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TNO-report

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Atmospheric deposition of pesticides, PAHs and PCBs in the Netherlands (translation of R2002/606)

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Preface

Research into atmospheric deposition has been carried out since the 1980's within the framework of the Interdepartementale Werkgroep Atmosferische Depositie (Interdepartmental Working Party Atmospheric Deposition, IWAD). During the first years, virtually only model calculations were used to map deposition. The uncertainty in the results of these calculations, however, was large and the demand for monitoring data increased. After an extensive preliminary study, a large monitoring programme was started in 1999 aimed at gaining a better insight into the deposition of pesticides, PAHs and PCBs. The results of this monitoring programme are extensively discussed in this report, and conclusions are drawn.
With the results of this study, the large uncertainty in earlier estimates of the atmospheric deposition has for many substances been reduced to acceptable levels. This means that the report provides a good basis for the development of a well-balanced policy aimed at improvement of the environmental quality.

The study was financed by contributions of the Ministry for Housing, Spatial Planning and the Environment (VROM), the Ministry of Public Works (V & W) (via RIKZ, the Dutch National Institute for Coastal and Marine Management), RIZA, the Institute for Inland Water Management and Waste Water Treatment and the Directorate IJsselmeergebied), STOWA, the Foundation for Applied Water Management Research, the province Noord Holland, RIVM, the National Institute of Public Health and the Environment, and TNO.

The members of IWAD, chaired by the Ministry of VROM, served as project advisory board. We thank the various members and chairmen (initially Douwe Jonkers, now Hans Meijer) for their contribution in all stages of the study. We particularly thank Ton van der Linden (RIVM), Remi Laane (RIKZ) and Rob Faasen (RIZA) for their extensive comments on draft versions of this report.

In 2003 the Dutch version of the report was translated into English externally.

Short summary

Estimates over the last ten years have shown that the atmosphere is an important diffuse source of pollution for surface water. Organic compounds such as PAHs, PCBs, pesticides and metals are among the pollutants. The significance of this process for pesticides was stressed again in a 1998 report of the Gezondheidsraad (The Dutch Health Council) . The estimates of the input from the atmosphere were based on model calculations. These models calculate the dispersion of substances that are emitted into the atmosphere. The uncertainties in these estimates, however, were very large, also because they had not been validated. An uncertainty factor of five to ten was estimated for pesticides. A research project was started in 1999 in the context of the Interdepartementale Werkgroep Atmosferische Depositie (IWAD; Interdepartmental Working Party Atmospheric Deposition). The objective of this project was to determine atmospheric deposition on the Dutch territory. The project was carried out by TNO-MEP and was financed by contributions of the Ministry for Housing, Spatial Planning and the Environment (VROM), the Ministry of Public Works (via RIKZ, RIZA and TNO target funding), the Ministry of Agriculture, Nature Management and Fisheries (via TNO target funding), and STOWA. The programme was extended later in 1999 by contributions of the Province Noord Holland and the Department of Public Works, Directorate IJsselmeergebied. RIVM made an additional contribution in 2001.
Precipitation and air samples were taken from summer 1999 up to December 2001 at 18 locations spread over the Netherlands. The samples were analysed for pesticide, PAH and PCB concentrations. In addition to the measurements, model calculations were carried out with the OPS model of RIVM, based on emission estimates. The emission estimates of pesticides in the Netherlands were prepared by ALTERRA on the basis of pesticides use figures in the Netherlands. The emission estimates of other substances and of pesticides from surrounding countries were produced by TNO on the basis of land use data and Dutch emission data.

The monitoring data have been analysed extensively; here two views are relevant. On the one hand, atmospheric deposition presents a chronic load into the Dutch surface water for a number of substances. The significance of this source (the load) can be compared with other sources such as leaching. On the other hand, atmospheric deposition may in the vicinity of the source in certain periods possibly lead to high concentrations in surface water. In the absence of criteria for precipitation and air for most substances, a comparison with MTR (maximum permissible level) values for surface water provides insight. The study yielded the following conclusions:

- There is a strong relationship between the use of pesticides in certain periods and areas and the pesticides deposition patterns throughout the year and throughout the country. The annual deposition may also, by differences in use, differ by a factor two from year to year.

- Substances that are no longer authorised, are in the year 2001 still found in precipitation and air in the Netherlands, of which the sources must partly be sought in the Netherlands. Sources from surrounding countries seem to make a significant contribution to the input of substances such as *atrazine* and *trifluralin*. This is probably also true for a number of long forbidden, persistent substances such as *endrin* and *o,p-DDE* (a transformation product of DDT). The contribution from surrounding countries is much less significant for most other substances.
- The concentration of eighteen different pesticides and four PAHs in precipitation regularly exceeds the MTR level for surface water. Furthermore, the concentrations of twenty two pesticides and some PAHs exceed the drinking water criterion. The average concentration of Borneff PAHs exceeds the drinking water criterion in the year 2000.
- Atmospheric deposition is a significant source of some pesticides and PAHs into inland surface water by:
  1. The measured input into the surface water (expressed as annual load) in the Netherlands as result of atmospheric deposition of pesticides is of the same order of magnitude as the input by drift or by lateral leaching.
  2. The measured input by atmospheric deposition of PAHs (annual load) is almost just as large as the other forms of input.
  3. For HCB, PCBs and *pentachlorophenol*, the atmospheric route is much more important than other routes.
- Calculations for some substances with a simple model for surface water show that the average annual load by atmospheric deposition alone probably does not lead to an exceedance of the MTR in surface water, but atmospheric deposition can contribute to the input by other sources, which may result in an earlier exceedance. And for some pesticides, the high load by atmospheric deposition may in certain periods of the year and in a specific region probably lead to an exceedance of the MTR in that period.
- The model calculations show that for some substances the NR (Negligible Risk) level in surface water *can indeed* be exceeded by atmospheric deposition alone.
- The input into Dutch soil (annual load) has also been estimated. The absence of knowledge and reliable data, however, makes estimates of deposition on the soil rather uncertain. Comparison with the input by other sources is often difficult because sound data are lacking. For nature areas, however, atmospheric deposition often is the only source.
- Generally, the OPS model appears to be suitable for calculation of the deposition of pesticides. The difference between measured and calculated depositions was less than a factor two for well over half of the substances. Larger differences were found for the other substances. These differences are probably caused by inaccuracies in the emission estimates that have been used. The differences between calculated and measured deposition of two different PAHs were a factor two to three.

The conclusions above are derived from the current study. Despite the broad set-up, some restrictions are of course associated with to the chosen approach:

- A representative product package has been chosen. An important group of pesticides such as the bis-dithiocarbamates (*maneb* etc.) and some important substances such as *glyphosate, diuron* and *carbendazim*, however, have not, or only to a limited extent, been included in the study.
- The location of the monitoring stations was aimed at obtaining a nation-wide picture with specific attention for differences between the regions. The monitoring locations were therefore situated in such a way that effects by local sources were minimal. Deposition could be considerably higher in the direct vicinity of sources, e.g. within a radius of one kilometre around important sources, where it could easily cause an exceedance of the MTR levels.
- Besides the direct load there is the indirect load. This is the deposition on land that gets into the surface water though leaching. The conclusions above are based on the measured deposition excluding this indirect load. The size of this indirect load, however, is very uncertain. First estimates show that this route could be just as large as direct deposition, as a result of which total atmospheric deposition could become twice as high.
- Comparison with criterion concentrations such as the MTR is only possible for individual substances. The effect of deposition by a combination of a large number of substances has not been studied.

Recommendations

- Monitoring of atmospheric deposition

  The results of the study give an impulse to a strategy aimed at deposition monitoring in the Netherlands. This consists of a combination of deposition estimates on the basis of model calculations and a restricted national monitoring network. It is recommended to include the results of the model calculations formally in the Emission Registration. This procedure would provide those responsible for water quality control and other interested parties with consistent deposition estimates in their (control) area. Objectives of the monitoring network include determination of the actual load with PAHs, PCBs and pesticides, model testing, and monitoring of developments in the presence of pesticides in coherence with changes in the product package.
- Authorisation policy

  In view of the found significance of atmospheric deposition, arguments seem to be present to include the atmospheric deposition route in the authorisation procedure of pesticides. There are good possibilities to convert the available knowledge into an instrument that can be used in the authorisation evaluation. Such possibilities could be investigated in a pilot study.

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Extended summary

S.1 Introduction

Substances get into surface water via various routes, where direct discharges, runoff, and drift are important. Substances can also be transported to the earth surface from the atmosphere, bound to precipitation or through direct uptake from the air. Recently conducted model calculations show that for many surface waters atmospheric deposition makes a significant contribution to the total input with (persistent) organic compounds. This is in particular true for the pesticides. The atmosphere is, e.g., for the North Sea the most important source of many pesticides. The atmosphere does also for fresh surface water often make a significant contribution to the total input. The Gezondheidsraad (Health Council) published an advice about the significance of the atmospheric dispersion of pesticides in 1998.
The uncertainty in the estimates that have been made is, however, very large. One of the reasons for this is the lack of good data about the emission of the different substances. Furthermore, measurements that enable validation of the estimates are lacking

S.1.1 The project

The Interdepartementale Werkgroep Atmosferische Depositie (Interdepartmental Working Party Atmospheric Deposition, IWAD) has already for some years been initiating research aimed at reducing the uncertainty in the estimates of the deposition of persistent compounds. After a first study, in which model calculations were carried out, an approach was set up to reduce the uncertainty by means of measurements. Subsequently, a first strategy for a monitoring programme was developed. This was followed by the development of monitoring methods in a pilot study, testing and small-scale implementation.
This study indicated that it is quite well possible to conduct measurements of deposition. Finally, a survey was held among all bodies involved in the problem of atmospheric deposition, such as water quality control bodies, Department of Public Works, and Ministries. The survey served to analyse the information requirement of these stakeholders. An extensive programme was set up on the basis of the results of that survey.
The programme was started in spring 1999; the objective was formulated as follows:
Determination of the input into the coastal waters and the North Sea (with the emphasis on the Dutch Continental Shelf) and the Dutch territory (water and soil) with persistent organic compounds (POPs) from the atmosphere and identification of the sources. The study results in insight into the atmospheric deposition at national as well as regional level. In the future, a strategy will have to be developed which is aimed at the monitoring of developments in deposition.

The programme was financed by contributions of the Ministry for Housing, Spatial Planning and the Environment (VROM), the Ministry of Public Works (via RIKZ, RIZA and TNO target subsidy), the Ministry of Agriculture, Nature Management and Fisheries (LNV, via TNO target subsidy), and STOWA. The programme was extended later in 1999 by contributions of the Province Noord Holland and the Department of Public Works, Directorate IJsselmeergebied. Financing of the monitoring programme stopped in autumn 2001. An additional contribution by RIVM, however, made it possible to carry out measurements throughout the total year 2001. This means that a total of two and a half monitoring years are now available.

After a preparatory phase during which sampling methods were developed, evaluated and tested, actual sampling was started in autumn 1999. There have been extensive reports on the results of the preparatory phase. Intermediate reports were prepared after the first half year and the first year during which measurements were carried our. An extensive report about the results in the calendar year 2000 was published in 2001.
This report now presents the results of the complete programme. This extended summary first describes the programme, followed by a presentation and discussion of the results. Finally, conclusions are drawn.

S.1.2 The processes

The substances studied are released into the atmosphere in different ways. PAHs are released during burning processes, PCBs evaporate from transformers and capacitors that are no longer in use, and pesticides evaporate during and after application. After getting into the air, the substances are transported via the air, and diluted. During transport through the air they are exposed to various processes. The most important are: adsorption to air-borne particles, transformation by ozone or radicals, photolysis, and deposition. Many of the substances studied here have a low vapour pressure and will rapidly be adsorbed to air-suspended material. They are then transported through the air as particles (fine dust or aerosol). Atmospheric deposition is the process during which substances are transported to the surface of the earth (land as well as water surface) where they are absorbed. Wet deposition is the process where substances are absorbed in precipitation. Dry deposition is the process where substances get into contact with the surface of the earth through turbulence where they are absorbed. Easily soluble substances can simply be absorbed in surface water.
Measurement of wet deposition is relatively simple. The concentration of a substance in precipitation is determined. Wet deposition is then the average concentration multiplied by the amount of precipitation. Measurement of dry deposition is quite difficult. In this study it is restricted to a measurement of the concentration in air. The deposition flux is calculated from this concentration by using the so-called dry deposition rate. This deposition rate depends on the substance properties and the properties of the surface on which deposition takes place.

S.2 Method

The programme includes measurements and model calculations.

S.2.1 Measurements

Precipitation and air samples were during a period of over two years taken at eighteen locations At most stations samples were taken during periods of four weeks. At three stations air samples were collected weekly. The air samples were collected by using high-volume pumps that suck in air through a filter-foam combination and an adsorbent. Precipitation samples were collected with a so-called wet-only sampler. The samples were analysed for a large number of compounds by means of gas chromatography coupled to mass spectrometry (GC/MS). The samples were analysed for their PCB, PAH and pesticide concentrations, including the seven *indicator PCBs* and the *sixteen PAHs from EPA*. It was also investigated to which extent PAHs were present in the gaseous phase or bound to particles. The pesticides concentration was determined in all samples; the PAH and PCB concentrations in the samples from nine stations.
A very large group of crop protection products is used in the Netherlands and other European countries. In the Netherlands alone, over 200 active substances are authorised as pesticides for use in agriculture. It is impossible to determine low concentrations of all used substances by means of one sampling method and analytical method. To restrict costs, it has therefore been decided to make a selection from the substances that are present, after which a specific method has been developed which enables determination of a large group of important substances. The following aspects have played a role in the selection:

- the presence of substances in surface water and in precipitation
- preferences of the stakeholders in the study
- a representative selection, considering use (herbicides, insecticides, etc.), cultures, and authorisation status (substances with or without authorisation)
- the possibility to be able to determine the substance in the expected, low, concentration.

During the first year, the samples from four locations have been analysed for a supplementary group of substances. The analytical package has on the basis of these results been adapted in the second year. During the first year, the presence of *triphenyltin* and some polar pesticides such as *carbendazim* and *diuron* has also been investigated at four locations. The limit of detection of this last group, however, was relatively high, which made the chance of detecting these substances small.

The distribution of the sampling locations over the Netherlands is such that a reasonable spatial distribution over the country is achieved. Furthermore, locations have been selected near areas with specific agricultural activities, with the associ-

ated use of pesticides. This method achieves a good spatial coverage as well as insight into the situation in the different regions with different agricultural activities. The locations of the monitoring stations have been selected carefully, with special attention for their position in relation to sources and the influence of obstacles such as buildings. The measurements at these locations present a good picture of the atmospheric deposition in the areas concerned and also give a good indication of the variation in atmospheric deposition over the Netherlands. According to a very rough analysis, the monitoring stations are representative at scales of 50 by 50 km. The choices that have been made mean that deposition closer to sources does not show up in the current monitoring network. Insight in the deposition at this scale requires a different experimental set-up.
Extra monitoring stations, financed by the province, were set up in Noord-Holland, resulting in a higher density of the monitoring stations in this province.

S.2.2 Model calculations

The dispersion of substances through the atmosphere is calculated with a dissipation model, in which the processes by which substances are removed from the atmosphere, such as dry deposition (vertical transport to the surface of the earth), wet deposition and degradation are taken into account.
A model enables calculation of the deposition over an area on the basis of data on the spatial distribution of emissions of substances, their physico-chemical properties, and meteorological conditions. Model calculations are attractive because they give insight into the relationship between emissions (and changes in these emissions) and deposition. In addition, calculations can be made for each location and depositions can be derived for a specific region (such as catchment areas).
These properties make models extremely suitable to study the possible effects of policy measures on deposition. The uncertainty in model calculation results, however, is often so large that the suitability for use in policy development is limited.
In the study described in this report, a first judgement is made about the quality of model calculations, on the basis of a comparison of measured and calculated concentrations and depositions for a large number of substances.

An additional advantage of model calculations is the possibility of so-called *source analysis*. Because the models enable establishment of a relationship between emission at a certain location and deposition somewhere else, it can be investigated to what extent certain sources are responsible for the deposition in an area.
In the study described here, calculations have been carried out on the basis of data about the emission from the Emissieregistratie (Emission Registration) (for PAHs) and an emission calculation for pesticides carried out by ALTERRA in the context of the evaluation of the MJP-G (Multi-Year Crop Protection Plan). The estimates of emissions of the other substances were taken from the Emission Registration.
The emissions of pesticides in surrounding countries were calculated byTNO.

S.3 Monitoring results

The measurements were extensively analysed. A large number of data, such as the average concentration and deposition of all substances, are presented in this report. These are discussed and compared with quality criteria for surface water. For a number of frequently found substances, the measured values have been compared with values calculated with the OPS dispersion model.
The most important results and conclusions are briefly discussed below.

S.3.1 Concentration in air

The PAH concentration in air samples could usually easily be measured. Determination of the PCB and pesticides concentrations was much more difficult. Many substances were found only a few times. A total of about 50 different pesticides were found one or more times in air, of which *chlorpropham, dichlobenil, DNOC, fluazinam, hexachlorobenzene, pentachlorophenol, procymidon, propachlor, triallate, trifluralin, chlorothalonil, kresoxim-methyl* and *vinclozolin* occurred frequently. In addition, some very persistent (and long forbidden) substances such as *endrin* and *telodrin, o,p-DDD* and *o,p-DDE* were found a few times. A large number of substances more recently forbidden in the Netherlands, such as *atrazine, endosulfan* etc. were regularly found. *Aldrin, deltamethrin, dicofol, disulfoton, fenitrothion, fenthion, phosphamidon, heptenophos, metamitron, methomyl, parathion-methyl, triadimenol* and *2.4-D* could not be detected in air samples.

S.3.2 Concentration in precipitation

The PAH concentration in precipitation samples could also easily be determined. PCBs and many pesticides could be detected much less frequently, although the detection was often less problematic than in the air samples. Also in precipitation, about 50 different pesticides were found, including a number of forbidden persistent substances such as *o,p-DDD* and a large number of substances recently forbidden in the Netherlands such as *trifluralin* and *atrazine*.
The following substances could not be detected in precipitation: *aldrin, bitertanol, deltamethrin, demeton-S-methyl, dicofol, disulfoton, endrin, phosphamidon, heptachlor, heptachlorepoxide, heptenophos, methomyl, o,p-DDE, telodrin, triadimenol* and *triazophos*.
A number samples were analysed by means of an LC-MS method, which is capable to detect the more polar pesticides. Of this group only *isoproturon* was regularly found.

S.3.3 Distribution over the phases

As stated above, substances in air are partitioned over the gaseous phase and the particle phase (suspended dust or aerosol), depending on their vapour pressure and particle concentration. This partitioning determines how the substance behaves in the atmosphere. The partitioning of a substance over these two phases is therefore important for the modelling of the dispersion through the air and deposition. The partitioning of the PAHs and PCBs over gaseous phase and aerosol phase, and the presence of substances in air or in precipitation can well be explained on the basis of theoretical considerations. For the pesticides, the agreement between the theoretical and observed partitioning is poor. No extensive research has been carried out into an explanation for this deviation. The quality of the parameters used in the equations such as vapour pressure and Henry coefficient, knowledge about the rate at which gaseous phase and particle phase reach an equilibrium in relation to the transport distance through the atmosphere, as well as measuring artefacts related to the limit of detection, probably play a role in this.

The extent to which a substance is absorbed in precipitation also depends on various physical parameters such as vapour pressure and solubility in precipitation. For PAHs and PCBs, there was good agreement between the expected ratio between the concentration in precipitation and the concentration in air on the one hand and the observed ratio on the other. For the group of pesticides, these ratios showed no agreement whatsoever.

S.3.4 Comparison with criteria

Currently there is hardly a quality criterion for the concentration of pesticides, PCBs and PAHs in air or precipitation. There is only a criterion for *benzo[a]pyrene* in air. The MTR value for the *benzo[a]pyrene* concentration (1 ng/m^3) in air, however, was not exceeded at any of the monitoring locations.

To still be able to make a judgement about the level of the observed concentrations, a comparison has been made with the quality criteria for surface water. Comparison with these criteria does therefore have no formal or scientific significance but serves as a first indication. The concentration in precipitation of eighteen different pesticides exceeded the MTR level for surface water one or several times. This concerned seven insecticides, four herbicides, four fungicides and two substances that are used as insecticide/acaricide. Furthermore, *methiocarb*, a substance with a wide field of use, was found above the MTR level. *Dichlorvos* and *chlorothalonil* were in more than 20% of the precipitation samples found above the MTR level. The *triphenyltin* concentration also often exceeded the MTR, mainly in Veendam and Yerseke. The concentration of this substance in Anna Paulowna and Lelystad sporadically exceeded the MTR. Of the PAHs, the *benzo[a]-anthracene*, *benzo[k]-fluorantene*, *fenantrene* and *fluorantene* concentrations sporadically exceeded the MTR level for surface water.

The concentration in precipitation of twenty two pesticides did one or several times exceed the drinking water criterion of 100 ng/l: eight fungicides, ten herbicides, two insecticides and two substances that are used as insecticide and acaricide. In particular the level and frequency of the exceedance for *propachlor* and *chlorpropham* are noteworthy. *DNOC,* a substance wide a wide field of use, exceeded the drinking water criterion at all stations. Of the polar pesticides, only *isoproturon* exceeded the drinking water criterion.

Markedly high *fenantrene*, *fluorante*ne and *pyrene* concentrations were also found. In the year 2000, the average Borneff PAH concentration in precipitation was slightly higher than the criterion of 200 ng/l. In 2001, the concentration was about 25% lower than this value. The b*enzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, indeno[g,h,i-cd]fluorantene* and *fluorantene* concentrations separately did a few times exceed the criteria of 200 ng/l in precipitation samples.

Exceedance of criteria in surface water as result of atmospheric deposition
It has been investigated by means of a simple model for the dispersion of substances in surface water for some substances to what extent atmospheric deposition may lead to exceedance of the quality criteria for surface water (MTR and NR, respectively). This investigation included various characteristic aquatic systems of which pools that are almost exclusively fed via precipitation are the most sensitive for exceedance of critical levels. These calculations show that the annual average load will probably not easily lead to exceedance of the MTR (maximum permissible level). This load did, however, for a number of substances lead to exceedance of the NR level (target level) in various waters.
Currently, criteria are only available for single substances. It is not clear to what extent the combination of a large number of substances leads to effects. This could, e.g., be investigated by means of bio-essays.

