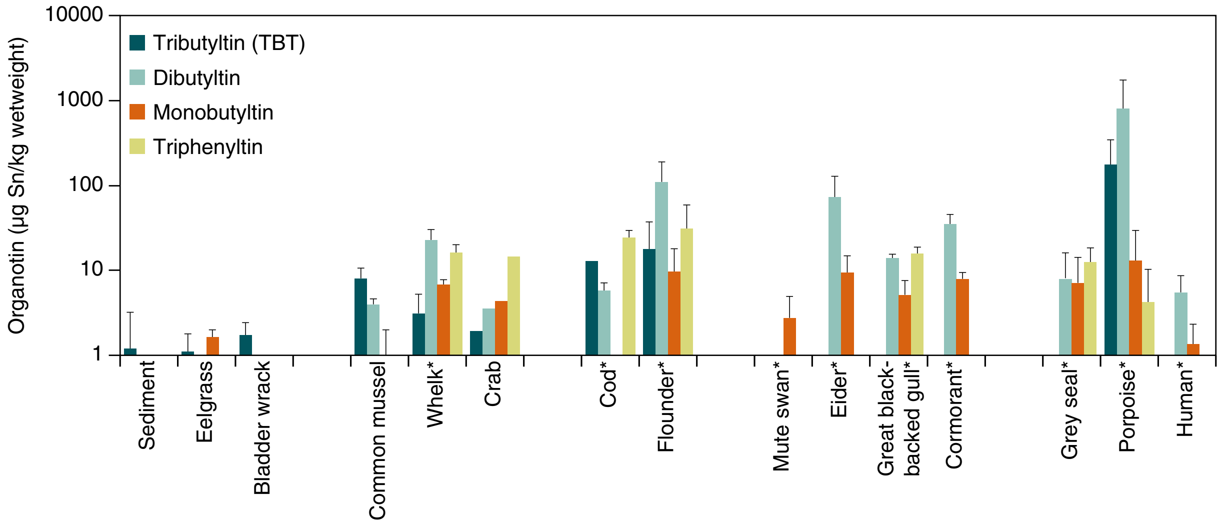


Figure 6.2. TBT and other organotin compounds accumulate in the different stages of the food chain of the sea. The highest concentrations were detected in porpoises. The data presented here are from the Belt Sea with the exception of the grey seal which are from the Swedish part of the Baltic Sea and people from Funen. * indicates samples of analysed liver. Notice the logarithmic scale.

6.13 Medicines

In NOVA-2003 only liquid manure was analysed for medicines, and of these only antibiotics. Antibiotics were detected in 30 of 45 samples. The highest concentrations were detected in liquid manure from pigs.

According to data from the freshwater fish farms, the consumption of medicines and auxiliary substances has been at the same level for the period 1998-2003. An investigation of the occurrence of antibiotics in the aquatic environment surrounding freshwater fish farms indicates that antibiotics used in the fish farms could be detected in the water phase of the watercourses downstream of the fish farms (Pedersen et al. 2004). Antibiotics were also detected in the sediment of fish farms. Concentration in the sediment of a single antibiotic from a fish farm was approx. 3 times higher than that found in liquid manure from pigs.

Pedersen et al. 2004 investigated resistance trends in connection with treatment with antibiotics. The resistance level among the investigated fish-pathogenic bacteria was low in the fish farms and at the same level as in watercourses unaffected by fish farms.

6.14 Future implications

Optimisation of monitoring of hazardous substances and heavy metals

Six years of monitoring of hazardous substances and heavy metals in NOVA-2003 have showed that some substances do not occur or only occur in insignificant quantities in the environment. These substances were eliminated in connection with the revision of the monitoring programme, and the programme was optimised to include only substances that had been proved relevant to monitor. The list of substances was reduced from approx. 250 subtances to approx. 190. The phased out substances were mainly among the pesticides. A few new substances that previous investigations had proved relevant to include were added, among these brominated flame retardants.

Strategy for “new” substances

In connection with the revision of the monitoring programme, screening analyses of “new” substances were performed parallel with “routine” monitoring. The results of the screening analyses will serve as the basis for the decision whether it is relevant to include the analysed substances in the routine monitoring. For example, a screening analysis will be performed for the few substances on the list of prioritised substances in the Water Frame Directive that are not already encompassed by the monitoring programme. In 2005/06, organotin compounds, PFAS (Perfluorinated Alkylated Substances), and beryllium in groundwater will be included in the screening analyses and in 2006/07 pharmaceuticals and triclosan will be included.