The model calculations have been made for nation-wide representative levels. Deposition of most substances, however, follows a strong pattern over the year. In addition, there is a wide variation in deposition over the country. The result of the calculations suggest that the high depositions that are in some areas measured in spring and summer could lead to exceedances of the MTR level during that period. Earlier research already showed that deposition closer to sources can still be much higher. Calculations with an atmospheric dispersion model show that deposition can at a distance of one kilometre from a source be a factor fifty higher than at a distance of ten kilometres. This means that the chance of critical levels being exceeded there is much higher. Deposition could still be much higher close to important source areas.

S.4 Input by atospheric deposition (loads)

The dry deposition load at the monitoring locations has been calculated on the basis of the concentration measured in air and an estimate of the deposition rate, based on simple models. This dry deposition is together with the measured wet deposition the total deposition on surface water and soil. The contribution of wet deposition to the total load by atmospheric deposition is lowest for the volatile PAHs and PCBs and highest for the less volatile PAHs and PCBs. For pesticides, the ratio between wet and dry deposition differs strongly for the various substances.

The table summarises the deposition on surface water in the Netherlands (excluding the large waters), Waddenzee, IJsselmeer and the Dutch soil for the year 2001. The deposition estimates are always based on data from the stations that are most representative for the area concerned. The depositions estimated for North Sea are uncertain because the course of the concentration over the North Sea has not been taken into account. Deposition on the part of the North Sea under consideration is probably lower than indicated here.
The deposition estimates for the province Noord-Holland have been included upon request of this province.

Table 1 Atmospheric deposition of pesticides on surface water in the Netherlands in the year 2000 (excluding large waters 2 793 km^2), surface water in Noord-Holland (800 km^2), Waddenzee (2 600 km^2), IJsselmeer[1] (1 928 km^2) and soil in the Netherlands (36 783 km^2) in (kg/yr). The estimates are based upon measurements of wet deposition, air concentration and an estimate of the dry deposition velocity

Atmospheric deposition	**Surface water The Netherlands 2001**	**Surface water Noord-Holland 2001**	**North Sea[2] 2001**	**Waddenzee 2001**	**IJsselmeer[1] 2001**	**Soil 2001**
	kg/yr	**kg/yr**	**t/yr**	**kg/yr**	**kg/yr**	**kg/yr**
2,4-D	4	1	1	5	2	49
Atrazine	25	6	3	17	13	333
Bentazon	7	1	0.04	1	6	65
Captan	85	29	4	30	53	432
Chlorpropham	290	142	15	382	218	2100
Chlorpyriphos-methyl	1	0.2	0.02	1	1	5
Chlorothalonil	38.1	15	3	61	30	298
Diazinon	1	1	0.1	2	1	11
Dichlobenil	443.7	140	42	352	356	362
Dichlorvos	19.3	4	0.2	5	8	132
Dimethoate	3	0.2	0.1	0	1	25
DNOC	1209	324	181	1207	879	14057
Endosulfan II	0.8	0.1	0	1	*0*	2
Epoxiconazole	6	*0*	-	*0*	-	39
Ethofumesate	34	7	5	23	19	267
Fluazinam	53	11	4	48	56	556
Fluroxypyr	9	4	*0*	10	3	13
Heptachlor epoxide	1.8	0		1	-	*0*
Hexachlorobenzene	2	1	0.1	2	1	8
Kresoxim-methyl	16	4	2	19	10	172
Lindane	23	8	2	15	11	248
MCPA	13	5	3	16	6	165
Mecoprop	12	4	1	13	6	153
Methiocarb	7	3	0.2	11	3	83
Metolachlor	6	2	1	3	1	72
Mevinfos	3	1	0.4	4	2	31
o,p'-DDD	1.1	0.02	0.5	1	1	14
Pentachlorophenol (PCP)	28	11	3	20	23	137
Pentachlorobenzene	0.6	0.3	0.1	1	1	6
Pirimicarb	1.1	0.3	*0*	0	1	13
Pirimiphos-methyl	352	88	50	312	397	456
Procymidone	13	6	1	14	6	109
Propachlor	194	50	15	188	279	1685
Propoxur	1	0.5	0.2	1	2	19
Terbutylazine	29	6	3	16	12	264
Tetrabromobisphenol A	7	3	0.3	2	5	92
Tolclofos-methyl	12	13	0.1	43	11	74
Triallate	80	14	10	69	47	115
Trifluralin	85	32	11	88	54	124
Vinclozolin	107	31	3	117	126	687

[1] IJsselmeer refers to the Public Waters in the IJsselmeer area.

[2] The deposition estimate for the North Sea is uncertain (see text).

Table 2 *Atmospheric deposition of PAHs and PCBs on surface water in the Netherlands (2 793 km^2), surface water in Noord-Holland (800 km^2), North Sea (508 126 km^2), Waddenzee (2 600 km^2), IJsselmeer[1)](1 928 km^2) and soil in the Netherlands (36 783 km^2) in (kg/yr). The estimates are based upon measurements of wet deposition, air concentration and an estimate of the dry deposition velocity*

Atmospheric deposition	**Surface water 2001**	**Surface water Noord-Holland 2001**	**North Sea[2)] 2001**	**Waddenzee 2001**	**IJsselmeer[1)] 2001**	**Soil 2001**
	kg/yr	**kg/yr**	**t/yr**	**kg/yr**	**kg/yr**	**kg/yr**
PAHs						
acenaphtene	454	139	92	836	268	498
acenaphtylene	44	13	2	78	23	98
anthracene	46	13	3	81	23	162
benzo[a]anthracene	19	6	3	35	12	207
benzo[a]pyrene	30	10	6	56	18	170
benzo[b]fluorantene	82	29	14	151	48	520
benzo[g,h,i]perylene	33	12	6	61	20	553
benzo[k]fluorantene	59	21	11	109	35	734
chrysene	56	17	8	105	35	426
dibenz[a,h]anthracene	7	3	1	14	5	74
fenantrene	1974	645	160	3553	1085	3176
fluorantene	710	262	68	1307	419	3646
fluorene	950	265	127	1738	551	953
indeno[1,2,3-cd]pyrene	32	11	7	58	18	263
naphthalene	1786	567	102	3448	1215	931
pyrene	382	121	32	692	215	1304
PCBs						
PCB-101	0.6	0.1	0.3	1.3	0.5	4.8
PCB-118	0.5	0.01	0.03	1.4	0.7	6.5
PCB-138	0.5	0.1	0.01	1.1	0.5	6.3
PCB-153	0.7	0.1	0.1	1.4	0.6	9.0
PCB-20	1.3	0.4	0.5	2.3	0.7	14.2
PCB-28	1.0	0.2	0.5	1.7	0.5	11.9
PCB-52	1.5	0.3	0.6	3.2	1.3	15.0
PCB-8	0.4	0.1	0.1	0.7	0.2	2.5

1) IJsselmeer refers to the Public Waters in the IJsselmeer area.

2) The deposition estimate for the North Sea is uncertain (see text).

PAHs and PCBs

High deposition values to surface water in the Netherlands were found for *fenantrene*, *naphthalene* and *fluorene*, *fluorantene* and *acenaphtene* with 0.5 tot 4 t in the year 2000. Deposition of most PAHs decreased in 2001 in comparison with the year 2000, by an average 40 per cent. Only the deposition of *dibenz[a,h]-anthracene* increased slightly. The deposition of PCB-52 on surface water in the Netherlands was relatively high in 2000, with over 4 kg per year. The deposition of the other PCBs ranged from 0.5 and 2.5 kg per year. The deposition of PCBs in the year 2001 had decreased by 30 per cent in comparison with the year 2000.

Pesticides

High deposition values for surface water in the Netherlands in the year 2000 were found for *DNOC* (2.4 t per year for the Netherlands), *dichlobenil* (0.7 t) and *chlorpropham* (0.6 t per year), *propachlor* (0.3 t per year) and *triallate* (0,2 t per year). High depositions of *chlorothalonil* (almost 0.1 t per year) and *vinclozolin* (0.08 t per year) were also found.

The average deposition of a number of pesticides in the Netherlands showed a strong decrease in 2001 in comparison with 2000: the deposition of *metolachlor* in 2001 is one fifth of the level in 2000, the deposition of *triallate* and *dimethoate* in 2001 is almost one third of the level in 2000, the deposition of *chlorpropham*, *DNOC*, *lindane* and *procymidon* is almost halved, the deposition of *propachlor* and *dichlobenil* decreased by a quarter to a third. The deposition of *2,4-D*, *bentazon*, *fluazinam*, *methiocarb* and *mevinphos* increased by a factor 2 to 4 in 2001.

This analysis shows that depositions and concentrations of pesticides can from year to year differ by a factor two. These differences are probably associated with differences in use from year to year. In 2001, e.g., *S-metolachlor* was often used instead of *metolachlor*. The difference between the concentration of most PAHs in the two monitoring years is much smaller.

The deposition of *carbendazim* and *glyphosate* was only estimated by means of model calculations. This means that the uncertainty in these estimates is high

Regional differences

There is a strong variation in deposition over the country. This is associated with the use of these substances in the different regions. The difference in deposition between the different regions can reach a factor of ten. Striking patterns were found for substances such as *fluazinam*, *dimethoate* and *chlorothalonil* that are used in potato growing, and substances such as *procymidon*, *tolclofos-methyl*, *vinclozolin* and *chlorpropham* that are used in bulb growing and in potato storage. A good relationship with cropping could not established for all substances. The distribution of the deposition of *atrazine* and *trifluralin* over the country indicates that these substances are probably transported to the Netherlands from surrounding countries. The deposition pattern for *terbutylazine* also suggests a considerable contribution from surrounding countries.

Re-emission

It was investigated to what extent dry deposition is affected by the presence of substances in the compartment of the environment on which deposition takes place. This may occur in case a high concentration of a poorly soluble substance is present in water. Discharges, drift or other processes such as wet deposition may cause the presence of the substance. High concentrations of the substance in the water and low concentrations in the air will restrict the dry deposition of the substance and in the extreme case even fully reverse the direction of the flux in which case there will be no dry deposition but emission from water to air. To gain insight into the significance of this process, surface water samples (IJsselmeer, Oosterschelde and North Sea), soil, and grass were taken in December 2000 and were analysed with the standard analytical package. These monitoring data and model calculations with a simple exchange model indicate that dry deposition of a number of substances is restricted or does not take place at all. Although the number of monitoring data were only small, there are concrete indications that *atrazine, hexachlorobenzene* and *triallate* are sooner evaporating from surface water than that they are absorbed in surface water by dry deposition. The substances already present in the surface water limit the deposition of dichlobenil and mecoprop. The dry deposition flux of these substances is probably overestimated. Proper insight into the actual significance of this re-emission process requires more detailed research.

S.5 Discussion

S.5.1 Comparison with earlier studies

With about 0.1 % of the amount of active substance used, the atmospheric deposition load with pesticides is about a factor ten higher than the estimate given in the MJP-G. The MJP-G estimate, however, has been made for all pesticides used in the Netherlands and is therefore possibly not representative of the group discussed here.
The reports by the Hoofdinspectie Milieuhygiëne (Main Inspectorate Environmental Hygiene) also include an estimate the input into surface water by atmospheric deposition for some of substances. These reports seem to overestimate the atmospheric deposition of the various PAHs by a factor three to four.

S.5.2 Trends

The deposition on the Netherlands as derived from the measurements is usually lower than the atmospheric deposition calculated with dispersion models for the year 1990. The current deposition of PAHs and PCBs is a factor four and a factor two lower, respectively, than was estimated for the year 1990. The depositions of *endosulfan, lindane* and *trifluralin,* however, differ a factor 10 to 50. The low values for the deposition of *parathion-ethyl, mevinphos, pentachlorophenol, simazine, bentazon* and *atrazine* are also noteworthy. The differences are probably caused by a strong decrease in the use of these substances over the last five years. Precisely the substances that received most attention at that time have meanwhile been replaced by other substances. Despite this trend, *non-authorised* substances are still regularly found. These are substances such as *atrazine, endosulfan, propachlor, trifluralin* and *mevinphos*. These substances were all found in precipitation in 2001. For *atrazine,* which is from the year 2000 no longer authorised in the Netherlands, *trifluralin* and *lindane,* sources from surrounding countries also seem to make a significant contribution.

S.5.3 Atmospheric deposition compared with other sources

Except from the atmosphere, surface water is burdened with substances via various other sources. The input by atmospheric deposition has been compared with the input via other routes. Few data, however, are available.
For the PAHs and some other substances, a comparison with the input via other sources could be made on the basis of the Milieumonitor (Environment Monitor of the Hoofdinspectie Milieuhygiëne). For all substances, the measured atmospheric deposition is of the same order of magnitude as the direct input and the direct input via effluents. The atmospheric route is by far the most important route for *HCB, PCBs* and *pentachlorophenol*.

The input into the different environmental compartments with crop protection products by processes such as drift and lateral leaching have been estimated in the context of the MJP-G evaluation. This estimate can for a restricted number of substances be compared with the direct atmospheric deposition on surface water in the Netherlands derived from the measurements. The contribution of atmospheric deposition to surface water input for this group of substances amounts to between 10 and 140% of the input by drift. It is noteworthy that the input by atmospheric deposition is much higher for *fluazinam*. Averaged over all studied substances, about 0.1% of the used amount of active substance gets into the surface water of the Netherlands via direct atmospheric deposition.

The MJP-G study also shows that the average input of pesticides via drift and lateral leaching amounts to 0.22% of the used amount of active substance. The input by atmospheric deposition is at least of the same order of magnitude, certainly when indirect input is taken into account. For some substances, such as *propachlor* and *vinclozolin,* deposition to water, with about one per cent of the used amount of active substance, can even be qualified as high. An important difference, however, is that the input via drift only gets onto a small part of the surface water whereas atmospheric deposition gets onto the total Dutch surface water. This means that drift will in a restricted number of small waters lead to high concentrations whereas atmospheric deposition leads to lower concentrations of pesticides in all waters.

Part of the deposition on soil will also reach the surface water via leaching and runoff. Estimates of the contribution of this indirect deposition via leaching and runoff from the soil, however, are relatively uncertain. The contribution by this indirect load has not been included in the estimates presented here. The contribution of indirect deposition is, according to a first estimate (for the ZHEW area), of the same order of magnitude as direct atmospheric deposition.

S.5.4 Model calculations

For twenty substances, which were regularly detected, model calculations have been carried out as well. The calculations were carried out with the OPS model, on the basis of the emissions for pesticides by agriculture as prepared by Alterra and the data from the emission registration for *fluorantene* and *benzo[a]pyrene*. In addition, the emission of substances in surrounding countries was - specifically for this study - estimated. All calculations were carried out for the year 2000.
The agreement between the measured and calculated wet deposition differed strongly between the various pesticides. The quality of the comparison is poor for *bentazon, dimethoate, tolclofosmethyl, DNOC* and *pirimiphos-methyl*. For the first three substances this is probably caused by the quality of the measuring results where only few concentrations were found above the limit of detection. The cause is less clear for *pirimiphos-methyl* and *DNOC*. There was a good fit for c*hlorpropham, chlorothalonil, ethofumesate, dichlorvos, procymidon, propachlor, terbutylazine* and *vinclozolin*. The average difference between calculated and measured deposition for these substances was less than a factor two. This suggests that the estimates that have been made of the magnitude of the emission and the variation over the country and the calculations of the dispersion through the atmosphere are reasonable. Larger differences, up to a factor ten, were found for *fluazinam, dichlobenil, dimethoate* and *triallate*. The results of the comparison between the calculated and measured deposition of *fluazinam* are noteworthy. The calculated levels of the substance are a factor ten higher than the measured levels, whereas the correlation between the concentrations at the various locations is reasonable. Extensive research to find an explanation for the differences that were found, has not been carried out.
On the basis of the differences that were found for *atrazine, dichlobenil, DNOC* and *triallate* it is concluded that the estimated emissions for these substances present no good picture of the actual situation or that the parameters that have been used for transformation or deposition are incorrect.

The deposition calculated for *fluorantene* (a gaseous PAH) and for *benzo[a]pyrene* also showed a considerable difference from the measured deposition. The model underestimates the measured deposition of *benzo[a]pyrene* by a factor three. The model overestimates the wet deposition of *fluorantene* by a factor two.

S.5.5 Contribution by the various source areas in the Netherlands and surrounding countries

The possibilities of the model to estimate the contribution of various source areas in the Netherlands to the deposition are illustrated by means of some examples. The deposition of *benzo[a]pyrene, procymidon, fluazinam, dichlorvos* and *chlorothalonil* as result of emissions in the different provinces has been calculated for all water quality areas. The contribution by the different source areas is strongly associated with the position in relation to the particular water quality area. The deposition of a large number of pesticides in the area of Zuiveringsschap Hollandse Eilanden and Values (ZHEW) is, for instance, for 25 tot 30% caused by emissions in the area whereas this contribution is about 90% for PAHs.

Model calculations also enable investigation of the contribution of surrounding countries to the deposition. Good estimates of the emissions in the surrounding countries, however, are lacking. In the context of the current study, estimates have been made on the basis of the use in the Netherlands in the various cultures and information about land use in surrounding countries.
The result for *atrazine* is interesting. In the calculations it was assumed that the substance was not used in the Netherlands. The good agreement between the calculated and measured *atrazine* concentration is again a strong indication that the deposition of this substance, which is not authorised in the Netherlands, is caused by emissions in surrounding countries. For *trifluralin* and possibly for *lindane*, surrounding countries do probably also make a significant contribution to the deposition.

S.6 Conclusions

The report above mainly deals with the scientific conclusions that can be drawn from the study. The conclusions that can be drawn from the study are in the following again summarised, this time focussed on policy development. The possibilities for monitoring developments in deposition in the years ahead are also discussed.

The following conclusions can be drawn on the basis of the monitoring programme described in this report:

- Associated with the use in certain periods and areas, the deposition of pesticides shows a strong variation throughout the year and throughout the country. The annual deposition can also, through differences in use, from year to year show differences up to a factor two.
- Substances that are no longer authorised are in the year 2001 still found in precipitation and air in the Netherlands, of which the sources should partly be sought in the Netherlands. For substances such as atrazine and trifluralin, sources outside the Netherlands seem to make a significant contribution to the input. This is probably also true for some long forbidden, persistent substances such as endrin and o,p-DDE (a transformation product of DDT). The contribution from surrounding countries is less important for most other substances.
- The concentration of eighteen different pesticides and four PAHs in precipitation regularly exceeds the MTR level for surface water . The concentration of twenty two pesticides and some PAHs do also exceed the drinking water criterion. The average concentration Borneff PAH exceeds the criterion for drinking water in the year 2000.

- Atmospheric deposition forms an important input into inland surface water by some pesticides and PAHs:
  1. The measured atmospheric deposition into surface water (expressed as annual load) with pesticides in the Netherlands is of the same order of magnitude as the input by drift or lateral leaching.
  2. The measured input by atmospheric deposition of PAHs (annual load) is almost just as large as the other forms of input.
  3. For HCB, PCBs and *pentachlorophenol*, the atmospheric route is much more important than other routes
- Calculations for some substances with a simple model show that the annual average input into surface water by atmospheric deposition alone does probably not lead to exceedance of the MTR in surface water. Atmospheric deposition can, however, contribute to the input by other sources, which may result in earlier exceedance. For some pesticides, the high input by atmospheric deposition in certain periods of the year and in specific regions may, however, lead to exceedance of the MTR in that period.
- The model calculations do for a number of substances indicate that the NR (Negligible Risk) level in surface water can be exceeded by atmospheric deposition alone.
- Atmospheric deposition into soil in the Netherlands (annual load) has also been estimated. Due to lacking knowledge and reliable data, estimates of the deposition on soil, however, are relatively uncertain. Comparison with the input by other sources is often difficult due to the lack of good data. For nature areas, however, atmospheric deposition is often the only source.
- Generally, the OPS model is suitable for calculation of the deposition of pesticides. The difference between measured and calculated depositions was less than a factor two for over half of the substances. Larger differences were found for the rest of the substances. These differences are probably caused by inaccuracies in the emission estimates that have been used. The differences between calculated and measured deposition of two different PAHs were a factor two to three.

The conclusions above have been derived from the current study. Despite the broad approach, there are of course some restrictions that are associated with the set-up that has been chosen:

- A representative product package has been selected. An important group of pesticides such as the bis-dithiocarbamates (maneb etc.) and some important substances such as glyphosate, diuron and carbendazim, however, are not -or only to a limited extent- included in the study.
- The location of the monitoring stations was aimed at obtaining a national picture with specific attention for differences between the regions. The monitoring locations are therefore situated in such a way that the influence by local sources is minimal. Deposition in the direct surrounding of sources, e.g. within a radius of one kilometre around important sources, could be considerably higher and could easily lead to exceedance of MTR levels.

- Besides direct input, there is also indirect input. This is deposition on land which gets into the surface water through runoff. The conclusions above are based on the measured deposition excluding this indirect deposition. The magnitude of this indirect input, however, is very uncertain. First estimates show that this route could be equally large as direct deposition, as a result of which the total atmospheric input could still become twice as high.
- Comparison with criteria concentrations such as the MTR is only possible for individual substances. The effect of the deposition by a combination of a large number of substances has not been investigated.

S.7 Recommendations

S.7.1 Research

The deposition values presented here have been derived from measurements at eighteen stations spread over the country. This results in a representative picture of deposition on the Netherlands. The use of measurements makes the quality of the estimated loads relatively high in comparison with the estimates that were made in the past on the basis of model calculations. The uncertainty in the calculated surface water input was estimated at a factor 5-10. Deposition was in this study for the first time also determined on the basis of measurements. A comparison between measurements and model calculations shows that the model is often capable to estimate wet deposition within a factor two. This seems to justify the conclusion that deposition can properly be determined by means of the current methods where it is important that the current pesticide emission estimates have been generated according to a new, detailed calculation method. Improvement of the quality can, in as far as the quality of the measurements is concerned, still be reached by:

1) Improvement of the sensitivity of the chemical analysis
An improved accuracy can still be reached for a number of substances by increasing the sensitivity of the chemical analysis for some substances. For most substances, however, the accuracy is sufficient. The sensitivity for polar pesticides, however, is generally too low.

2) Increasing the knowledge about dry deposition
An uncertain variable is the value of the dry deposition rate to soil, vegetation and surface water. The dry deposition is now derived from measurements of the concentration in air and an estimate of the deposition rate. The uncertainty in this deposition rate is fairly large and there is a need for validation of the estimates. The attention for the input of terrestrial ecosystems from the atmosphere is, except for acidifying substances, very limited. This is one of the reasons why little research has been carried out into the underlying processes and that knowledge is very limited. The process of re-emission also requires more knowledge.

3) Increasing the knowledge about indirect deposition
   The significance of indirect deposition (runoff etc) has not been included in this study. The uncertainty in the magnitude of this route is very large while the contribution is possibly large.

4) Improvement of the estimate of deposition on the North Sea
   Deposition on the North Sea has now been estimated on the basis of measurements close to the Dutch coast. This means that the course in concentration from coast to more open sea has not been taken into account. Improvement of these estimates is possible by conducting measurements at relevant locations. An extensive model study could also increase insight.

Apart from this, the quality of the model calculations of the deposition of pesticides can be increased by improvements in:

1) Data about the emission of pesticides in surrounding countries. The estimates used here are mainly based on data about Dutch agriculture. Good data about the use in surrounding countries are lacking.

2) The quality of physical-chemical parameters such as the Henry constant and vapour pressure. The uncertainty about the vapour pressure is especially large for substances with a low volatility. The knowledge about the transformation rate of substances in the atmosphere is very limited. This leads to large uncertainties about the contribution of the input by substances from sources from surrounding countries.

3) As mentioned above, there is a need for knowledge about the process of dry deposition and indirect deposition.

A very large amount of data have been collected in the study presented here. The analysis of the data is very time-consuming and was primarily aimed at main points. There is certainly room for a more detailed, scientific analysis of the monitoring data. Model calculations could of course play a role in an extensive analysis. A further analysis of the cause of the differences between the measured and calculated depositions such as this was observed for some substances could be the starting point for this activity.

S.7.2 Transfer of information regarding atmospheric deposition

The current study also holds leads for the future policy regarding atmospheric deposition. A number of aspects is reviewed below.

Monitoring atmospheric deposition in the future

The magnitude of the atmospheric deposition as diffuse source for surface water, as determined in this study, seems so large that it may be expected that the need for data about the significance of this process remains. Based on the comparison between the results of model calculations and measurements, the conclusion that suitable models are available seems also justified. It is also technically quite well possible to determine the deposition at a national scale by means of measurements. The costs of the monitoring programme seem reasonable.

Measurements as well as model calculations are therefore available for the development of a strategy aimed at deposition monitoring in the years ahead. The concrete objective and the possible costs in relation to available budgets are in particular relevant for the development of such a strategy. A combination of model calculations and measurements is the most likely option. A monitoring programme could then be aimed at mapping the distribution of the deposition over the country and at following the developments in the range of pesticides. Model calculations could be used for regionally mapping the deposition in relation to emissions and its developments.

TNO developed a strategy aimed at the information requirement of water quality control bodies in a study that was recently carried out for RIZA (Duyzer *et al*., 2002), following from which three lines can be distinguished.

1) Model calculations for the Netherlands as a whole
   It is proposed to carry out calculations with a dispersion model in a formal route on the basis of data about the emission to air. These data can be obtained on the basis of the Emission Registration, in which estimates of the emissions of pesticides could be included. The results of the calculations can also be included in the Emission Registration. The advantage of this formal approach is that the quality of the data is guaranteed and maintained. A resolution of the data on a 5 by 5 km scale enables each water quality control body to collect data.

2) Measurement of the deposition at a national scale
   This route is in particular important for pesticides. Because the product range is continually changing, measurements are required to observe and monitor developments. Measurements can also play an important role in monitoring the effects of policies and to monitor the use within and outside the Netherlands. (Large-scale) illegal use could also be detected. For obtaining a national picture, the efforts could be restricted to some monitoring stations. A good picture of the large differences in deposition over the Netherlands could probably be obtained by spreading five stations according to a dice configuration. This would enable mapping of large differences between North, South, East and West, for which it is important that these stations monitor a wide range of substances with a high sensitivity as well as regular adaptation of the range to developments. The input into specific regions could be estimated on the basis of model calculations based on detailed emission estimates. Model calculations can in any case yield a reasonable estimate.

3) Measurement of deposition at a regional scale
   Single monitoring stations can be set up in case data are required on a regional scale, for which a specific product package can be specified. Connection with the national monitoring activities enables comparison of the results.

S.7.3 Atmospheric deposition in the authorisation policy

The input into surface water in the Netherlands via atmospheric deposition appears to be of the same order of magnitude as the input via drift and lateral leaching. The contribution of indirect deposition can result in a considerable further increase of the load.
An important difference, however, is that all input by drift takes place on a relatively small water area whereas atmospheric deposition takes place all over the Netherlands. This means that input by drift will sooner lead to high concentrations and criterion exceedance. Atmospheric deposition causes a relatively low input to large areas. This means that the average level will not easily lead to criterion exceedances in those areas. Measurements, however, show at the same time that in concentration areas, in periods in which the substances are used, deposition can be up to a factor ten higher. Although this has not been investigated in the current study, it is clear that still closer to the source, within some kilometres from sources or source areas, concentration and deposition could still be much higher.
Atmospheric deposition thus seems to be an important diffuse source for surface water. High loads in certain periods in source areas probably lead to exceedance of MTR levels. In view of this situation it seems meaningful to investigate how atmospheric deposition can be brought into the authorisation policy.

The question is to what extent the deposition of crop protection products could be restricted. An extensive discussion of the possibilities falls outside the scope of this study. Here, we therefore limit ourselves to some remarks of a general nature. In a recently conducted study of CLM (Centrum voor Landbouw en Milieu; Centre for Agriculture and Environment) this subject is discussed in more detail. A first important fact is that deposition is virtually proportional to the emission to air. Furthermore, the model calculations show that the deposition in the different water quality areas can often be related to emissions in the area (within the province). This means that restrictions in use and emission in the Netherlands do directly lead to a reduction in deposition. Choosing substances with a shorter residence time in air, e.g. caused by a higher transformation rate, does also lead to a reduction of the deposition. The residence time in air could therefore be taken into account in the authorisation of pesticides. This, however, is a complex matter for which a large number of basic data are lacking. Moreover, the effect of a high transformation rate (i.e. a short residence in air) will be rather restricted in the direct vicinity of the source, at a scale of tens of kilometres where deposition is highest. The advantage of substances with a high transformation rate is more significant at larger distances from the source.

Sources in surrounding countries play a significant role in as far as a number of substances are concerned that are no longer from these sources. Here, the international authorisation policy would have to play a role.

Pilot study
A pilot study into the possibilities to include atmospheric deposition in the authorisation policy seems meaningful. It could be investigated to what extent physical-chemical parameters do in practice affect transport over medium-range distances and which other aspects are relevant here. The study could be aimed at the medium-range distance from the more or less direct vicinity of the source to hundreds of kilometres. Deposition in the direct vicinity of the source has not been investigated in the current study. Deposition in the direct vicinity is probably much higher than at the current monitoring stations. The chance of exceedance of criterion concentrations and effects in the direct vicinity of sources, within some kilometres, is much higher than at a national scale. Furthermore, it seems useful to make calculations for a number of realistic situations.

S.7.4 Other substances

The now completed study was especially aimed at organic compounds with the emphasis on persistent compounds and pesticides. Earlier research already showed that input from the atmosphere can also be important for other substances. Moreover, there are recent indications that the atmospheric deposition of metals, such as zinc, is higher than assumed before. Recently, it was also demonstrated that precipitation can have oestrogenic potential. This potential could be related to the presence of *organochlor* pesticides. The significance of these research results is still subject of further study.
It is suggested in a recently reported study that the input into estuaries and coastal waters with nutrients from the atmosphere may have a significant effect. An influence on known effects such as algal bloom and ultimately even a loss of biodiversity has been suggested. It is for the time being not clear to what extent the results of this North American study are also significant for Northern Europe. It seems meaningful to conduct - in the near future- an inventory study into the significance of the deposition of metals, oestrogenically active compounds, and nutrients for the quality of surface water.

1. Introduction

Substances get into surface water via different routes, where direct discharges, run-off and drift are important. Substances can also be transported from the atmosphere to the surface of the earth, bound to precipitation and by direct absorption from the air. Recently conducted model calculations show that input via the atmosphere makes a significant contribution to the total load with persistent organic compounds for many surface waters (Baart et al., 1995; Warmenhoven et al., 1989a). This is especially true for the pesticides. The atmosphere is, e.g., the most important source of many pesticides for the North Sea. The contribution for the studied substances was often higher than 90% (Baart et al., 1995). The atmosphere does often also make an important contribution to the input into fresh surface water (Warmenhoven et al., 1989b).
The uncertainty in the estimates, however, is large (Baart et al., 1995). One of the reasons for this is the lack of good data about the emission of the different substances. Moreover, measurements that enable validation of the estimates are lacking. In 1998, the Gezondheidsraad (Health Council) published a report and an advice in which the significance of the atmospheric route for pesticides was indicated (Van Dijk et al., 1998).

1.1 The programme

The Interdepartementale Werkgroep Atmosferische Depositie (IWAD; Interdepartmental Working Party Atmospheric Deposition) is already for some years been working on the initiation of research aimed at reducing the uncertainty in estimates of the deposition of persistent compounds. After a first study, in which model calculations were carried out (Warmenhoven 1989a, 1989b), a line was set out to reduce the uncertainty by means of measurements, followed by the development of a first strategy for a monitoring programme. In a pilot study, measuring methods were then developed, tested and implemented on a small scale (Baas and Duyzer, 1997). This study indicated that deposition measurements could well be carried out. Finally, a survey was held among all stakeholders in the issue of atmospheric deposition, such as water quality control bodies, Rijkswaterstaat (Department of Public Works), and Ministries (Duyzer et al., 1998). The information requirement of the stakeholders was investigated in this survey. An extensive programme was set up on the basis of the results of this survey.
The programme was started in spring 1999. The objective of the study was formulated as follows:
Determination of the load of the coastal waters and the North Sea (with the emphasis on the Dutch Continental Shelf) and the Dutch territory (water and soil) with persistent organic compounds (POPs) from the atmosphere and identification of the sources of this load. The study results in insight into the significance of atmos-

pheric deposition at national as well as regional level. In the future a strategy will have to be developed which is aimed at monitoring developments in deposition.

The programme was financed by contributions of the Ministry for Housing, Spatial Planning and the Environment (VROM), the Ministry of Public Works (via RIKZ, RIZA and TNO target subsidy), the Ministry of Agriculture, Nature Management and Fisheries (LNV, via TNO target subsidy), and STOWA. The programme was extended later in 1999 by contributions of the Province Noord Holland and the Department of Public Works, Directorate IJsselmeergebied. Financing of the monitoring programme stopped in autumn 2001. An additional contribution by RIVM, however, made it possible to carry out measurements throughout the whole year 2001. This means that a total of two and a half monitoring years are now available. Sampling was started in autumn 1999 after interim reports about the preparatory phase during which sampling methods were developed, evaluated and tested. An extensive report has been published about the results of the preparatory phase (Duyzer et al., 1999). Interim reports were prepared after the first half year and the first year in which measurements were carried out (Duyzer, 2000a, 2000b). An extensive report about the measurements in the calendar year 2000 was published in 2001 (Duyzer and Vonk, 2001).
The results of the complete programme are presented in this report.

1.2 The processes

The substances studied here enter the atmosphere in different ways. PAHs are released during burning processes, PCBs evaporate from transformers and capacitors that are no longer in use, and pesticides evaporate during and after application. After getting into the air, the substances are transported via the air and are diluted. During transport through the air they are exposed to various processes. The most important are: adsorption to air-borne particles, transformation by ozone or radicals, photolysis, and deposition. Many of the substances studied here have a low vapour pressure and will rapidly be adsorbed to air-suspended material. Deposition is the process during which substances are absorbed by the surface of the earth. Wet deposition is the process during which substances are absorbed in precipitation. Dry deposition is the process during which substances get into contact with the surface of the earth through turbulence where they are absorbed. Easily soluble substances can in this way simply be absorbed in surface water.

Measurement of wet deposition is relatively simple. The concentration of a substance in precipitation is determined. Wet deposition is then the average concentration multiplied by the amount of precipitation. Measurement of dry deposition is very difficult. In this study it is restricted to measurement of the concentration in air. The deposition flux is calculated from this concentration by using the so-called dry deposition rate. This deposition rate depends on the substance properties and the properties of the surface on which deposition takes place.

1.3 Monitoring programme

Precipitation and air samples were during a period of over two years taken at eighteen locations The distribution of the sampling locations over the Netherlands is such that a reasonable spatial coverage of the country is achieved. Furthermore, locations have been selected in areas with specific agricultural activities, with the associated use of pesticides. This method achieves a good spatial coverage as well as insight into the situation in the different regions with different agricultural activities. The locations of the monitoring stations have been selected carefully, with special attention for their position in relation to sources and the influences of obstacles such as buildings. The measurements at these locations yield a good picture of the atmospheric deposition in the areas concerned and also give a good indication of the variation in atmospheric deposition over the Netherlands.

1.3.1 Monitoring locations

The position of the different locations and the analyses carried out on the samples taken at those locations are presented in Table 3. The location Lelystad-Oostvaardersplassen has in the course of the year 2000 been dismantled in view of vandalism, for which a new location, Lelystad-Houtribsluizen, was taken into use. The position of the monitoring locations is shown in Figure 1.

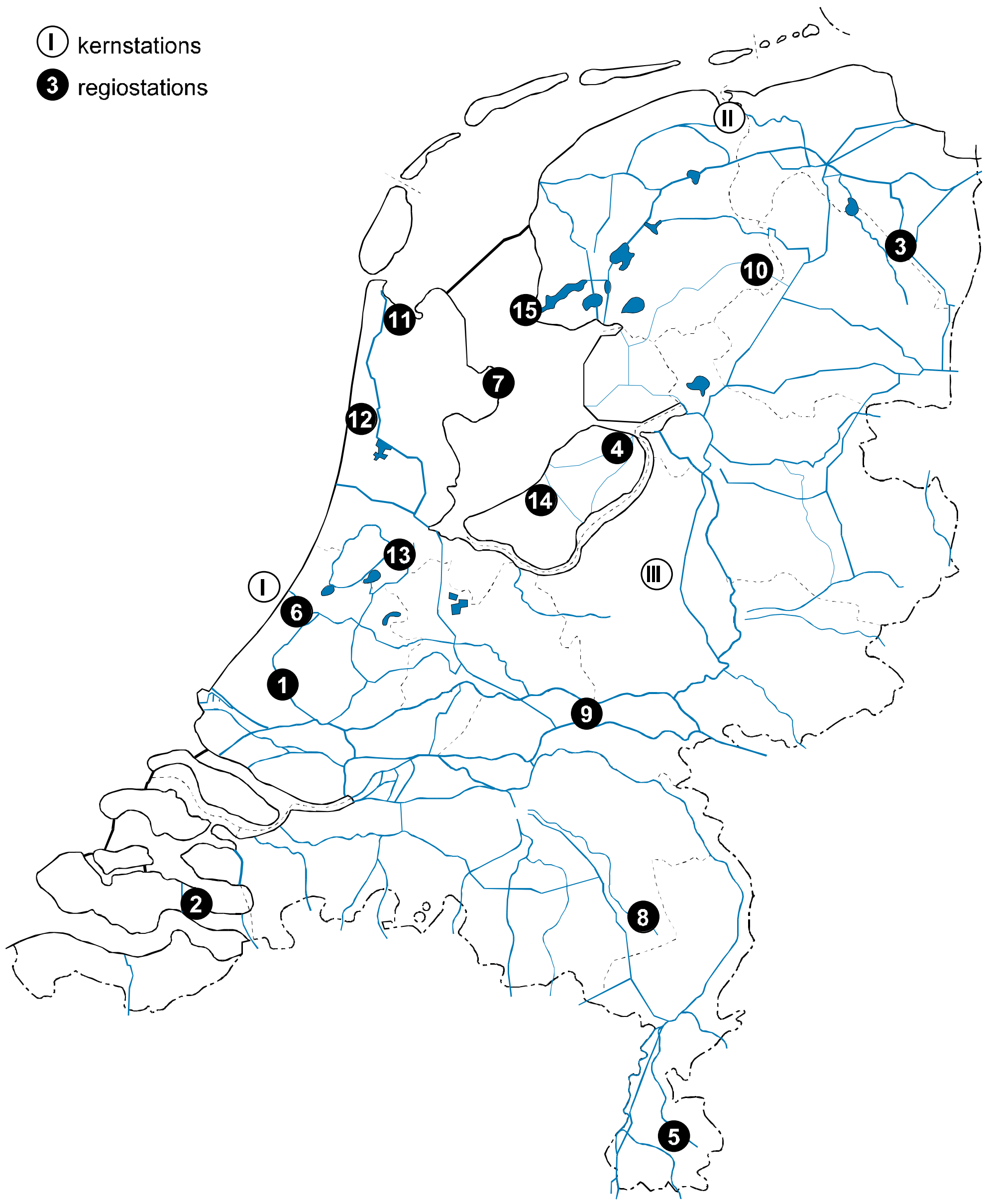


Figure 1 Location of the monitoring stations in the Netherlands. Kernstations = key station; regiostations = regional stations.

Table 3 *Position of locations where measurements were carried out. The co-ordinates and the 'controller' are presented, as well as the two-letter code for the station and the measurements carried out at the different locations.*

Number	**Name location**	**Code**	**Position**	**Type station**	**Co-ordinates**	**Controller**	**Start sampling**	**Extra measurements1)**
7	Andijk	(AN)	Noord-Holland (Oost)	R+	145.500-528.700	PWZN	16-9-1999	
13	Aalsmeer	(AL)	Noord-Holland (Zuid)	R+	119.500-478.300	Waterschap AGV	2-11-1999	
11	Anna Paulowna	(AP)	Noord-Holland (Noord)	R+	120.250-544.500	Waterschap the Hollandse Kroon	4-11-1999	OT, AL
9	Betuwe	(BE)	Betuwe	R	147.650-440.150	HHR Rivierenland	13-8-1999	
1	Delft	(DE)	Randstad	R	086.100-446.100	TNO	15-9-1999	AL
12	Egmond	(EG)	Noord-Holland (kuststrook)	R+	108.600-517.300	Waterschap the Lange Rond	5-11-1999	
10	Fochtelooerveen	(FO)	Zuid Oost Friesland	R	220.250-558.300	Natuurmonumenten	15-9-1999	
8	Groote Peel	(GP)	Noord-Limburg - Oost Brabant	R	184.200-370.800	Staatsbosbeheer	15-9-1999	AL
4	Ketelhaven	(KE)	Flevopolder	R	180.100-510.250	Fleverwaard	15-9-1999	
II	Lauwersmeer	(LA)	Waddenzee	K	211.460-600.200	Staatsbosbeheer	15-9-1999	
14*)	Lelystad	(LE)	Flevoland (Oostvaardersplassen)	R+	156.500-496.500	Staatsbosbeheer (regio Flevoland)	Ontmanteld 16-8-1999	OT
14	Lelystad	(LE)	Houtribsluizen	R		Rijkswaterstaat	15-9-2000	
I	Meetpost Noordwijk	(NO)	North Sea	K		RWS Dir. North Sea	8-9-1999	
III	Speulderveld	(SP)	Veluwe	K	177.700-476.000	RIVM	15-9-1999	
15	Stavoren	(ST)	Friesland	R+	153.500-533.500	Wetterskip Fryslan	8-12-1999	
3	Veendam	(VE)	Oost Groningen	R	256.450-5691.50	Zuiveringsbeheer Provincie Groningen	17-9-1999	OT, AL
5	Wijnandsrade	(WY)	Zuid Limburg	R	189.800-323.700	RIVM	15-9-1999	
2	Yerseke	(YE)	Zeeland	R	061.320-390.650	Waterschap Zeeuwse Eilanden	15-9-1999	OT
6	De Zilk	(ZI)	Zuid Holland coast	R	95.350-479.050	RIVM	15-9-1999	

R indicates a *regional station* where an air sample and a precipitation sample are taken every four weeks. The amount of pesticides is determined in both samples.

R+ indicates a *regional station* where an air sample and a precipitation sample are taken every four weeks. The amount of pesticides, PCBs and PAHs is determined in both samples.

K indicates a *key station* where a precipitation sample is taken every four weeks in which the amount of pesticides, PCBs and PAHs is determined. An air sample is taken each week, in which the same substances are determined.

**) dismantled*

1) AL = additional list of substances in precipitation and air samples in 2000
OT = organotin compounds in precipitation
PU = phenylurea compounds in precipitation

1.3.2 Sampling

An air sample and a precipitation sample were always, during periods of four weeks, taken at most stations (R, Table 3). The amount of pesticides was determined in both samples. At ten locations, the amount of PCBs and PAHs was determined besides the amount of pesticides (the so-called region plus stations, R+). At three key stations (K), spread over the country, air samples were collected weekly and precipitation samples monthly. These samples were analysed for pesticides, PCBs and PAHs. For key station Speulderveld, the fractions in which the substances in the air occurred in the particle phase and in the gaseous phase were determined.

1.3.3 Sampling method

The air samples were collected by sucking a known amount of air through a sampler by means of high-volume pumps. This sampler consists of a glass fibre particle filter, followed by a polyurethane-foam (PUF) in series with a styrene/diphenylbenzene polymer adsorbent (XAD). The particle filter and the absorbents are in the laboratory analysed to determine the concentrations of pesticides, PCBs and PAHs in the particle phase and gaseous phase. The method is extensively discussed by Duyzer et al. (1999). Precipitation samples were collected with a wet-only sampler. These samplers are only open when it rains, while precipitation is collected in a cooled (4°C) glass 10 litre bottle. There are a number of causes for a difference between the amount of precipitation collected with a wet-only sampler and the amount of precipitation collected by means of a standard rain collector. This is why the amount of precipitation, required for determination of wet deposition, is determined with a standard rain collector of the Hellman type, according to NEN 6585.

Figure 2 Precipitation collector and equipment for taking air samples at the Speulderveld location.

1.3.4 Chemical analysis

The air and precipitation samples have been analysed for a large number of compounds by means of gas chromatography coupled with mass spectrometry (GC/MS); the LC-MS method has been used for analysis of a number of polar pesticides.
The compounds for which the samples were analysed were selected on the basis of different criteria that have been discussed extensively in the research proposal. The samples have been analysed for their PCB, PAH and pesticides concentration, including the seven *indicator-PCBs* and the *sixteen PAHs* of *EPA*. It has also been investigated to what extent PAHs are present in the gaseous phase or bound to particles. The pesticides concentration was determined in all samples; PAH and PCB concentrations were determined in the samples of nine stations.
The selection of the pesticides for which the analyses should be carried out is more difficult. A very large group of crop protection products is used in the Netherlands and other European countries. In the Netherlands alone, three hundred pesticides are authorised for use in agriculture. A large group is found in surface water and in precipitation. The following aspects played a role in the selection:

- the presence of substances in surface water and in precipitation
- preferences of the organisations commissioning the study
- a representative selection, regarding field of use (herbicides, insecticides, etc.), crops, and authorisation status (substances with or without authorisation).