Effects on snails and fish

The sexual changes detected in snails by the NOVA-2003 monitoring are directly related to the impact by TBT. Other investigations have detected sexual changes in fish in watercourses, but a corresponding connection with discharged hazardous substances to the investigated watercourses has not yet been established, although foreign investigations indicate a possible connection. The investigation was performed by Århus County in cooperation with University of Southern Denmark (Århus Amt 2001).

The monitoring programme was extended in 2004 to include further effect studies. The detection of two-headed viviparous blenny fry in summer 2005 was one of the first results of these studies. The detections of two-headed viviparous blenny fry are ascribed to the impact of hazardous substances, but it has not been possible to identify one or several specific substances as being the cause.

Environment and health

The monitoring programme has been designed to describe only the status and trend in nature and the environment. There has been awareness of the effect of several of the substances encompassed by the monitoring programme on humans also, especially in relation to fertility. Humans can take up the substances through the air, drinking water or food.

The monitoring programme was extended in 2004 with analyses of organopollutants in precipitation and the air. The air in large cities has been monitored for concentrations of particles and a number of harmful substances since the beginning of the 1980s, solely in the interest of human health. Drinking water was analysed by the waterworks controls. The Danish Veterinary and Food Administration monitors chemical pollution in food, including a number of substances encompassed by the environmental monitoring.

Water Frame Directive and other directives

The Water Frame Directive is an EU directive whose purpose it is to ensure the protection of watercourses, lakes, coastal waters and groundwater.

The results of the monitoring of hazardous substances and heavy metals have served as a basis for the assessment of the environmental status of the Danish water bodies that the Danish counties have prepared in connection with the basic analysis of the Water Framework Directive. In relation to the Water Framework Directive the operational monitoring of water bodies at risk of non-compliance with the established objectives for the environment in 2015 will be planned on the basis of the basic analysis. This also applies for the assessment of the extent of operational monitoring of hazardous substances and heavy metals.

Monitoring of hazardous substances and heavy metals under NOVANA (the National Monitoring and Assessment Programme for the Aquatic and Terrestrial Environments 2004-2009) will, for instance, comply with the obligations of control monitoring described in the Water Framework Directive. If water bodies encompassed by NOVANA are at risk of non-compliance with the environmental objectives, NOVANA will comply with parts of the obligation of operational monitoring.

The marine monitoring programme will be carried out in coordination with the countries bordering the marine areas in question. This applies to the Baltic Sea via the HELCOM convention, the North Sea via OSPAR convention, and the Wadden Sea via the Wadden Sea Cooperation. In addition to monitoring of sediment, mussels and fish in the Wadden Sea, monitoring also includes the concentrations of a number of hazardous substances in birds' eggs (Essink et al., 2005).

Research needs

Two-headed viviparous blenny fry are ascribed to the impact of hazardous substances as are changes in the sexual organs of snails. Correspondingly, NOVANA encompasses effect studies in form of investigations of cell damage in mussels and monitoring of the activity of detoxification enzymes in fish. These effects are also ascribed to the impact of hazardous substances. A relationship between TBT and sexual changes in snails has been identified and documented, but the substances causing the other effects encompassed by the monitoring programme have not yet been identified. There is a need for further research to have these relationships identified.

P triesters are the group of substances among the organopollutants that are discharged from wastewater treatment plants in the highest quantities. There is limited knowledge about the effect of the substances on the environment. Considering the extent of the discharges there is a need to elucidate the possible effect of the substances on the environment.

The introduction of pesticides as low-dose pesticides, i.e. pesticides that are effective at extremely low concentrations and therefore applied in lower concentrations than traditional pesticides, gives rise to considerations whether to introduce new sampling methods, possibly in way of passive sampling where a substance is captured from the water for a period and bound to an adsorbent so that measurable concentrations are eventually achieved.

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This report presents the results of hazardous substances and heavy metals monitoring in the aquatic environment under the Danish National Monitoring Programme of the Aquatic Environment during the period 1998-2003 (NOVA-2003). The report describes the sources and occurrences of the substances in groundwater, watercourses, lakes and marine waters, and estimates the significance of these occurrences. The report is based on the annual reports prepared for each subprogramme by the Topic Centres. The latter reports are based on data collected and submitted by the Danish county authorities.

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