From a cost point of view it is impossible to determine low concentrations of all used substances by one sampling method and analytical method. To save costs, a

selection has been made from the substances that are present. Subsequently, a specific method has been developed which enables determination of a large group of important substances. Further details about the selection of the components for which analyses are to be carried out are given in Duyzer et al. (1999) and Duyzer and Vonk (2001). The complete list of analysed pesticides, PCBs and PAHs is given in Annex II.
During the first year, the samples from four locations have been analysed for a supplementary group of substances. The product package has in the second year been adapted on the basis of these analyses. In the year 2000, a number of samples was also analysed for a number of more polar compounds. The results of these analyses are given in Annex VIII.

1.4 Model calculations

The dispersion of substances through the atmosphere is calculated with a dispersion model in which the processes by which substances are removed from the atmosphere, such as dry deposition (adsorption to the surface of the earth without precipitation being involved), wet deposition (absorption in precipitation followed by absorption to the surface of the earth) and degradation, are taken into account.
A model enables calculation of the deposition over an area on the basis of data on the spatial distribution of emissions of substances, their physical-chemical properties, and meteorological conditions.
Model calculations are attractive because they give insight into the relationship between emissions (and changes in these emissions) and deposition. In addition, calculations can be carried out for each location and depositions can be derived for a specific region (such as catchment areas).
These properties make models extremely suitable to study the possible effects of policy measures on deposition. The uncertainty in model calculation results, however, is often so large that the suitability for use for policy development is disputable. The study described in this report presents a first judgement about the quality of model calculations based on the comparison of measured and calculated concentrations and depositions for a large number of substances.

An additional advantage of model calculations is the possibility of so-called *source analysis*. Because the models enable establishment of a relationship between emission at a certain location and deposition somewhere else, it can be investigated to what extent certain sources are responsible for the deposition in an area.

In the study described here, calculations have been carried out on the basis of data about the emission from the Emissieregistratie (Emission Registration) (for PAHs) and an emission inventory for pesticides carried out by ALTERRA in the context of the evaluation of the MJP-G (Multi-Year Crop Protection Plan).

2. Methods

2.1 Introduction

The way in which the measurement results are analysed is discussed in this chapter. Analysis of the measurement results is no simple matter. The concentrations of 16 PAHs, 10 PCBs and about 70 pesticides have been determined at 18 locations. Furthermore, supplementary monitoring data such as amount of precipitation, air temperature and sample volumes were collected separately. This yields almost 5000 monitoring data per month. Per year, this mounts up to over 50 000 monitoring data.
All data have been entered into a database from which summaries and profiles can be prepared in a relatively simple way. The possibilities to safeguard the quality of the monitoring data and derive representative monitoring results from the data is discussed below.

2.1.1 Analysis of the data

Determination of representative variables from the available monitoring data is often not simple because the concentration of some substances is not often found, and only slightly, above the limit of detection. This has been taken into account in determining the concentration of substances in the samples that have been taken. The limit of detection for each individual substance is estimated for each series of samples. All positive identifications of a substance, however, are reported. A value ‘0’ is only given in those cases that the substance cannot be identified. The effect of this procedure has been investigated in a sensitivity analysis. The procedure that has been followed is in Annex III.

The fact whether or not substances are present below the limit of detection does in particular present problems for substances such as pesticides, which may possibly during periods of the year not be present at all.
The substance may during three months exceed MTR values whereas it is not found in the other months. This then results in a very low calculated annual average and does not present a good picture of the presence of the substance.
In this case it is not beforehand clear how the data can best be presented. To overcome this problem, we do in this report besides average values also present the number of times that the substance is detected and that it exceeds the criteria if this would be the case.

Calculation of average variables
The average concentrations in air and precipitation are presented in the following chapter. The average concentration is calculated as the mathematical average of all samples. The samples in which the substance could not be detected have thus been

included in the calculation. This calculation method has also been used for the average concentration in the precipitation samples. Average concentrations of only those samples in which a substance was detected, were also calculated (see also Annex III).

2.2 Calculation of the atmospheric deposition from the monitoring results

The main objective of the study is determination of the input into the Netherlands with persistent organic compounds from the atmosphere. It is described in this chapter how deposition is derived from the monitoring data, where a distinction is made between dry and wet deposition. These are now discussed successively.

Wet deposition
Wet deposition is calculated from the measured concentration of substances in the precipitation ($C_{n,period}$) and the amount of precipitation over the four-weekly period (V_{period}).
The annual average wet deposition flux $F_{wet,year}$ (g/ha/year) is then calculated from the data for each four-weekly period:

$$F_{wet,year} = \sum_{period=1}^{13} \overline{c_{n,period} \cdot V_{period}}$$

Dry deposition
In case of dry deposition, substances (gases or particles) are absorbed to the surface of the earth without intervention of precipitation. Substances that are present in the air get into contact with the earth surface through turbulent air movements where they are absorbed. Easily soluble substances will, e.g., rapidly dissolve in surface water. Direct measurements of atmospheric deposition are in principle possible but they are hardly applied on a continuous basis. A hybrid method has been used in this study, which estimates the deposition on the basis of measurements of the aerial concentration and an estimate of the deposition rate.
The dry deposition flux ($F_{dry,year}$) is calculated from the measured annual average concentration of substances in air ($C_{air,year}$) and an estimate of the deposition rate (V_d):

$$F_{dry,year} = c_{air,year} \cdot v_d$$

The dry deposition rate of substances depends strongly on meteorological conditions and the surface on which deposition takes place. The deposition rate to water and soil has for the different substances been calculated in this study. The deposition rate to water has been calculated according to the method of Liss and Slater (1974) and the deposition rate to soil has been calculated by means of the *Simple-Box* model (Van der Meent, 1993). Further details are given in Annex I.

2.3 Calculation of atmospheric deposition by means of a dispersion and deposition model

2.3.1 Introduction

The calculation of dispersion and deposition of substances through the air first requires insight into the extent to which polluting substances are emitted into the atmosphere by the various sources. The way in which this information has been obtained is elaborated in Section 2.3.2.

After their emission, the substances will, depending on the meteorological conditions and their physical-chemical properties, disseminate via the atmosphere and eventually deposit on the surface of the earth. These processes can be described by means of a transport and deposition model (see Section 2.3.3).

The total input to surface water as result of atmospheric deposition does not only follow a direct route (atmospheric deposition) but there is also an indirect route. This indirect input causes an extra load into surface water in addition to the direct atmospheric deposition. Indirect deposition concerns the routes that are followed after a pollutant has been deposited on a different surface than water, such as run-off from the soil, hardened or not, input via sewage systems (treated/-untreated), etc. A method to derive indirect deposition besides direct deposition presented by Bleeker and Duyzer (2002). Indirect deposition has not been calculated in the study described here.

2.3.2 Emissions to air

The emission of a substance to the air is an important input parameter for a transport and emission model. The emission of pesticides is, generally, derived from the amount used in a certain area and the so-called emission factor. This emission factor, i.e., the fraction of a substance which evaporates during and after application, strongly depends on the vapour pressure of a substance. Various factors such as crop type, method of application, etc. also play a role. The uncertainty in the emission factor is usually high.
The amount of products used in a specific area is usually also uncertain. Here, the confidentiality of information also plays a role. A reasonable estimate of the total use in the Netherlands can usually, however, be made on the basis sales figures. The allocation to certain areas takes place on the basis of data about cultures.
The estimates of the emission of pesticides are therefore fairly uncertain while a consistent source of information such as the Emission Registration is lacking. This means that various information sources had to be used for the substances in the current study. These are briefly discussed below.

The emissions of *fluoranthene* and *benzo[a]pyrene* were derived from the Emission Registration (Bleeker and Duyzer, 2002).

Emissions from agriculture
The emissions (including the spatial distribution) of the Dutch agricultural sources have been calculated for the year 1998 and have in the context of this programme been made available by RIVM. The data have been prepared by the ALTERRA research organisation in the context of the evaluation of the Meerjarenplan Gewasbescherming 2000 (MJP-G- 2000; Multi-Year Crop Protection Plan). Because the use of substances varies from year to year, the emission data for the year 1998 have been adjusted on the basis of sales data for the years 2000 and 2001.

Emissions from surrounding countries
The emission of *fluoranthene* and *benzo[a]pyrene* has been derived from surveys carried out by TNO (see also Bleeker and Duyzer, 2002). Information about the emission of pesticides is not available in a uniform way for countries that are expected to make a significant contribution to the input in The Netherlands. These are primarily the countries: Belgium, Germany, England, France and Spain. To still be able to make an estimate, albeit fairly rough, of the emission in these countries, use factors for 1998, which have been derived from the Dutch data, have been used. These have then be combined with information about the spatial distribution of different types of land use, such as grassland, cereals, orchards, etc. The emission factors for the different pesticides have been derived from information about the total Dutch emission of the substance and the use of that substance according to CBS (Netherlands Bureau for Statistics).
The use of a substance per country has then been ‘scaled’ on the basis of information about the acreage on which the substance is (possibly) applied (EU, 2000). Such information was lacking for a number of substances so that estimates of the emissions from sources in surrounding countries could not be made in this way.
It will be clear that this procedure is not optimal and does not lead to the highest possible accuracy. The required data, however, are at this moment not (freely) available.

2.3.3 The transport and deposition model

The dispersion and deposition model OPS (Operationeel model Prioritaire Stoffen; Operational Model Priority Substances), developed by RIVM, has been used to calculate the dispersion and atmospheric deposition of the various polluting substances.

The OPS model
The OPS model calculates the concentration of a substance in air and precipitation at a certain place as result of an emission at a different place. Version 3 of the OPS model (Van Jaarsveld, 1989, 1995) has been used for the current study. The contri-

butions to the concentration and deposition at a certain place for which the calculation is carried out (receptor), are calculated separately for all sources by means of so-called back-casting trajectories. These trajectories describe the route followed by the air (holding the emitted substance) from the source to the receptor. The dispersion is calculated with a mathematical description of a plume, the so-called *Gaussian* plume formulation. The spatial resolution of the model is largely determined by the resolution of the emission files. The resolution may be of the order of 100x100 m around an individual point source; 5x5 km is a practical lower limit at a national scale.

The required input data are discussed:

Meteorological data

Meteorological parameters such as wind direction and wind speed do of course play an important role in the calculations mentioned above. The calculations reported here have been based on the meteorological data for the year 2000.

Substance-specific data

Substances may be degraded by reactions with ozone or hydroxyl radicals during transport through the atmosphere. In addition, there are losses and the concentration decreases through dry and wet deposition. The OPS model takes the effect of these loss processes into account. The OPS model holds substance-specific parameters for a number of substances. Substance-specific deposition rates have, e.g., been used, as these have also been used to calculate dry deposition from the measurements, derived by means of the *SimpleBox* model (Van der Meent, 1993) and the two film model for deposition to water (Liss and Slater, 1974) (Section 2.2.1 and Annex I). Besides, wet deposition is calculated by means of the so-called washout coefficient.

The degradation rate of pesticides in air has been derived by means of the programme OPWIN 1.88. (Atkinson, 1991). This programme calculates the transformation rate through hydroxyl (OH) radicals on the basis of structure characteristics of the substance.

Atmospheric deposition

Finally, the concentration above the receptor and deposition are calculated by means of the dispersion model, where the contribution of all sources is aggregated. The parameters used in the context of this study to calculate the deposition of pesticides are presented in Annex II.

The model yields the atmospheric deposition for each map square of 5x5 kilometre. The model can also determine the relationship between sources and deposition. The load of such a map square can be estimated, as well as the contribution of source areas (e.g., other provinces, catchment areas, surrounding countries) to the deposition on that map square.

3. Results

3.1 Introduction

The following subjects are successively discussed in this chapter:

- the concentration in air and in precipitation
- the input into surface water
- the comparison of modelled deposition with measured deposition
- the contribution of different source areas to the input into water management areas
- the modelling of the effect of atmospheric deposition on concentrations in some typical aquatic systems.

3.2 Concentrations in precipitation and air

3.2.1 Quality control

Duyzer et al. (1999) describe how the quality of sampling and chemical analyses were controlled during the programme. A good quality of the chemical analyses is therefore assumed when filling the database with the analytical results. Mistakes may, however, still arise as result of specific problems or artefacts during sampling. After filling the database, reviews have been prepared about the presence of each substance in air and in precipitation over the total database (see some examples below). All these reviews have been evaluated manually. This procedure ensures that outliers and changes in trends in the data are soon discovered, where the expected pattern of the presence of substances, etc. is taken into account. Extensive research into the quality of the analyses has been carried out in the preparatory phase of the study. The samples were analysed by means of the GC-MS method (see Duyzer et al., 1999). Despite identification on the basis of specific mass values, there is sometimes still doubt about the identification being correct. A check of the identification at a complete mass spectrum did for *azinphos-methyl, coumaphos* and *metabenzthiazuron* show that the identification on the basis of two peaks in the mass spectrum were not sufficiently conclusive. The analytical results of these substances have therefore not been included in this report.

Comparison with other measurements could still yield additional information about the quality of the monitoring results. These may be monitoring results described in the literature, or measurements simultaneously carried out by others. In the Netherlands, however, measurements of the now measured substances have hardly been carried out in recent years. Duyzer (2000b) already compared the PAH concentration measurements in this study with measurements carried out by RIVM. This learned that the ratios between the concentrations of the various PAHs found in the current study showed good agreement with the distribution found earlier by RIVM. Duyzer and Vonk (2001) still made a number of comparisons with measurements that were carried out in 1998 in the province Zuid-Holland (Van Zalinge, 2001) and measurements from an earlier TNO study in 1996 at monitoring locations De Zilk and Noordwijk.

RIVM is still conducting a restricted number of measurements of substances (including *lindane*) at monitoring location De Zilk of the national Meetnet Luchtkwaliteit (Monitoring Network Air Quality). Figure 3 shows the comparison of the monitoring results of RIVM and TNO. The course over time and the maximum values show reasonable agreement despite the differences in averaging periods. The agreement can be qualified as quite well. A significant difference between both results is the treatment of the samples in which lindane is not detected. For the RIVM samples, concentrations below the limit of detection are reported with a value. For the TNO samples, all positive identifications are reported. These differences in procedure lead to the apparent systematic difference in the results in the winter period.

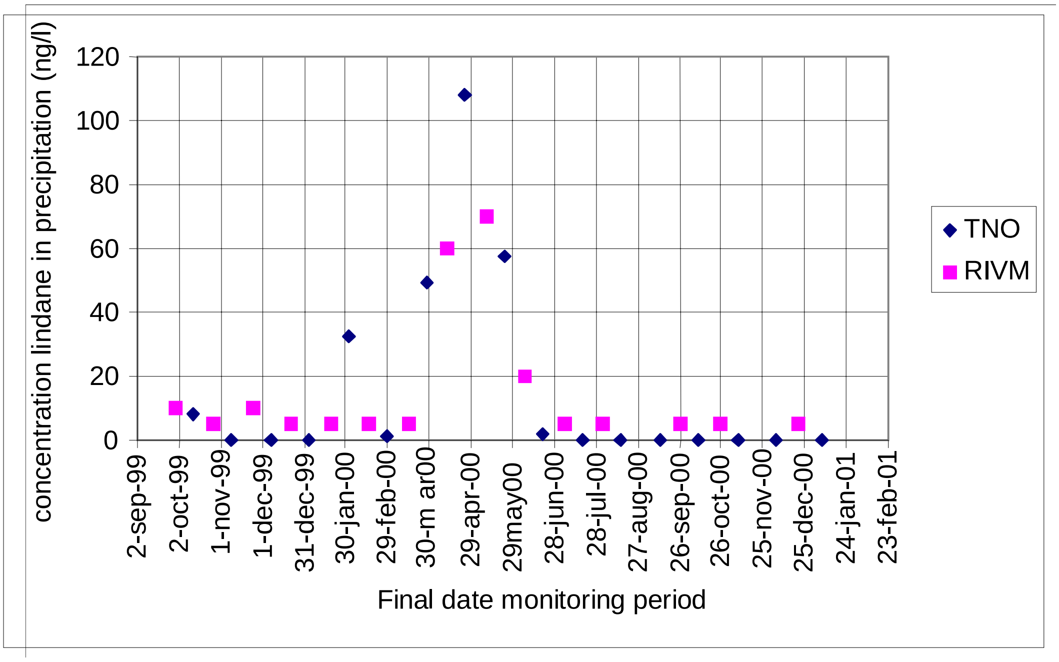


Figure 3 *Comparison of measured lindane concentrations in precipitation samples of RIVM monitoring station De Zilk.*

3.2.2 Partitioning over phases

As stated above, substances in air will partition over the gaseous phase and the particle phase (suspended substance or aerosol), depending on their vapour pressure and particle concentration. This partitioning determines the behaviour of the substance in the atmosphere. This means that the partitioning of a substance over both phases is important for the modelling of the dispersion through air and deposition. The partitioning of PAHs and PCBs over the gaseous phase and the aerosol phase and the presence of substances in air or in precipitation can well be described on the basis of theoretical considerations. For the pesticides, the agreement between theoretical and observed partitioning is poor. No extensive research has been carried out into an explanation for this deviation. The quality of the parameters used in the equations such as vapour pressure and Henry coefficient, the knowledge about the rate at which gaseous phase and particle phase reach an equilibrium in relation to the transport distance through the atmosphere, as well as measuring artefacts related to the limit of detection probably play a role in this.

The extent to which a substance is absorbed in precipitation also depends on various physical parameters such as vapour pressure and solubility in precipitation. For PAHs and PCBs, there was good agreement between the expected ratio between the concentration in precipitation and the concentration in air on the one hand and the observed ratio on the other. For the group of pesticides, these ratios showed no agreement whatsoever.

The partitioning of substances over the gaseous and aerosol phase and the ratio between the concentration in air and in precipitation is extensively discussed in Duyzer and Vonk (2001).

3.2.3 Concentration course during the monitoring period

Figures 4–11 show the concentration course over the years 2000 and 2001 for a number of pesticides, to which regulations are applicable. The figures illustrate differences in periods of application of the pesticides: a number is only used in a few months in spring. Examples are *propachlor* and *metolachlor*, whereas there are other pesticides, such as *trifluralin*, which are on the other hand used in autumn and winter. A number of pesticides such as *DNOC* and *dichlobenil* are used throughout the year and consequently also detected in air and precipitation samples.

Application in spring

Propachlor is a herbicide of which the authorisation has been withdrawn since August 2001. The substance is very persistent in water and is toxic to fish. It was still only authorised as an agriculturally indispensable substance for a number of fields of use, mainly in onion growing. Figure 4 and Figure 5 show the concentrations in air and precipitation for a number of stations where *propachlor* was detected most. The concentrations had already clearly decreased in the year 2001. Peak concentrations had decreased. *Propachlor* was in 2000 as well as in 2001 detected in about 20% of the precipitation samples, in concentrations exceeding the drinking water criterion of 100 ng/l.

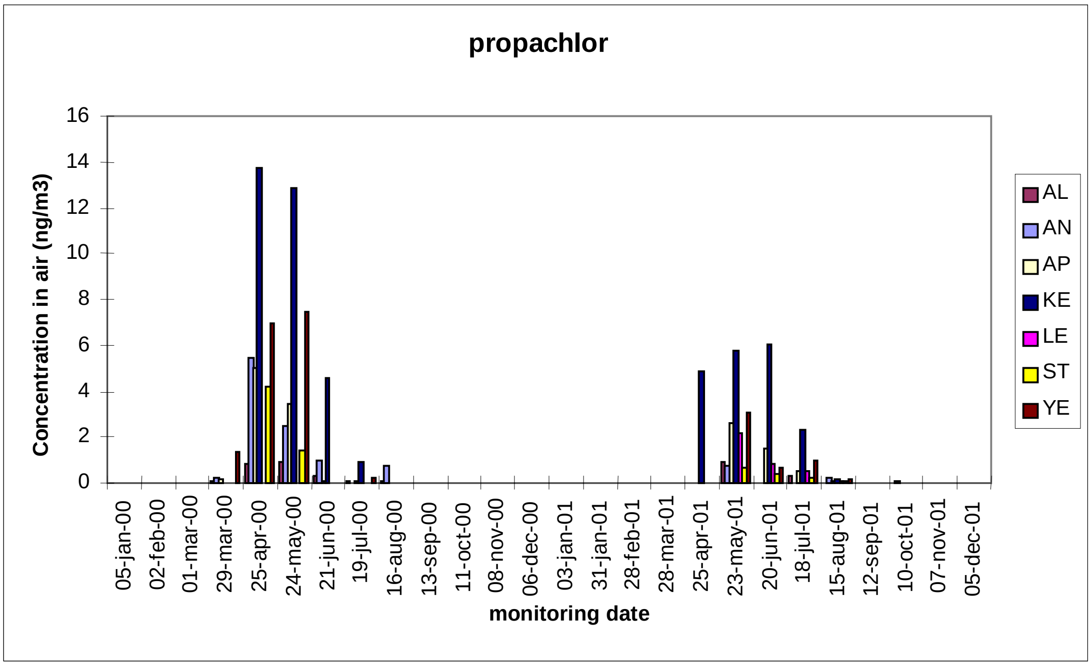


Figure 4 Course of the propachlor concentration in air for the different stations in 2000 and 2001.

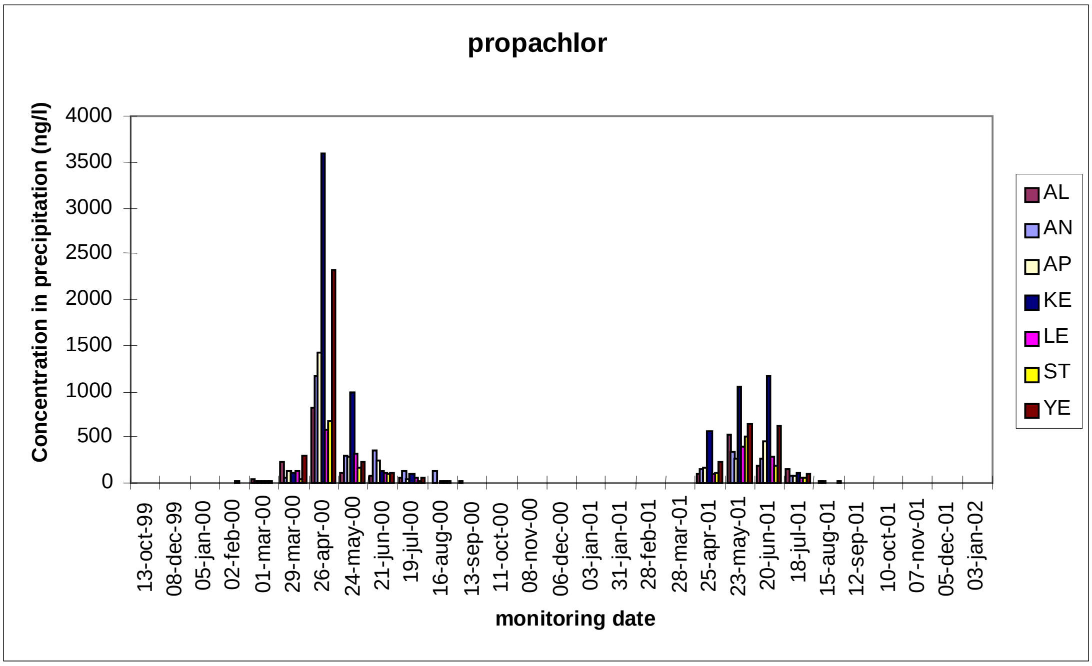


Figure 5 Course of the propachlor concentration in precipitation for the different stations in 2000 and 2001.

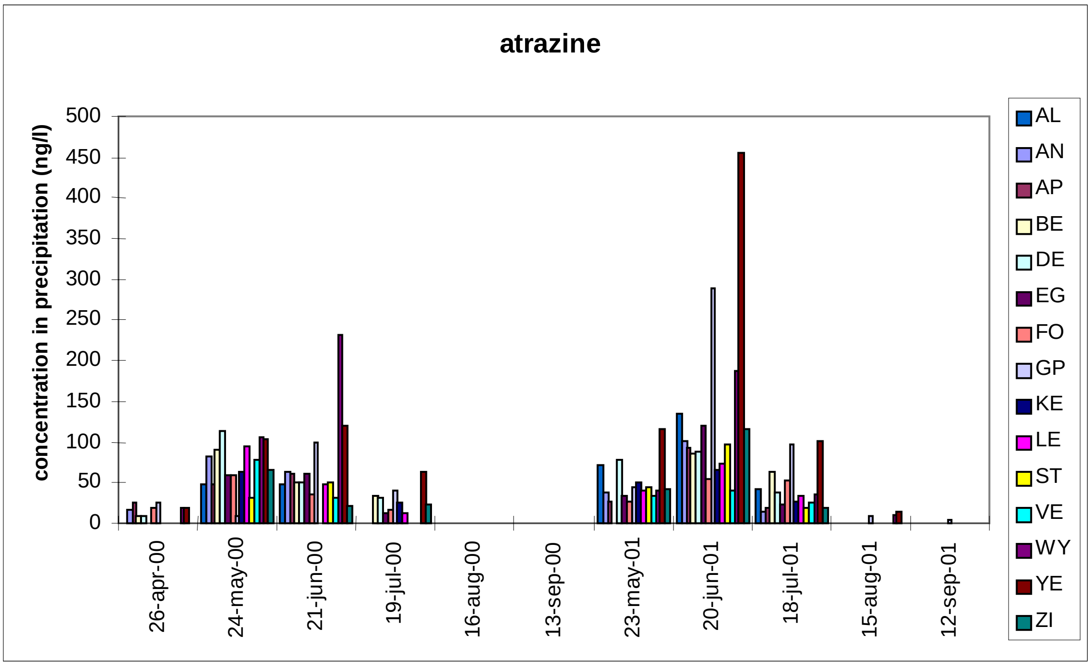


Figure 6 Course of the atrazine concentration in precipitation for the different stations in 2000 and 2001.

Atrazine (Figure 6) is a herbicide of which the authorisation was withdrawn in 1999 due to the persistence and leaching potential of the substance. The substance is still frequently found in precipitation samples, in the period April - September. The highest concentrations were in 2001 again found in the south of the country: Wynandsrade, Yerseke and the Groote Peel. The substance therefore seems to be originating from Belgium and France, where the product is not forbidden. Belgium has a lot of sandy soils on which maize is grown, in which crop *atrazine* is used a lot. The substance was, as in the year 2000, hardly found in air.

Application in autumn
Trifluralin (Figure 7) is applied as herbicide in winter cereals. It is no longer authorised in the Netherlands since 1999. *Trifluralin* is fairly persistent in the soil. The concentration pattern for *trifluralin* is in agreement with the use of the substance from September to December: high concentrations are found in winter. There is, however, also a peak in use from April - June 2001, where a gradient is visible from the south to the north of the Netherlands (high in Yerseke, Wynandsrade, Groote Peel, Egmond, and De Zilk, low in the northern provinces, despite the fact that a lot of winter cereals are grown in this area). The presence of *trifluralin* in precipitation samples is probably caused by emissions in surrounding countries, transported to the Netherlands via the air.

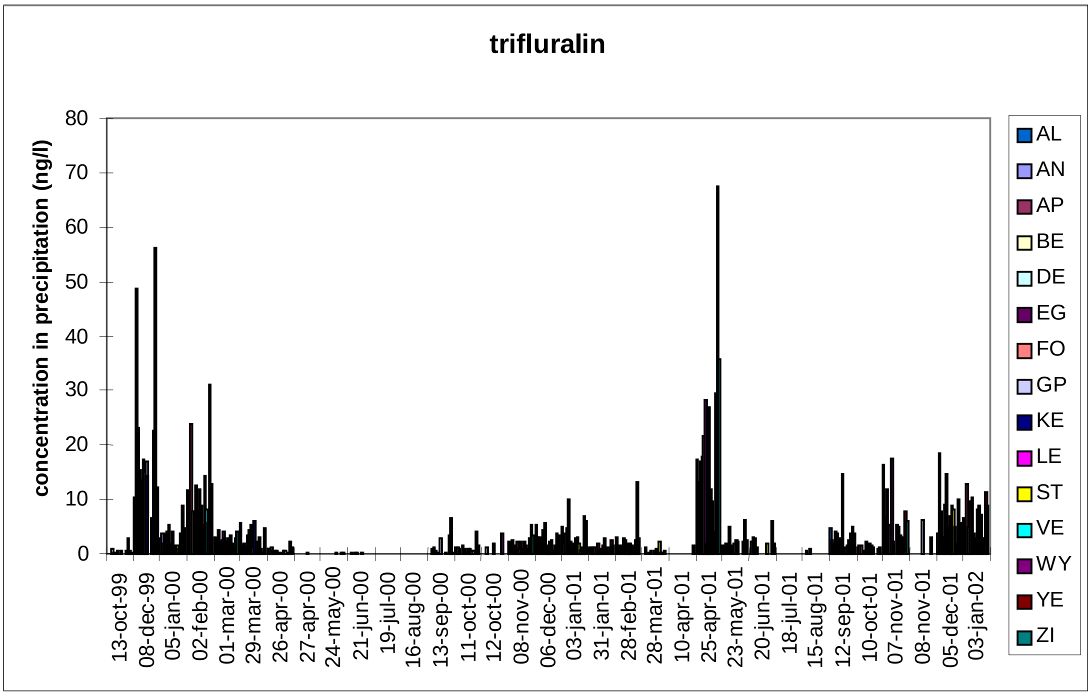


Figure 7 *Concentration course of trifluralin in precipitation for all stations from October 1999 to January 2001.*

Substances that are applied throughout the year

DNOC is a substance of which the authorisation was withdrawn in the course of the year 2000. The substance was applied as haulm desiccant and as herbicide, throughout the year.

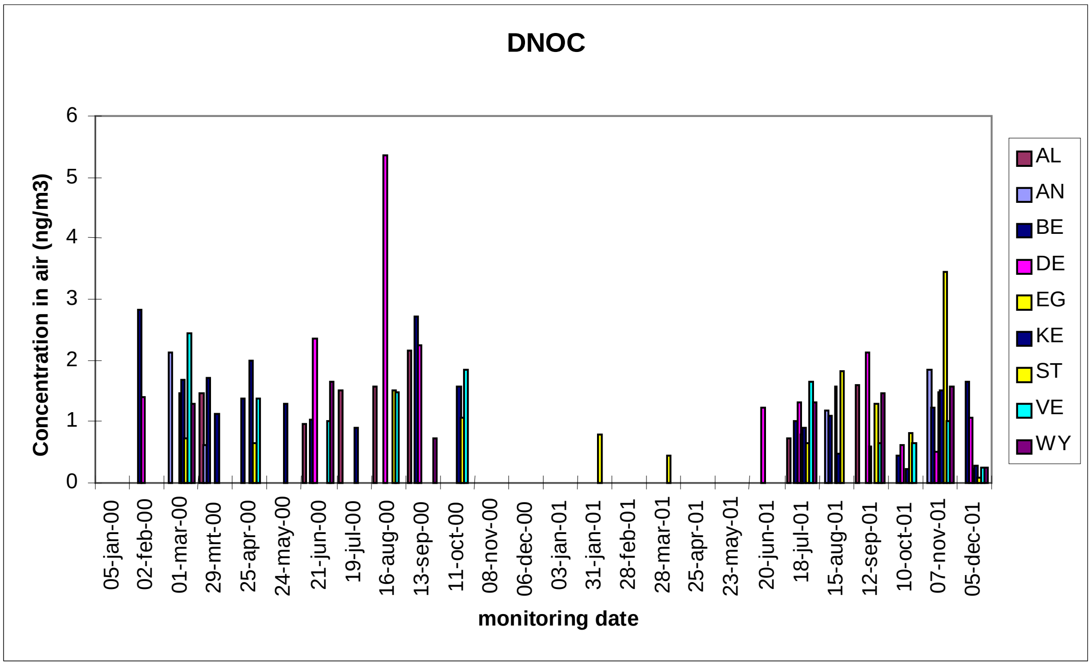


Figure 8 *Course of the DNOC concentration in air for the different stations in 2000 and 2001.*

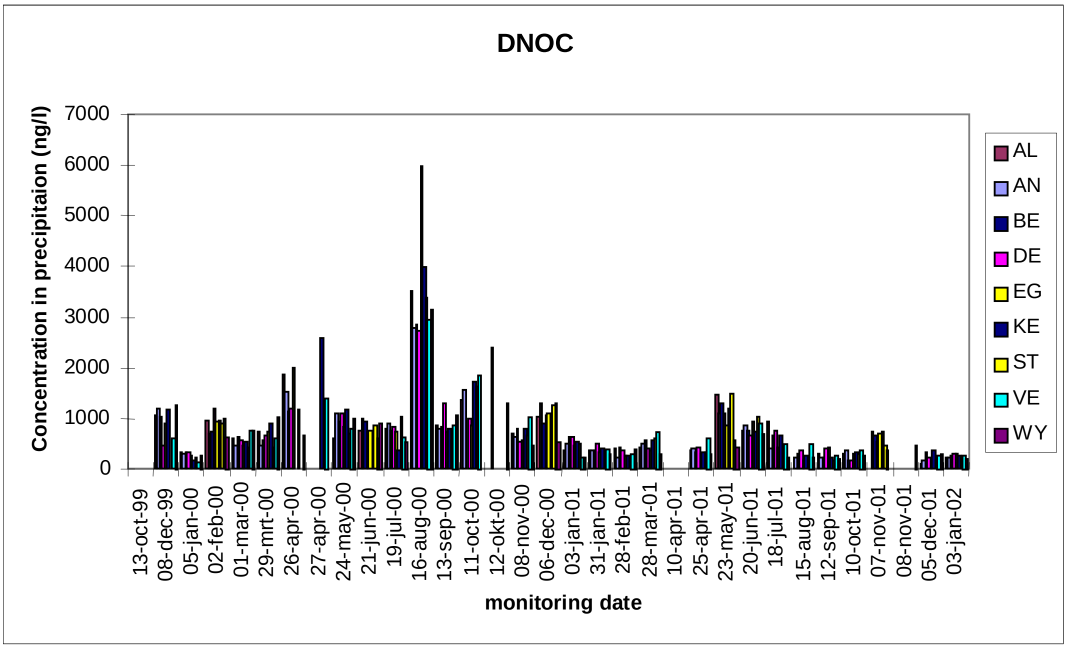


Figure 9 *Course of the DNOC concentration in precipitation for the different stations in 2000 and 2001.*

DNOC is forbidden due to its sensitivity for leaching to groundwater. Figure 9 shows a decreasing course in the concentrations in precipitation. This course does not show up in the concentrations in air samples (Figure 8). *DNOC* was still regularly found at all stations. Averaged over the year and over all stations, the concentrations in precipitation have about halved in the period 2000 to 2001. The concentration level, however, still exceeds the drinking water criterion of 100 ng/l.

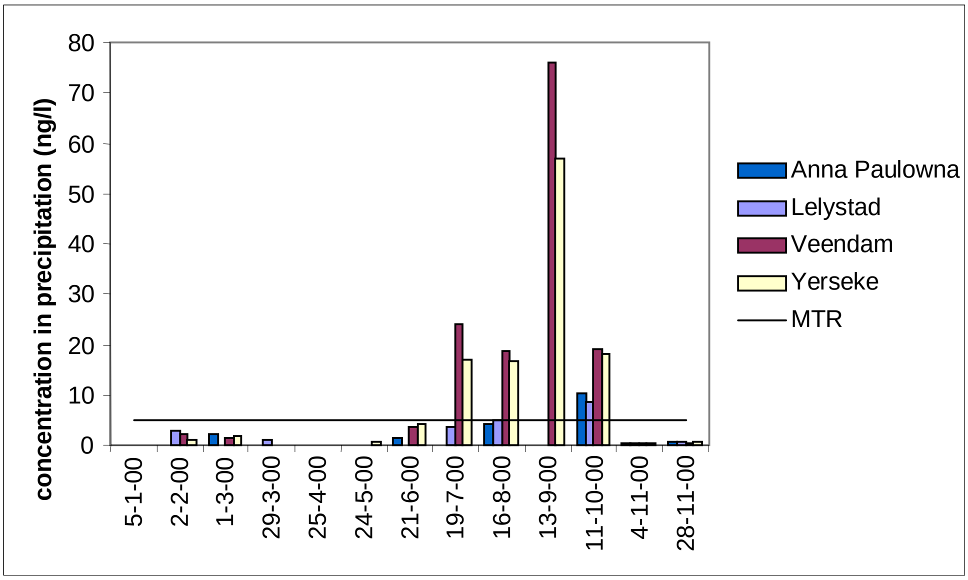


Figure 10 *Course of the triphenyltin concentration in precipitation for the four stations in 2000, and the MTR for surface water (ng/l).*

Of the tin compounds, *triphenyltin* was regularly found in precipitation in the year 2000 (Figure 10). The MTR for surface water was exceeded at all locations. *Triphenyltin* was used as fungicide in potato growing. The highest values were found in Yerseke and Veendam, where this crop is important.

Besides substances covered by regulations, there are still other interesting substances that merit discussion here. *Fluazinam* is a substance that becomes increasingly popular as result of the disappearance of other products. *Fluazinam* (Figure 11) is moderately degradable to persistent in the soil but it is degradable in water. It is a fungicide that is used for the treatment of planting stock (dipping), in potato growing and in the field in bulb growing. This substance was in the year 2000 already found in precipitation at levels above the drinking water criterion; the number of samples that exceeded the drinking water criterion had in 2001 increased to over 4%. It was found in high concentrations in air and precipitation samples at the monitoring locations Veendam, Ketelhaven, Anna Paulowna, Yerseke and Wynandsrade.

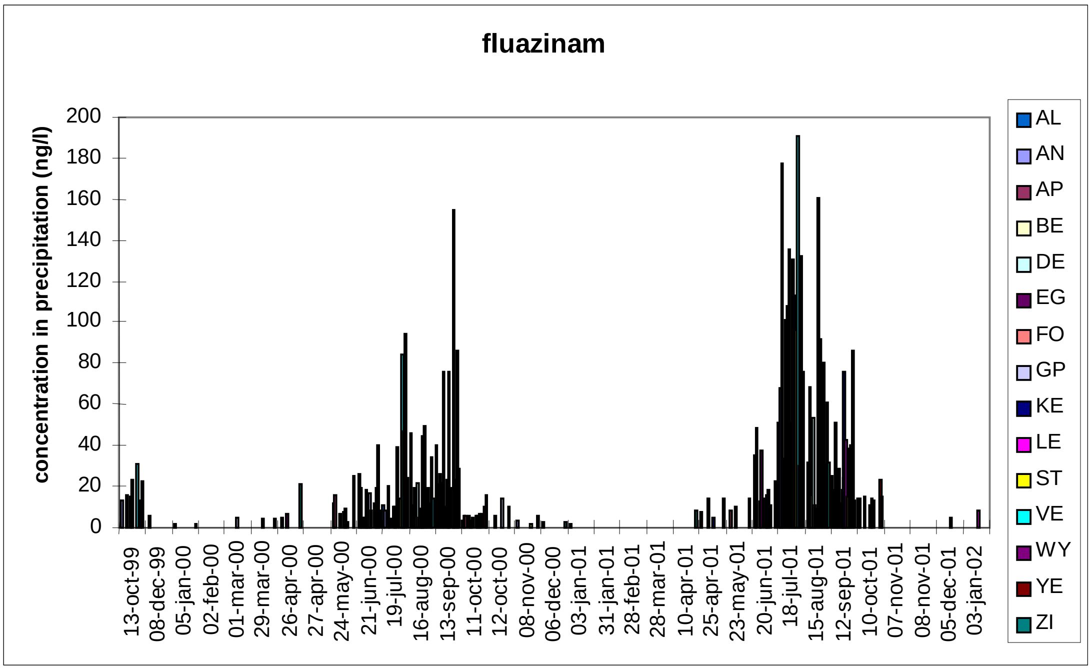


Figure 11 Concentration course of fluazinam in precipitation for the different stations in the years 2000 and 2001.

The concentration course in precipitation for the substances *ethofumesate* and *terbutylazine* is similar to that of *fluazinam*.

Ethofumesate is a herbicide that is used against annual weeds in field vegetable production (beet) and in grassland. The substance was in the year 2000 determined at four locations and in 2001 at all 18 locations. In comparison with the year 2000, the concentration at the four locations had increased in 2001. *Ethofumesate* was in

in Yerseke, Wynandsrade and Ketelhaven found in precipitation in concentrations exceeding the drinking water criterion.
Ethofumesate is in sugar and fodder beet applied from March to May, in ryegrass in spring or autumn, and on grassland in autumn. The measurements in the year 2000 only show peaks from April to June. In November 2001, a maximum concentration well above the drinking water criterion was found at monitoring station Aalsmeer.

Terbutylazine is used as herbicide in the culture of potatoes and peas. The substance is persistent in the soil, and very persistent in natural water. *Terbutylazine* is included in the re-evaluation/withdrawal list of the MJP-G. At least one metabolite can leach to the groundwater. The concentration in precipitation has increased in 2001. The number of times that the criterion for drinking water is exceeded has also increased.

Polar pesticides
In spring 2000, some precipitation samples were submitted to Omegan laboratories for analysis of the concentration of polar pesticides, where the limited amount of sample sometimes was a problem. Only *isoproturon* was found regularly. The *isoproturon* concentration did in Wijnandsrade a few times exceed the criterion for drinking water. *Diuron*, *pencycuron*, *metoxuron* and *metabromuron* were also found a few times. *Carbendazim* could not be detected. The analytical results are presented in Annex VIII.

3.2.4 Average concentrations in air

About 50 different pesticides, 18 PAHs and seven PCBs could be detected in air. The annual average concentrations, percentages in which the substances were found, and the average concentrations of the samples in which the substances were found in air in the years 2000 and 2001, are presented in Annex II.
Aldrin, 2.4-D, diazinon, deltamethrin, dicofol, disulfoton, fenitrothion, fenthion, phosphamidon, heptenophos, metamitron, methomyl, parathion-methyl, and *triadimenol* could not be detected in air.
In the year 2001, *bentazon*, *captan*, *ethofumesate* and *pentachlorobenzene* were more frequently found in air than in the year 2000. The substances *hexachlorobenzene*, *lindane* and *procymidon* were considerably less found in air in 2001 than in 2000.

Figure 12 shows the average concentrations of pesticides that were regularly found in air samples.

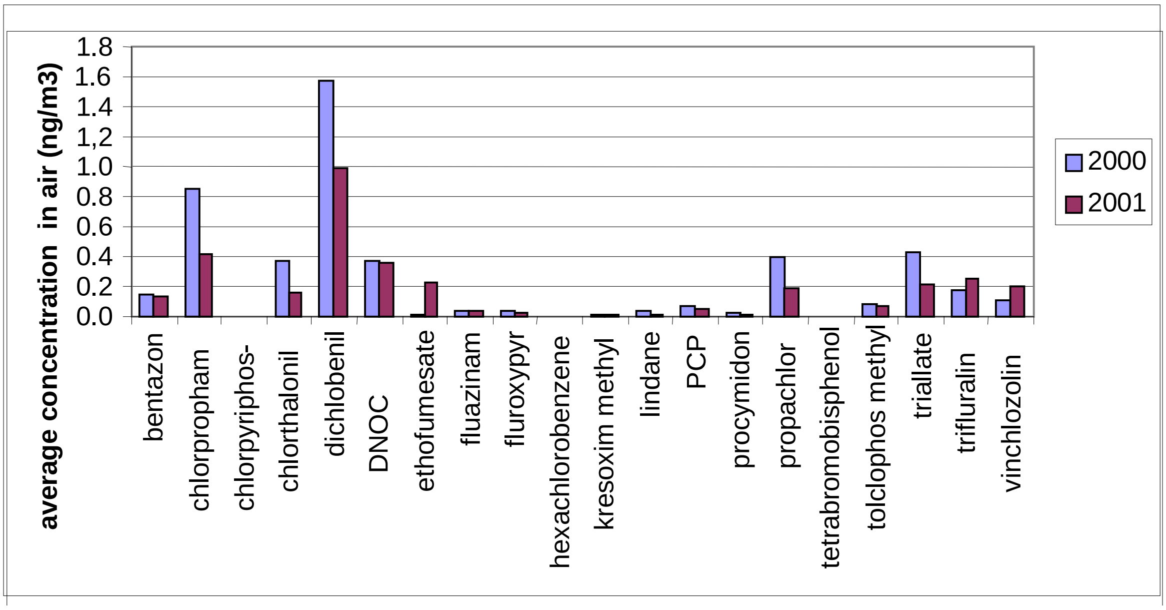


Figure 12 Annual average concentration of pesticides in air samples of substances that were regularly found.

The figure does for most herbicides show a decrease in concentration in 2001, where *chlorpropham*, *dichlobenil* and *propachlor* show the largest decrease. The concentration of the herbicide *ethofumesate* has strongly increased in 2001. The concentrations of the fungicides *chlorothalonil* and *procymidon* have decreased in 2001; the concentration of *vinchlozolin* has increased in 2001; *chlorothalonil* and *vinclozolin* were in 2001 determined at all eighteen locations so that the average concentrations between both years cannot be compared directly. It is also noteworthy that only one insecticide, *lindane*, is regularly found in air. The explanation for this phenomenon is probably that insecticides are applied in smaller amounts than herbicides and fungicides.

The percentage samples in which the PAHs were detected in air is roughly the same in both years. Only *benzo[a]-anthracene*, *chrysene* and *dibenz[a,h]-anthracene* were found less frequently in 2001. The PAH concentrations in air in both years are comparable (Figure 13).

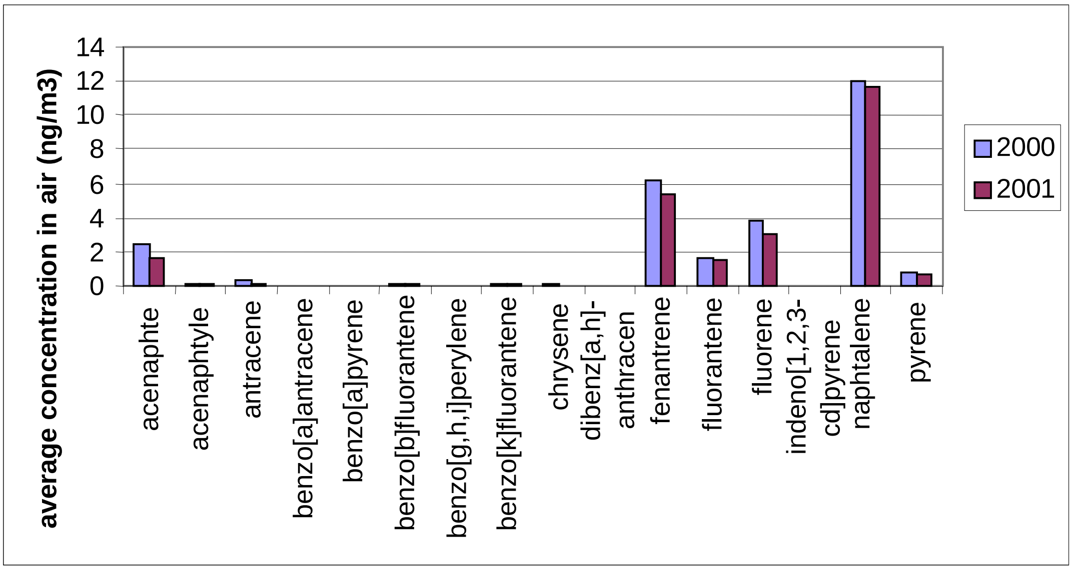


Figure 13 Annual average concentration of PAHs in air samples in the years 2000 and 2001.

The percentage PCBs found in air samples in 2001 has decreased in comparison with the year 2000 (Figure 14), in particular the PCBs 138, 153, 28 and 8. PCB-180 is no longer detected. These differences are probably also caused by a necessary change in the analytical procedure, which resulted in a change in the limit of detection. The other concentrations in the air samples in 2001 are comparable to the concentrations in the year 2000.

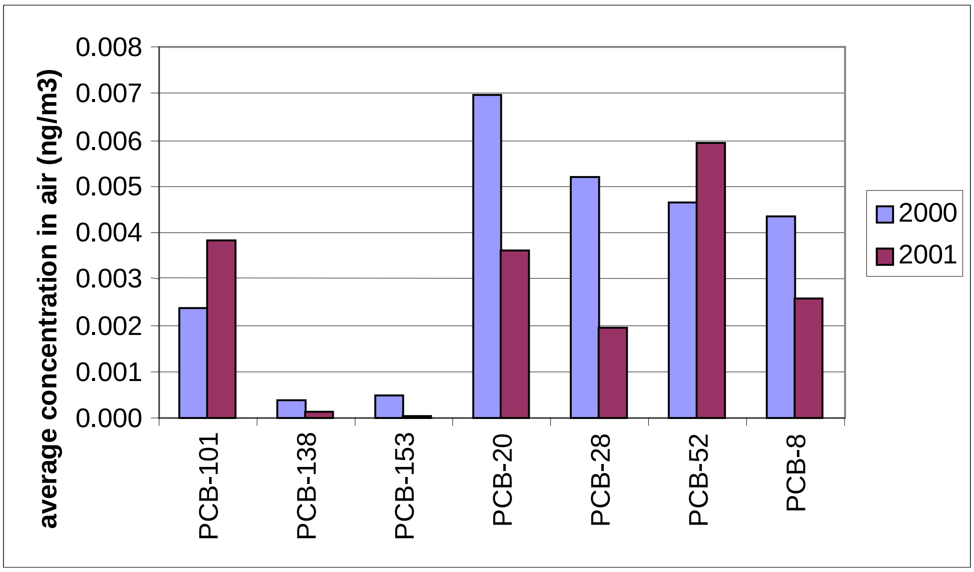


Figure 14 Annual average PCB concentrations in air samples in the years 2000 and 2001 .

The variation between both years was larger for the pesticides concentrations than for the PCB and PAH concentrations. This is probably caused by differences in use, and thus emission, of the pesticides in both years, whereas the emission of PAHs and PCBs has not changed much.

3.2.5 Average concentrations in precipitation

Pesticides

The pesticides concentrations in precipitation are presented in Annex II. About 50 pesticides could be detected in precipitation. *Aldrin, bitertanol, deltamethrin, dicofol, disulfoton, endrin, phosphamidon, heptachlor, heptachlor-epoxide, heptenophos, methomyl o,p-DDE, telodrin, triadimenol* and *triazophos* could not be detected in precipitation.

Figure 15 shows the annual average concentrations of those pesticides that were found in more than ten per cent of the precipitation samples.

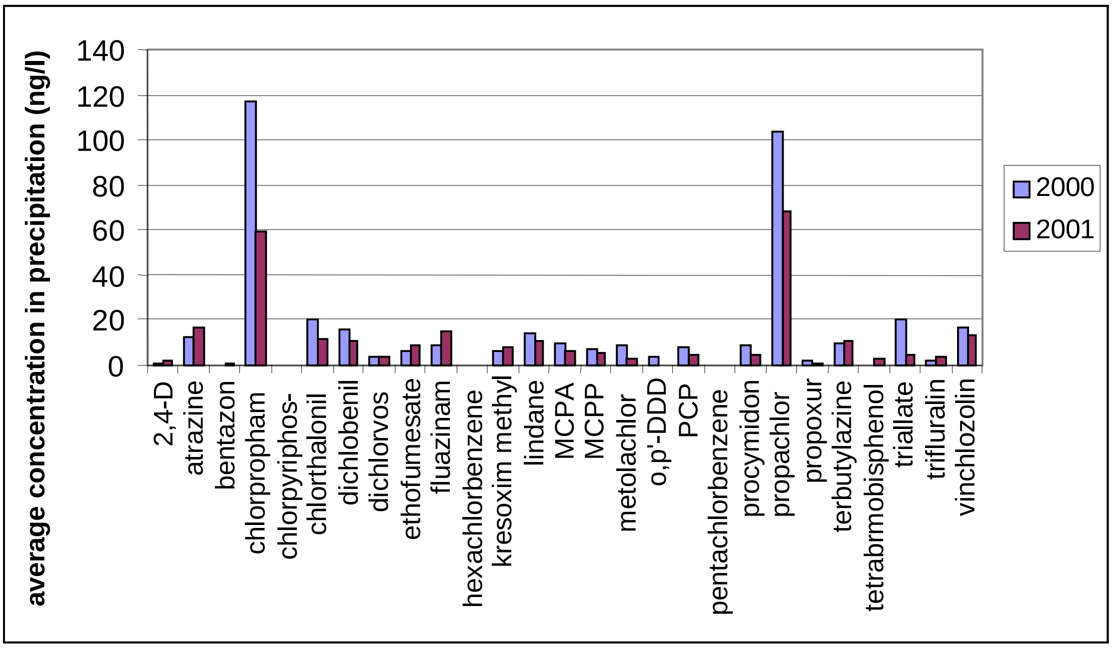


Figure 15 Annual average pesticides concentrations in precipitation, over all monitoring stations.

The large number of herbicides and fungicides in this figure is again noteworthy. The concentrations of most herbicides and fungicides have in 2001 decreased by a factor two, e.g., *DNOC, chlorpropham* and *propachlor*. Exceptions are *ethofumesate* (H), *terbutylazine* (H) and *fluazinam* (F), of which the concentration increased in 2001. The concentration of herbicides that are no longer authorised in the Netherlands, *atrazine* and *trifluralin*, did also increase in 2001.

Comparison with criterion concentrations

For most substances in the study there is no quality criterion for concentrations in air or precipitation. A quality criterion has only been derived for *benzo[a]pyrene* for open air. This value of 1 ng/m^3 was not exceeded. In order to gain some insight into the height of the found concentration, the pesticides concentrations in precipitation have been compared with the Maximum Tolerable Risk level (MTR) for surface water (Table 4). Formally, this value has no significance for precipitation but it gives a first impression of the possible toxicity of the observed concentration. In aquatic systems that are only fed by precipitation, the concentration could rise to the level of the concentration in precipitation. Loss processes in surface water and dilution will usually be the cause that the concentration in surface water never reaches that level. On the other hand, the input via precipitation will not easily lead to MTR exceedance in case the concentration in the precipitation is *below* the MTR level.

The criterion for drinking water production is another water quality criterion. This neither has a formal significance for precipitation but it does again give an indication. The pesticides that exceeded the drinking water criterion are listed in Table 5. Also substances that are no longer authorised, e.g. due to high persistence or toxicity, can still be found in precipitation (and air) (Table 6).

The concentration in precipitation of eighteen different pesticides did one or more times exceed the MTR level for surface water. These were seven insecticides, four herbicides, four fungicides and two substances that are used as insecticide/acaricide (Table 4). *Methiocarb,* a substance with a broad field of use, was also found above the MTR. *Triphenyltin,* a substance that was only determined at four locations, did also regularly exceed the MTR level. *Dichlorvos* was in 2000 as well as in 2001 only found above the MTR. The number of samples in which the substance was found had increased in 2001. The following substances did only in 2001 exceed the MTR: *methiocarb, pirimiphos-methyl, terbutylazine* and *trifluralin*. *Pirimiphos-methyl* was only determined in 2001.

Table 4 MTR-exceeding substances in precipitation, with the percentage month samples that exceed the MTR, the maximum MTR exceedance and the use.

Substance	**Use**	**MTR** ng/l	**% > MTR** 2000	**% > MTR** 2001	**Maximum MTR exceedance (x MTR) 2000**	**Maximum MTR exceedance (x MTR) 2001**
captan	F	110	0	1	0	9
chlorfenvinfos	I	2	1	1	11	18
chlorpyriphos-methyl	I	3	2	0	2	0
chlorothalonil *	F	10	51	24	32	26
diazinon	I	37	0.4	0	1	0
dichlorvos	I	0.7	21	29	88	69
endosulfan I	I/A	20	0.4	0.4	1	2
parathion-ethyl	I	2	1	0	12	0
methiocarb	Broad	16	-	7	-	6
metolachlor	H	200	0.4	0	1	0
mevinphos	I	2	2	3	7	5
propachlor	H	1300	1	0	3	0
pirimiphos-methyl	I/A	2	-	4	-	498
propoxur	I	10	3	2	7	2
pyrazophos	F	40	0.4	0	8	0
terbutylazine*	H	190	0	0.4	0	2
trifluralin	H	38	0	0.4	0	2

* number of locations in 2001 increased from 4 to 18

I: insecticide A: acaricide H: herbicide F: fungicide

In the years 2000 and 2001 twenty two pesticides exceeded the drinking water criterion, of which eight fungicides, ten herbicides, two insecticides and two substances that are used as insecticide and acaricide (Figure 16, Table 5). Of the fungicides, *procymidon*, which was in 2000 still notable due to the height of the exceedance, was in 2001 still only found in precipitation at a level below the drinking water criterion. Seven herbicides exceeded the drinking water criterion; *MCPA* and *metolachlor* did in 2001 no longer exceed the drinking water criterion. The exceedance of *chlorpropham*, notable in the year 2000, is now considerably reduced in number of times that the substance was found and concentration level. Of the insecticides, *dimethoate* no longer exceeded the criterion, whereas *pirimiphos-methyl* did in 2001 exceed the criterion. Substances for broad use, such as *DNOC* and *methiocarb*, did also exceed the criterion in 2001, where the concentration level of the forbidden substance *DNOC* was halved. *DNOC* did also in 2001 exceed the drinking water criterion in almost all precipitation samples!

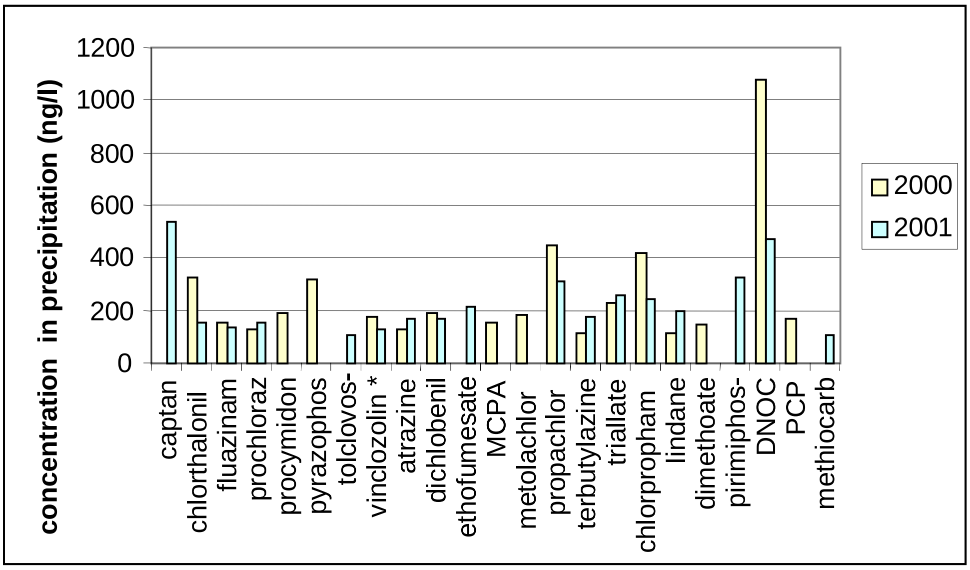


Figure 16 *Average concentrations of pesticides that exceeded the drinking water criterion of 100 ng/l. The average was calculated from the samples that exceeded the criterion.*

Table 5 Substances that exceeded the drinking water criterion in precipitation (100 ng/l), with the percentage of samples that exceeded the criterion, and the average concentration in those samples.

Substance	% samples > drinking water criterion 2000	% samples > drinking water criterion 2001	Average > drinking water criterion (ng/l) 2000	Average > drinking water criterion (ng/l) 2001
Fungicides				
captan	0	1.3	0	537
chlorothalonil *	2.6	1.8	325	149
fluazinam	0.4	4.4	155	136
prochloraz	0.4	0.4	126	152
procymidon	0.9	0	193	0
pyrazophos	0.4	0	320	0
tolclofos-methyl	0	0.4	0	105
vinchlozolin *	2.6	1.3	174	131
Herbicides				
atrazine	2.6	4.8	130	166
dichlobenil	1.3	0.4	191	167
ethofumesate	0	1.8	0	215
MCPA	0.4	0	155	0
metolachlor	1.8	0	183	0
propachlor	21.1	19.3	446	312
terbutylazine *	5.3	2.6	115	173
triallate	2.2	0.4	226	260
chlorpropham	22.8	14.5	419	245
Insecticides				
lindane	3.9	1.3	117	196
dimethoate	1.8	-	144	-
pirimiphos-methyl	-	3.1	-	328
Other				
DNOC	99.6	98.2	1075	471
PCP	0.4	0	165	0
methiocarb	0	0.9	0	103

* number of locations in 2001 increased from 4 to 18

\- not determined

Atrazine, DNOC, propachlor, trifluralin and *mevinphos* were, despite the withdrawal of the authorisation in the Netherlands in 2000, found in precipitation. Table 6 shows the concentration and the frequency of occurrence of these substances.

Table 6 Substances that are no longer authorised in 2001. The table shows the use of the substance, the percentage of the precipitation samples that exceeds the limit of detection (DL) and the average concentration in the year 2000 and 2001.

Substance in precipitation	**Authorisation withdrawn in the year**	**Use**	**% > DL 2000**	**% > DL 2001**	**Concentration 2000 (ng/l)**	**Concentration 2001 (ng/l)**
atrazine	1999	H	21	24	12	16
DNOC	2000	B	98	98	1061	463
endosulfan II		I/A	0	0	1.5	0.05
propachlor	2000	H	41	35	104	68
trifluralin	1999	H	27	44	2.2	3.7
mevinphos	2000	I	0	1	0.2	0.1

I: insecticide A: acaricide H: herbicide F: fungicide

The *atrazine* (Figure 6) and *trifluralin* (Figure 7) concentrations did even slightly increase in 2001. The presence of both herbicides in air and precipitation in the Netherlands suggests a contribution from surrounding countries.

The authorisation of a number of pesticides, which had a temporary authorisation in view of their agricultural indispensability, was withdrawn in the course of 2001. These are the substances *carbaryl, carbofuran, chlorpyriphos, chloridazon, dichlorvos, fenbutatinoxide, parathion-ethyl, penconazole, pirimiphos-methyl* and *simazine*.

Table 7 Substances no longer authorised in the course of 2001. The table shows the use of the substance, the percentage of the precipitation samples that exceeds the limit of detection, and the average concentrations in the years 2000 and 2001.

Substance in precipitation	**Authorisation withdrawn in the year**	**Use**	**% > DL 2000**	**% > DL 2001**	**Concentration 2000 (ng/l)**	**Concentration 2001 (ng/l)**
chlorothalonil [1]	2001	F	54	36	20	11
chlorpyriphos[2]	2001	I	2	0	0.4	0.2
diazinon	2001	I	0.4	0	1	0.3
dichlorvos [2, 3]	2001	I	0.4	10	4	3
lindane[4]	2001	I	14	6	14	11
mecoprop	2001	H	2	5	7	6
pirimiphos-methyl [2]	2001	I/A	-	4	-	10.3
pyrazophos	2001	F	0.4	-	2	0
simazine [2]	2001	H	1	1	1	1
triallate	2001	H	68	10	21	4.4

- not determined

I: insecticide A: acaricide H: herbicide F: fungicide

[1] alternately forbidden/not forbidden.

[2] substances that had the status ‘agriculturally indispensable’

[3] authorised for veterinary use as insecticide

[4] substance since 1990 only authorised for seed treatment.

PAHs

The PAH concentrations in 2001 were slightly lower than in the year 2000. Concentrations of *benzo[a]pyrene*, *benzo[b]fluorantene*, *chrysene* and *pyrene* had decreased considerably. The substances *acenaphtylene* and *dibenz[a,h]-anthracene* were in 2001found more frequently in precipitation (Annex II). Figure 17 shows the annual average concentrations of PAHs in precipitation, for 2000 and 2001. The concentration of five PAHs did a few times exceed the MTR for surface water. Markedly high concentrations were found for *fenantrene, fluorantene* and *pyrene*. In the year 2000, the sum of the concentrations of the Borneff PAHs in precipitation was on average higher than the criterion of 200 ng/l voor drinking water (210 ng/l). In 2001, the concentration was about 25% lower than this value. *Benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, indeno[g,h,i-cd]pyrene* and *fluorantene* did in 2000 separately exceed this value in precipitation samples. In 2001, only *fluorantene* did separately exceed the criterion.

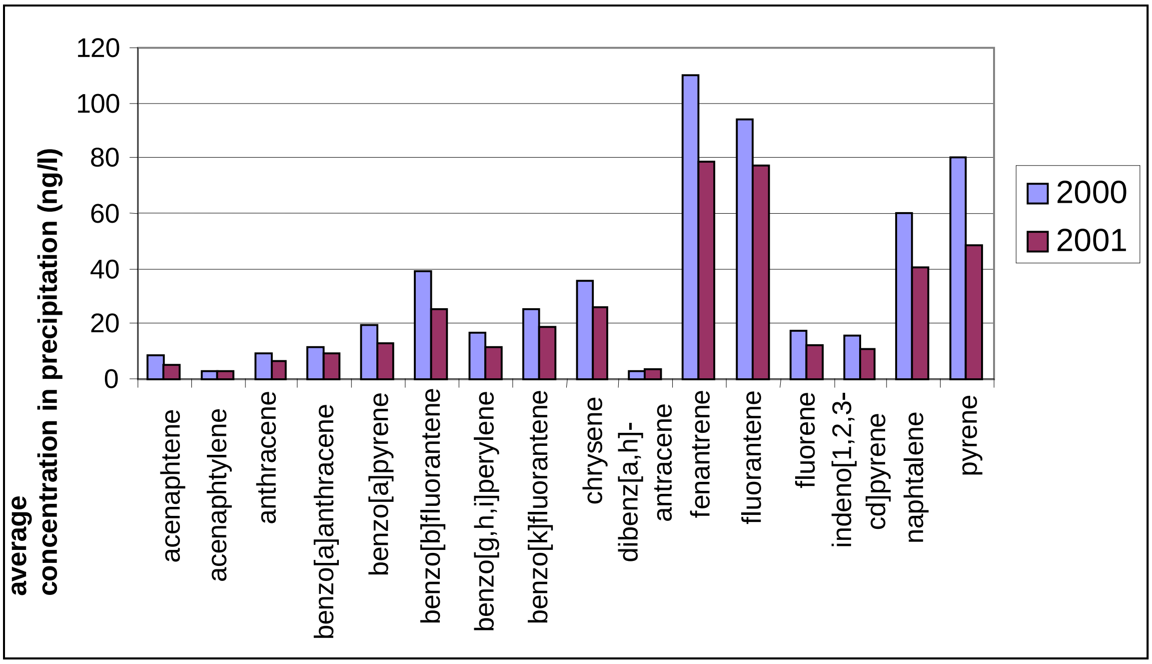


Figure 17 Annual average concentrations of PAHs in precipitation (ng/l) in 2000 and 2001.

PCBs

Het percentage PCBs found in precipitation as well as in air samples had in 2001 decreased in comparison with the year 2000. This is in particular true for the PCBs 101, 118, 138 and 180. The average concentrations over all samples had also decreased.

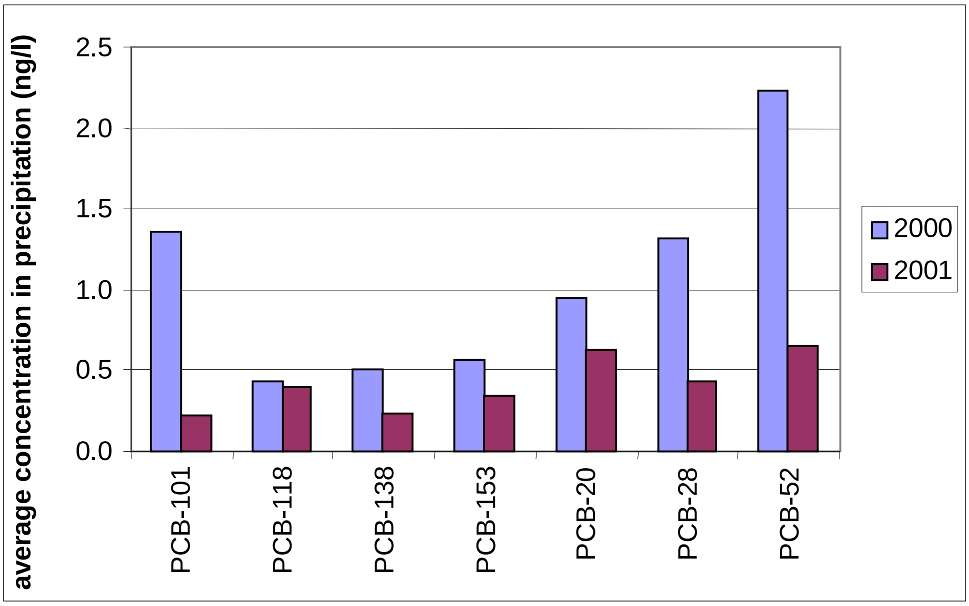


Figure 18 Annual average concentrations of PCBs in precipitation (ng/l) in 2000 and 2001.

3.3 Input into soil and surface water by atmospheric deposition

An estimate of the dry deposition rate to soil and surface water has in this report been made for those substances that are regularly found in precipitation and air samples, according to the method presented earlier in this report. An estimate has been made of the total deposition on the surface water of the Netherlands and on the soil of the Netherlands. For IJsselmeer, Noord-Holland and Waddenzee, the atmospheric load has been calculated on the basis of measurements at the monitoring stations that are considered representative. Deposition on the IJsselmeer has been calculated on the basis of the data of the surrounding stations Stavoren, Ketelhaven, Andijk and Lelystad. Deposition on the province Noord-Holland has been calculated on the basis of all stations in Noord Holland: Anna Paulowna, Andijk, Egmond and Aalsmeer. Deposition on the North Sea has been calculated by means of measurements of the monitoring locations Noordwijk and Lauwersmeer. This deposition is probably an overestimation of the actual deposition on the North Sea because concentrations of the pesticides that are used in the Netherlands will decrease with the distance from the coast; this has not been taken into account. The average atmospheric deposition of the rest of the surface water and soil of the Netherlands has been calculated from the average wet deposition and dry deposition for all monitoring stations.

Table 8 *Total atmospheric deposition of pesticides on the surface water of the Netherlands (2 793 km^2), the surface water in Noord-Holland (800 km^2), the North Sea (508 126 km^2 in* ***t/yr****), the Waddenzee (2 600 km^2), the IJsselmeer (1 928 km^2)$^{1)}$ and soil of the Netherlands (36 783 km^2) in (kg/yr). The reliability for the substances printed* ***bold*** *is good. The reliability for the other substances is low (see also text). The Italic "0" means that the substance has not been detected. The estimates are based upon measurements of wet deposition, air concentration and an estimate of the dry deposition velocity*

Atmospheric deposition	**Surface water NL 2000 kg/yr**	**Surface water NL 2001 kg/yr**	**Surface water Noord-Holland 2001 kg/yr**	**North Sea$^{1)}$ 2001 t/yr**	**Wadden-zee 2000 kg/yr**	**Wadden-zee 2001 kg/yr**	**IJssel-meer$^{2)}$ 2000 kg/yr**	**IJssel-meer$^{2)}$ 2001 kg/yr**	**Soil 2000 kg/yr**	**Soil 2001 kg/yr**
2,4-D	1	4	1,2	1	0,8	4,8	0,9	2,3	15	49
aldrin	*0*	-	-	-	*0*	-	-	-	*0*	-
atrazine	24	25	6	3	15	17	14	13	313	333
bentazon	2	7	1	0,04	*0*	1	1	6	19	65
bitertanol	39	-	-	-	*0*	-	-	-	*0*	-
captan	17	85	29	4	12	30	15	53	104	432
chlorfenvinfos	1	1	1	*0*	*0*	1	1	*0*	5	4
chlorpropham	557	290	142	15	1333	382	362	218	3765	2100
chlorpyriphos-methyl	1	1	0,2	0,02	3	1	1	1	10	5
chlorothalonil	80	38,2	15	3	84	61	-	30	280	298
chloridazon (pyrazon)	2	-	-	-	1	-	1	-	8	-
deltamethrin	*0*	-	-	-	*0*	-	-	-	*0*	-
demotion-S-methyl	6	-	-	-	20	-	-	-	2	-
diazinon	1	1	1	0,1	2	2	*0*	1	14	11
dichlobenil	685	443,7	140	42	809	352	413	356	499	362
dichlorvos	18	19,3	4	0,2	4	5	3	8	129	132
dicofol	*0*	-	-	-	*0*	-	-	-	*0*	-
dimethoate	6	3	0,2	0,1	0,1	0,4	6	1	81	25
disulfoton	*0*	-	-	-	*0*	-	-	-	*0*	-
DNOC	2391	1207	324	181	2201	1207	1709	879	29248	14057
endosulfan I	1	2	*0*	*0*	*0*	*0*	0,4	*0*	3	10
endosulfan II	4	0,8	0,1	*0*	2	1	1	*0*	38	2
endrin	2	-	-	-	8	-	-	-	*0*	-
epoxiconazole	6	-	-	-	*0*	-	-	-	39	-
ethofumesate	11	34	7	5	10	23	-	19	104	267
parathion-ethyl	0,3	0,2	*0*	0,2	0,1	*0*	0,4	*0*	3	0,05
fenitrothion	0,1	-	-	-	0,3	-	*0*	-	1	-
fenthion	0,1	-	-	-	*0*	-	-	-	1	-
fluazinam	31	53	11	4	14	48	16	56	261	556
fluroxypyr	20	9	4	*0*	26	10	-	3	80	13
phosphamidon	*0*	-	-	-	*0*	-	*0*	-	*0*	-
heptachlor	*0*	0,7	*0*	*0*	*0*	*0*	*0*	*0*	*0*	9
heptachlor epoxide	2	-	-	-	1	-	-	-	*0*	-
heptenophos	*0*	-	-	-	*0*	-	-	-	*0*	-
hexachloor-1,3-butadiene	1	-	-	-	3	-	-	-	0,1	-
hexachlorobenzene	2	2	1	0,1	3	2	2	1	11	8
isoxaflutole	5	-	-	-	*0*	-	-	-	7	-
kresoxim-methyl	12	16	4	2	24	19	-	10	81	172
lindane	39	23	8	2	30	15	26	11	313	248
malathion	0,1	-	-	-	*0*	-	*0*	-	1	-
MCPA	16	13	5	3	15	16	9	6	206	165
mecoprop	12	12	4	1	7	13	7	6	155	153
metamitron	1	-	-	-	2	-	-	-	9	-
methiocarb	0,1	7	3	0,2	*0*	11	*0*	3	0,5	83
methomyl	*0*	-	-	-	*0*	-	-	-	*0*	-
parathion-methyl	0,1	-	-	-	*0*	-	*0*	-	1	-
metolachlor	24	6	2	1	10	3	8	1	256	72
mevinphos	2	3	1	0,4	3	4	*0*	2	5	31

Atmospheric deposition	**Surface water NL**	**Surface water NL**	**Surface water Noord-Holland**	**North Sea**[1)]	**Wadden-zee**	**Wadden-zee**	**IJssel-meer**[2)]	**IJssel-meer**[2)]	**Soil**	**Soil**
	2000	**2001**	**2001**	**2001**	**2000**	**2001**	**2000**	**2001**	**2000**	**2001**
	kg/yr	**kg/yr**	**kg/yr**	**t/yr**	**kg/yr**	**kg/yr**	**kg/yr**	**kg/yr**	**kg/yr**	**kg/yr**
o,p'-DDD	7	1,1	0,02	0,5	5	1	-	1	48	14
o,p'-DDE	1	-	-	-	2	-	-	-	*0*	-
pentachlorbenzene	0,5	0,6	0,3	0,1	2	1	-	1	3	6
pirimicarb	0,5	1,1	0,3	*0*	0,3	0,1	0,5	1	5	13
pirimiphos-methyl	4	352	88	50	11	312	6	397	3	456
prochloraz	2	3	2	0,9	*0*	*0*	1	4	27	35
procymidon	24	13	6	0,5	30	14	15	6	170	109
propachlor	312	193	50	15	314	188	442	279	2205	1685
propoxur	2	1	0,5	0,2	1	1	3	2	29	19
pyrazophos	5	0,3	*0*	0,07	1	0	3	0	28	0,2
simazine	4	3,5	3	*0*	2	10	3	2	27	22
telodrin	0,4	-	-	-	1	-	-	-	0	-
terbutylazine	15	29	6	3	6	16	-	12	148	264
tetrabromobisphenol A	0,03	8	3	0,3	0,02	2	-	5	0,2	92
tolclofos-methyl	15	12	13	0,1	49	43	2	11	101	74
triadimenol	*0*	-	14	-	*0*	-	-	-	*0*	-
triallate	201	80	-	10	174	69	171	47	725	115
triazophos	1	-	-	-	*0*	-	-	-	*0*	-
trifluralin	59	85	32	11	50	88	44	54	74	124
vinclozolin	78	106	31	3	185	117	-	126	446	687

1) The estimates of the deposition on the North Sea are uncertain (see text).

2) The IJsselmeer is defined as the public waters in the IJsselmeergebied.

The reliability for the substances printed **bold** is reasonable (see also Annex III). The other substances were found so little in exceedance of the limit of detection that the reliability of the estimated deposition is low. This applies for *aldrin, bitertanol, demeton-S-methyl, diazinon, endrin, fenthion, heptachlorepoxide, heptenophos, hexachlorbutadiene, malathion, o,p'-DDE, p,p-DDT, prochloraz, telodrin, triademol, triazophos*. The calculations that have been carried out to investigate the effect of the limit of detection are elaborated in Annex III. In case the uncertainty in the estimated deposition as result of the limit of detection is larger than a factor two and the substance was found in a low frequency (< 5%) in precipitation and air, the reliability is considered low.

The ratio between the contribution of wet and dry deposition is also discussed in Annex III.

High values for the deposition to the surface water in the Netherlands were in 2000 found for *DNOC* (2.4 t per year for the Netherlands), *dichlobenil* (0.7 t) and *chlorpropham* (0.6 t per year), *propachlor* (0.3 t per year) and *triallate* (0.2 t per year). A high deposition was also found for *chlorothalonil* (almost 0.1 t per year) and *vinclozolin* (0.08 t per year).

The average deposition of a number pesticides is the Netherlands in 2001 had strongly decreased in comparison with 2000: the deposition of *metolachlor* in 2001 is one fifth of the level in 2000, the deposition of *triallate* and *dimethoate* in 2001 is almost one third of the level in 2000; deposition of *chlorpropham, DNOC, lin-*

dane and *procymidon* has almost halved, deposition of *propachlor* and *dichlobenil* have decreased by a quarter to a third. The deposition of *2,4-D*, *bentazon, fluazinam*, *methiocarb* and *mevinphos* has in 2001 increased by a factor 2 to 4. Of the substances that were in 2001 measured at 18 locations instead of four, the deposition of *ethofumesate, kresoxim-methyl, terbutylazine, tetrabromobisphenol* and *vinclozolin* also increased by a factor 2 to 4.
Annex VI gives further information about the measurements in the province Noord-Holland, where three extra monitoring locations were set up upon request of the province. The nationally highest deposition of a number pesticides is found at the monitoring locations Anna Paulowna and Andijk (see Annex III). This is associated with the specific location of the monitoring stations in relation to source areas. The mentioned monitoring stations were found to be relatively strongly burdened with substances that are used in bulb growing and potato growing such as *chlorpropham, tolclofosmethyl, pyrazophos, procymidon, chlorpyriphos methyl* and *chlorothalonil.*

Table 9 *Atmospheric deposition of PAHs and PCBs on the surface water of the Netherlands (2 793 km^2), Waddenzee (2 600 km^2), North Sea (508 126 km^2 in* ***t/yr****), IJsselmeer (1 928 km^2) and the soil of the Netherlands (36 783 km^2) in (kg/yr). The reliability for the substances printed* ***bold*** *is good. The reliability of the other substances is low (see also text). The estimates are based upon measurements of wet deposition, air concentration and an estimate of the dry deposition velocity*

Atmospheric deposition	**Surface water NL**	**Surface water NL**	**Surface water Noord-Holland**	**North Sea** [1)]	**Wadden-zee**	**Wadden-zee**	**IJssel-meer**[2)]	**IJssel-meer**[2)]	**Soil**	**Soil**
	2000	**2001**	**2001**	**2001**	**2000**	**2001**	**2000**	**2001**	**2000**	**2001**
	kg/yr	**kg/yr**	**kg/yr**	**t/yr**	**kg/yr**	**kg/yr**	**kg/yr**	**kg/yr**	**kg/yr**	**kg/yr**
PAHs										
acenaphtene	700	454	139	92	712	461	463	268	800	498
acenaphtylene	52	44	13	2	19	22	29	23	128	98
anthracene	178	46	13	3	60	35	176	23	361	162
benzo[a]anthracene	36	19	6	3	23	17	22	12	336	207
benzo[a]pyrene	60	30	10	6	36	31	48	18	425	170
benzo[b]fluorantene	138	82	29	14	72	78	113	48	1131	520
benzo[g,h,i]perylene	53	33	12	6	33	34	41	20	810	553
benzo[k]fluorantene	89	59	21	11	46	61	80	35	1099	734
chrysene	132	56	17	8	71	46	88	35	545	426
dibenz[a,h]anthracene	8	7	3	1	4	10	6	5	79	74
fenantrene	2416	1974	645	160	1863	1442	1481	1085	4735	3176
fluorantene	861	710	262	68	553	590	560	419	4744	3646
fluorene	1254	950	265	127	1142	711	793	551	1378	953
indeno[1,2,3-cd]pyrene	49	32	11	7	29	34	38	18	487	263
naphthalene	1965	1786	567	102	1188	1017	1489	1215	1630	931
pyrene	484	382	121	32	274	296	269	215	2128	1304
PCBs										
PCB-101	2.5	0.6	0.1	0.3	3.6	0.8	1.4	0.5	29.4	4.8
PCB-118	0.8	0.5	0.01	0.0	0.8	0.1	0.5	0.7	10.9	6.5
PCB-138	0.9	0.5	0.1	0.0	0.7	0.2	0.5	0.5	10.8	6.3
PCB-153	1.1	0.7	0.1	0.1	0.8	0.4	0.7	0.6	13.0	9.0
PCB-180	0.4	0.2	0.1	0.0	0.1	0.0	0.6	0.3	6.7	2.8
PCB-20	2.1	1.3	0.4	0.5	2.1	1.7	0.9	0.7	19.6	14.2
PCB-28	2.6	1.0	0.2	0.5	3.2	1.4	1.1	0.5	27.2	11.9
PCB-35	0.2	0.2	0.1	0.0	0.3	0.2	0.0	0.2	1.5	1.9
PCB-52	4.2	1.5	0.3	0.6	8.4	2.1	2.0	1.3	45.2	15.0
PCB-8	0.9	0.4	0.1	0.1	0.4	0.2	0.4	0.2	6.8	2.5

1) The estimates of the deposition of the North Sea are uncertain (see text).

2) The IJsselmeer is defined as the public waters in the IJsselmeer area.

Good measurement of the deposition of all PAHs and PCBs appears to be possible with the chosen method. Just as the concentrations in precipitation, the deposition of most PAHs had in 2001 decreased in comparison with the year 2000, by an average of 40 per cent. Only the deposition of *dibenz[a,h]-anthracene* increased slightly. The deposition of PCBs had in the year 2001 decreased by 30 per cent in comparison with the year 2000. Many of the maximum deposition values for PCBs and PAHs were found at the coast and in Noord-Holland: Egmond, Aalsmeer, and Noordwijk (Annex III).

3.4 Calculation of atmospheric deposition by means of a dispersion model

The atmospheric deposition over the Netherlands calculated on the basis of the monitoring results enables evaluation, and possibly improvement, of the quality of models that calculate the dispersion and deposition of pesticides. These models can then be used in policy studies because this enables quantification of the relationship between emission and deposition. This makes it possible to compute the effect of policy scenarios.

Model calculations of the dispersion and deposition of a number of pesticides and PAHs that were found a lot in the measurements were made by using data on the emissions from agriculture in the Netherlands and rough estimates of emissions in surrounding countries. Model calculations carried out with emissions in the Netherlands yield the contributions from the Netherlands to the deposition in the Netherlands; calculations with emissions from surrounding countries yield the contribution by surrounding countries to the deposition in the Netherlands. The sum of both contributions yields the total deposition in the Netherlands.

No measurements have been carried out for *carbendazim* and *glyphosate.* The deposition has therefore been derived from model calculations carried out for those substances. The following table shows the calculated load. The uncertainty in the results is unknown because monitoring data are lacking.

Deposition of carbendazim *and* glyphosate *to the surface water of the Netherlands, the IJsselmeer, the surface water in Noord-Holland and the soil of the Netherlands expressed in kg/year as derived from model calculations.*

	Surface water	**IJsselmeer**	**Surface water Noord-Holland**	**The soil of the Netherlands**
carbendazim	16	6	6	278
glyphosate	68	40	35	1613

3.4.1 Comparison of model results with measured wet deposition in the Netherlands

The model calculations, in which emissions and meteorological data for the year 2000 have been used, have been compared with measured wet deposition of the year 2000 (Figure 19). For this comparison only substances have been chosen that were found in more than ten per cent of the samples and for which sufficient data were available. Comparison of dry deposition is more difficult because substances were found less frequently. Furthermore, the estimates made by means of the model as well as the estimates on the basis of measurements are based on the same deposition rate.

In some cases, the depositions calculated for the year 2000 have been compared with the monitoring results in the year 2001. The emission figures were in that case adjusted for changes in the use of substances in 2001. This applies for *chlorothalonil, ethofumesate, terbutylazine, tolclofos-methyl* and *vinclozolin* which were in 2001 determined at all 18 locations and in 2000 only at four locations. In view of the strong correlation between measured depositions in the year 2000 and 2001, this comparison of measurements from the year 2001 can still present a picture of the quality of the modelling.

Table 10 shows the correlation between measured and modelled deposition and the correlation coefficient of the regression between both.
There was a reasonable fit between models and measurements for a number of substances. The correlation between measured and modelled deposition was between a half (model overestimates a factor two) and two (model underestimates a factor two). This applies for *atrazine, chlorpropham* (2000), *ethofumesate* (Figure 29), *procymidon* (Figure 23), *propachlor* and *terbutylazine* (Figure 25). There was also a good correlation between measurements and model calculations for these substances. This indicates that the model properly calculated the spatial distribution of those substances and that the spatial distribution of the emissions was good. Figure 22 an 23 show the course of the wet deposition of *atrazine* and *procymidon* over the Netherlands; *procymidon* is an example of a substance that is used a lot on a local scale (in bulb growing). This picture emerges from the calculated as well as the measured concentration. The *atrazine* picture is completely different. The use of this substance is no longer permitted since 1999. Nevertheless, measurable concentrations of the substance are still found in precipitation. The fit between the measured course of the deposition over the country and the deposition calculated on the basis of emissions only in surrounding countries supports the picture that the *atrazine* that was found does probably originate from surrounding countries.

For *chlorothalonil (*Figure 27*), DNOC* (2001), and *vinclozolin (*Figure 31), the order of magnitude of the modelled deposition was good (factor ½ to two), but the correlation between model and monitoring results was poor.
The ratio between measured and modelled deposition was higher than ten (model underestimates by more than a factor ten) or lower than one tenth (model overestimates more than a factor 10) for *dichlorvos (*Figure 26) and *fluazinam*. The correlation between calculated and measured concentration was reasonable for those substances. This could mean that the allocation of the emissions in the Netherlands is still fairly good, but that the estimates of the source strength were less accurate

The comparison between measured and calculated wet deposition was poor, regarding the correlation as well as the order of magnitude of the deposition, for *bentazon, dichlobenil, dimethoate, pirimiphos-methyl, tolclofos-methyl* and *triallate* (2000). The correlation was good for *triallate* in 2001 (Figure 30).

Table 10 Notes on the comparison between de measured and modelled concentrations of pesticides in the Netherlands. Modelling was done with the emissions in the Netherlands plus surrounding countries.

Substance	**Remark** [2)]
atrazine	Emissions underestimated
bentazon	Quality of the measurements possibly insufficient
chlorpropham	Within factor 2
chlorothalonil[1)]	Within factor 2
dichlobenil	Emissions underestimated
dichlorvos	Within factor 2
dimethoate	Quality of the measurements possibly insufficient
DNOC	Emissions underestimated
ethofumesate[1)]	Within factor 2
fluazinam	Emissions overestimated
procymidon	Within factor 2
propachlor	Within factor 2
terbutylazine[1)]	Within factor 2
tolclofosmethyl[1)]	Quality of the measurements possibly insufficient
triallate	Emissions underestimated
vinclozolin[1)]	Within factor 2

1) Substances measured at four locations.

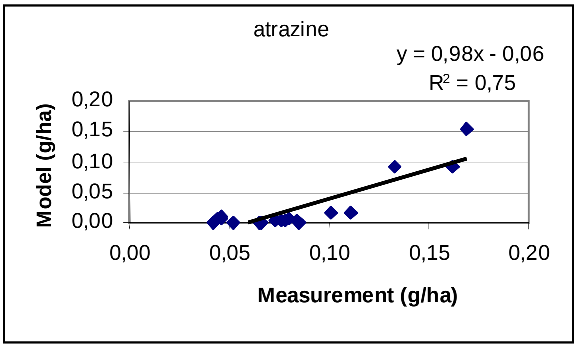
2) The remarks ‘Emissions underestimated’ or ‘Emissions overestimated’ indicate that model parameters such as emissions, transformation rates or wash-out coefficients are probably incorrect. A deviation in the emission factor, however, is most likely.

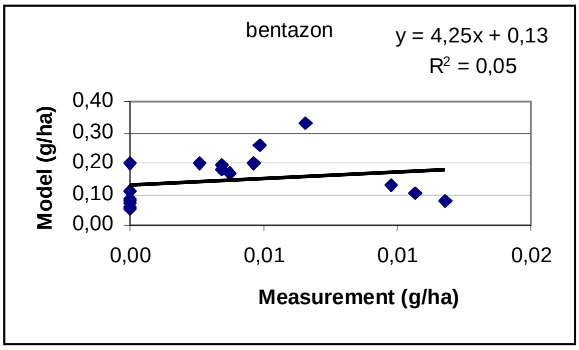
This table and figures 19 and 20 show that the model:

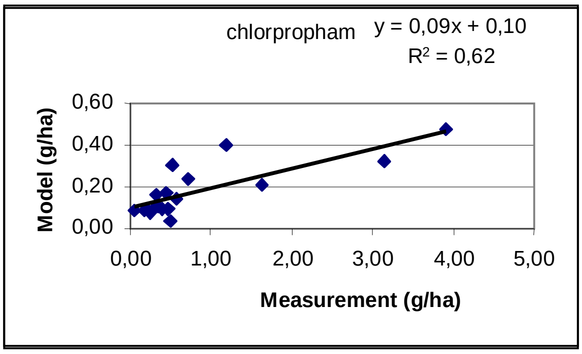
– calculates the right order of magnitude of deposition for atrazine, chlorpropham, chlorothalonil, dichlorvos, ethofumesate, procymidon, propachlor, terbutylazine (2001) and vinclozolin,

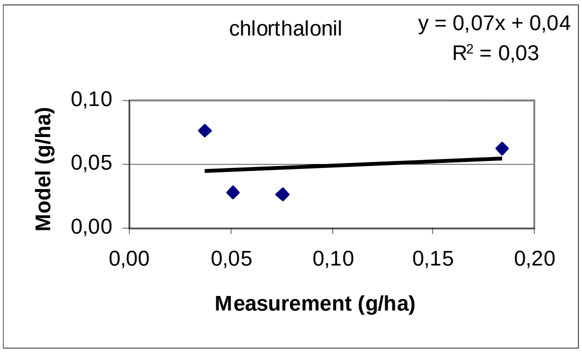
– strongly overestimates the deposition of bentazon, dimethoate and terbutylazine (2000),

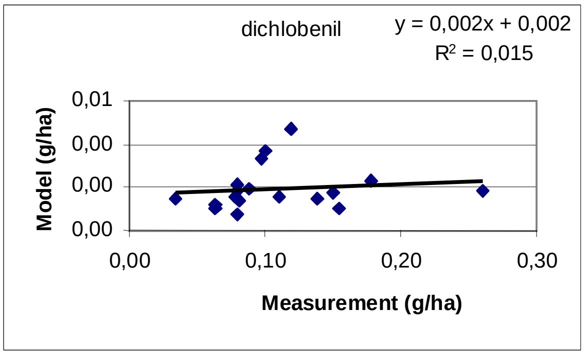
– strongly underestimates the deposition of dichlobenil and triallate, despite the fact that contributions from surrounding countries have not been calculated.

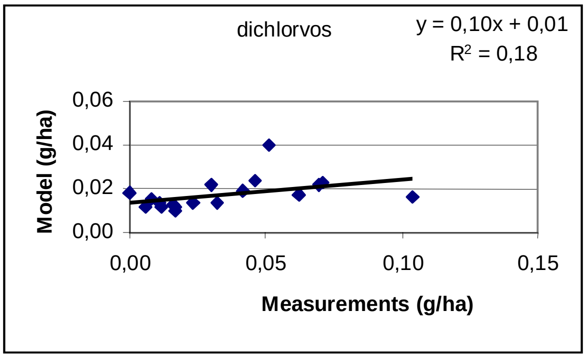

atrazine
y = 0,98x - 0,06
R^2 = 0,75
Model (g/ha)
0,20
0,15
0,10
0,05
0,00
0,00
0,05
0,10
0,15
0,20
Measurement (g/ha)

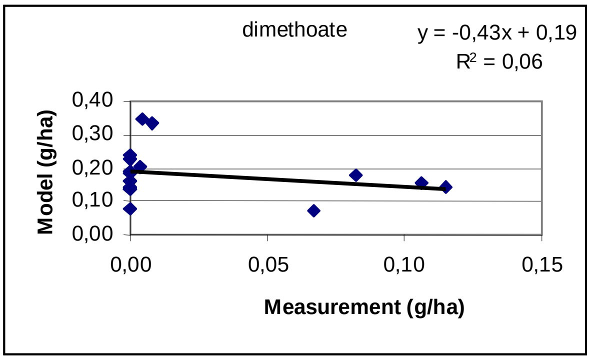

bentazon
y = 4,25x + 0,13
R^2 = 0,05
Model (g/ha)
0,40
0,30
0,20
0,10
0,00
0,00
0,01
0,01
0,02
Measurement (g/ha)

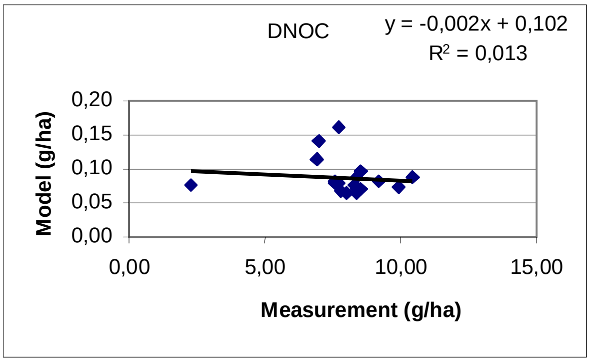

chlorpropham
y = 0,09x + 0,10
R^2 = 0,62
Model (g/ha)
0,60
0,40
0,20
0,00
0,00
1,00
2,00
3,00
4,00
5,00
Measurement (g/ha)


chlorthalonil
y = 0,07x + 0,04
R^2 = 0,03
Model (g/ha)
0,10
0,05
0,00
0,00
0,05
0,10
0,15
0,20
Measurement (g/ha)


dichlobenil
y = 0,002x + 0,002
R^2 = 0,015
Model (g/ha)
0,01
0,00
0,00
0,00
0,00
0,10
0,20
0,30
Measurement (g/ha)


dichlorvos
y = 0,10x + 0,01
R^2 = 0,18
Model (g/ha)
0,06
0,04
0,02
0,00
0,00
0,05
0,10
0,15
Measurements (g/ha)


dimethoate
y = -0,43x + 0,19
R^2 = 0,06
Model (g/ha)
0,40
0,30
0,20
0,10
0,00
0,00
0,05
0,10
0,15
Measurement (g/ha)


DNOC
y = -0,002x + 0,102
R^2 = 0,013
Model (g/ha)
0,20
0,15
0,10
0,05
0,00
0,00
5,00
10,00
15,00
Measurement (g/ha)

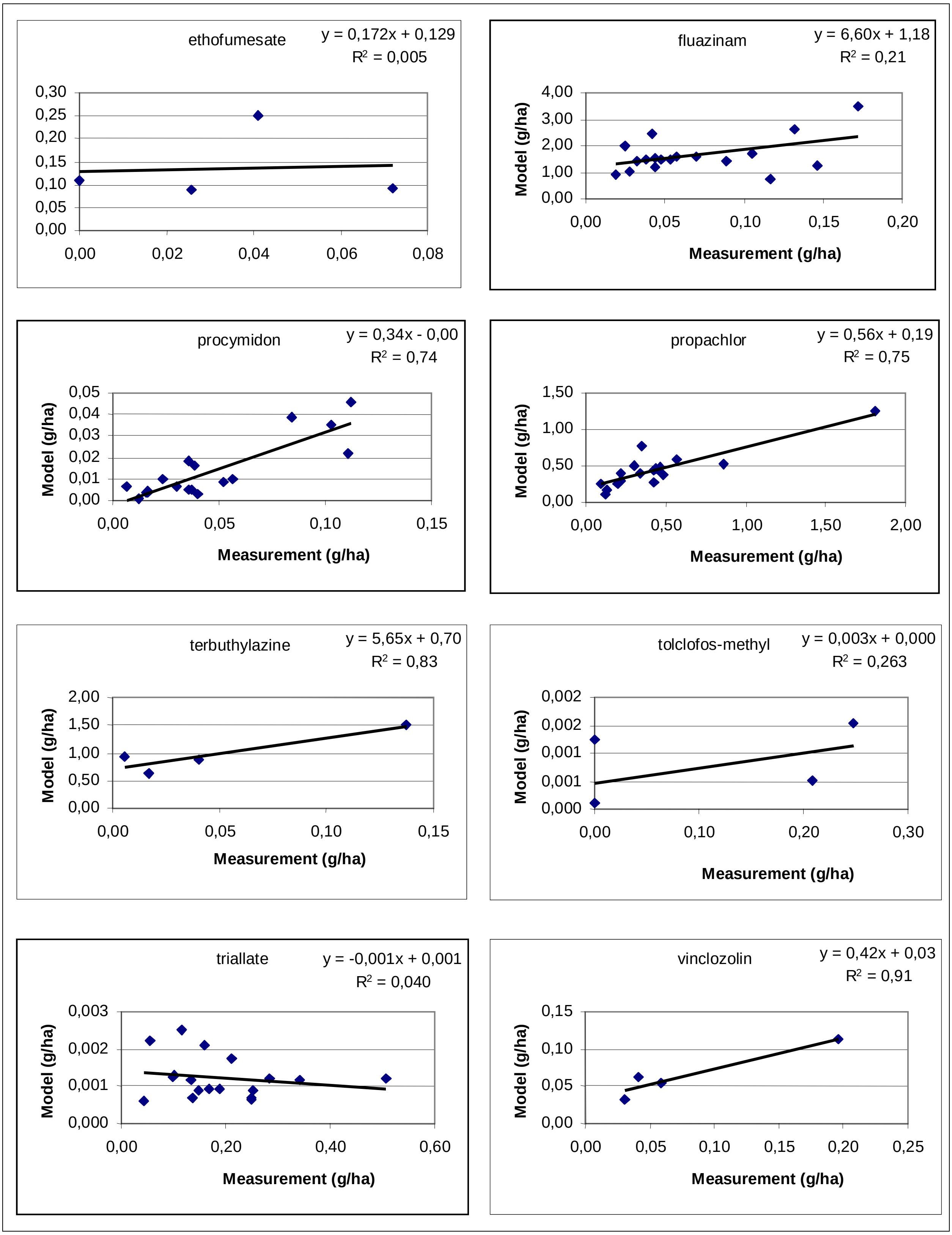


Figure 19 *Comparison between values calculated with the OPS model and measured values of the wet deposition of a number of important pesticides in 2000.*

The comparison between the model results and measurements in 2001 at 18 monitoring stations for *chlorothalonil*, *ethofumesate*, *terbutylazine*, *tolclofos-methyl* and *vinclozolin* is shown in Figure 20.

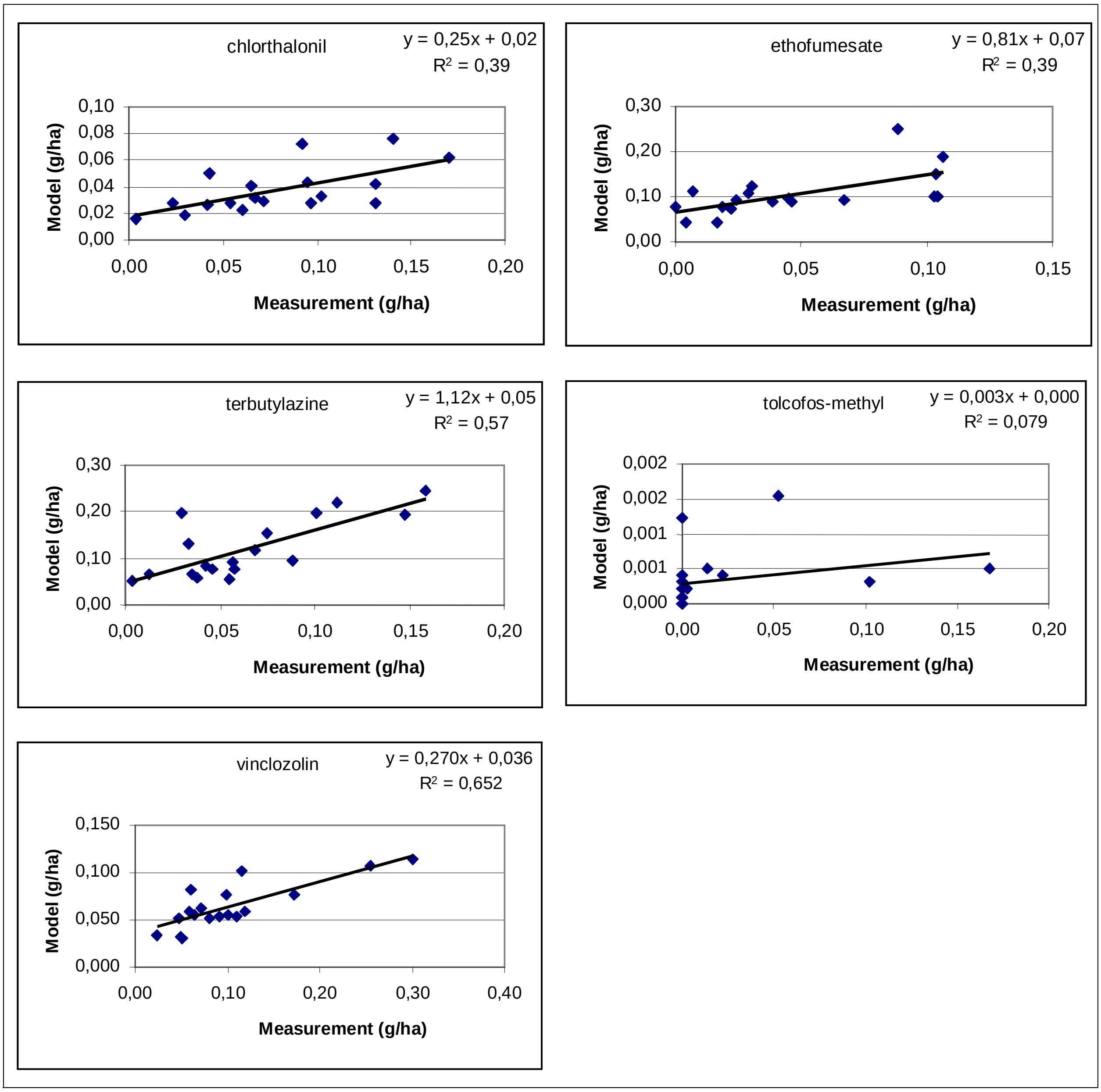


Figure 20 *Comparison of the wet deposition of a number of important pesticides for the year 2001 calculated with the OPS model and the measured values for which the emission figures of 2001 were used and the meteorological data of 2000. These are pesticides that were in 2000 determined at four locations and in 2001 at eighteen locations.*

The calculated wet deposition of the PAHs *benzo(a)pyrene* and *fluoranthene* has also been compared with the measured values. Figure 21 still shows some correlation between measurement and model for *benzo(a)pyrene.* The model, however, underestimated the measured wet deposition by a factor of about three. There is also little correlation between the measured and calculated deposition of *fluoranthene*. The estimate of the absolute level of the deposition is still reasonable.

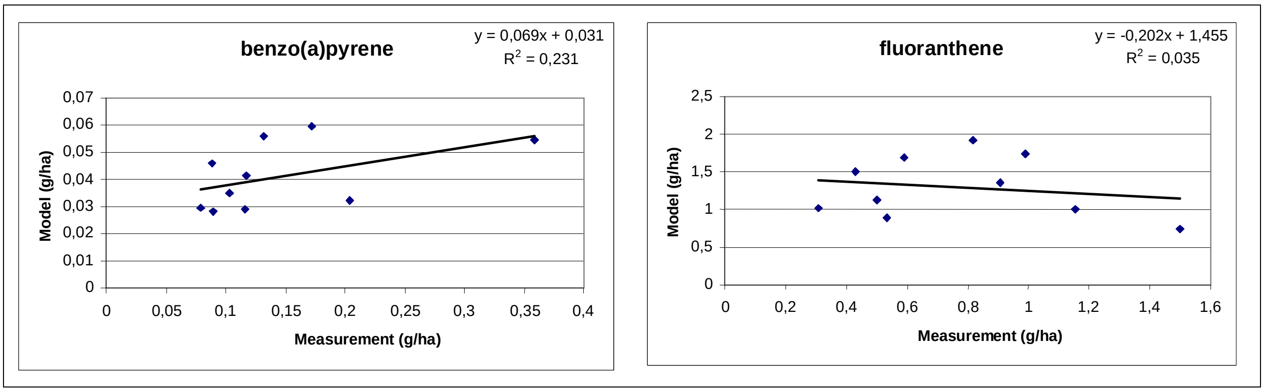


Figure 21 Correlation between calculated and measured wet deposition of fluoranthene and benzo[a]pyrene in year 2000.

The correlation is poor for: *DNOC, dimethoate, pirimiphos-methyl, tolclofos-methyl* and *triallate.*
No estimates were available about the emissions of *dichlobenil* in surrounding countries.

Figures 22 - 30 show the spatial distribution of the measured and modelled wet deposition of *atrazine*, *procymidon*, *propachlor*, *terbutylazine*, *ethofumesate* and *triallate* over the Netherlands.

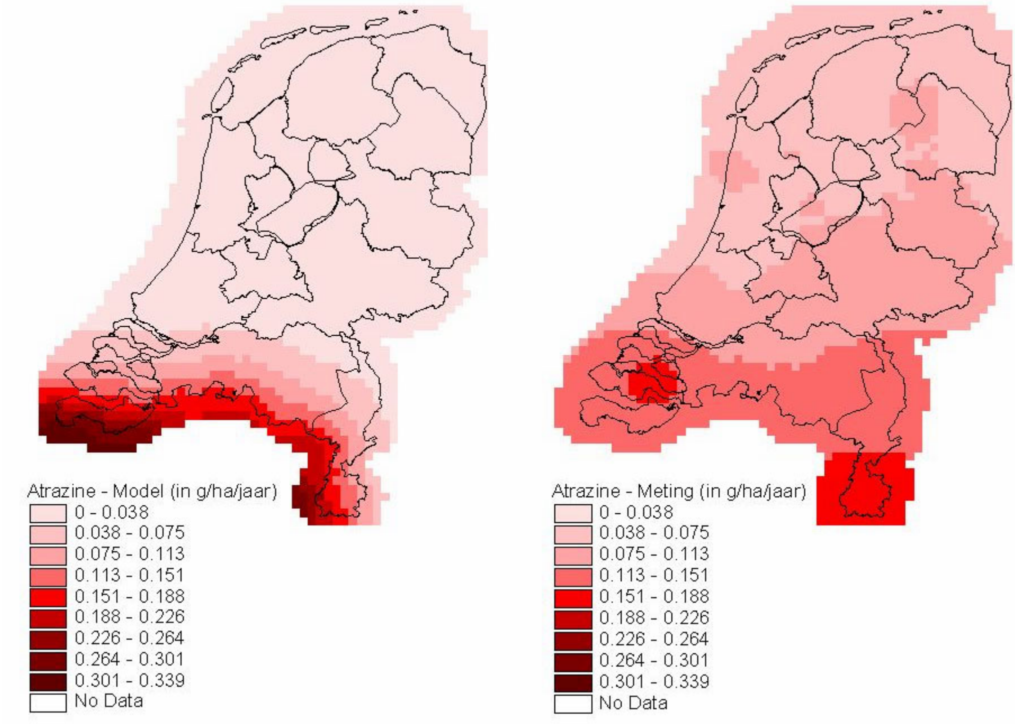


Figure 22 Spatial distribution of the modelled (Model) and measured (Meting) wet deposition of atrazine (g/ha/yr) for the year 2000.

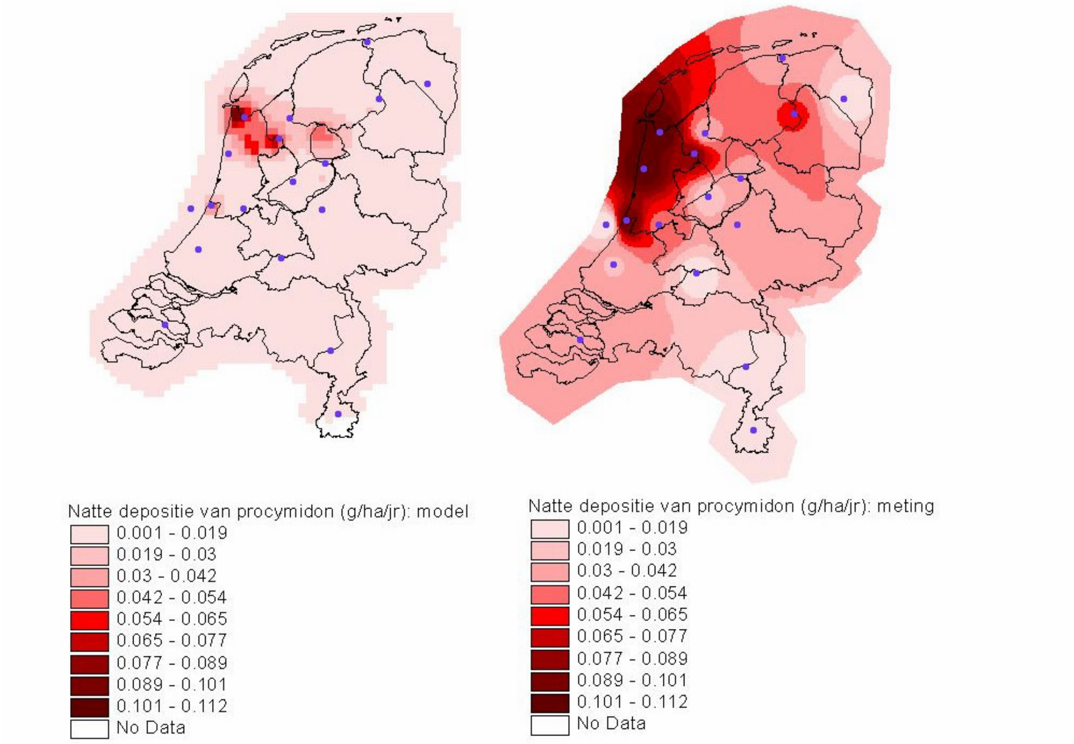


Figure 23 Spatial distribution of the modelled (model) and measured (meting) wet deposition (Natte depositie) of procymidon (g/ha/yr) for the year 2000.

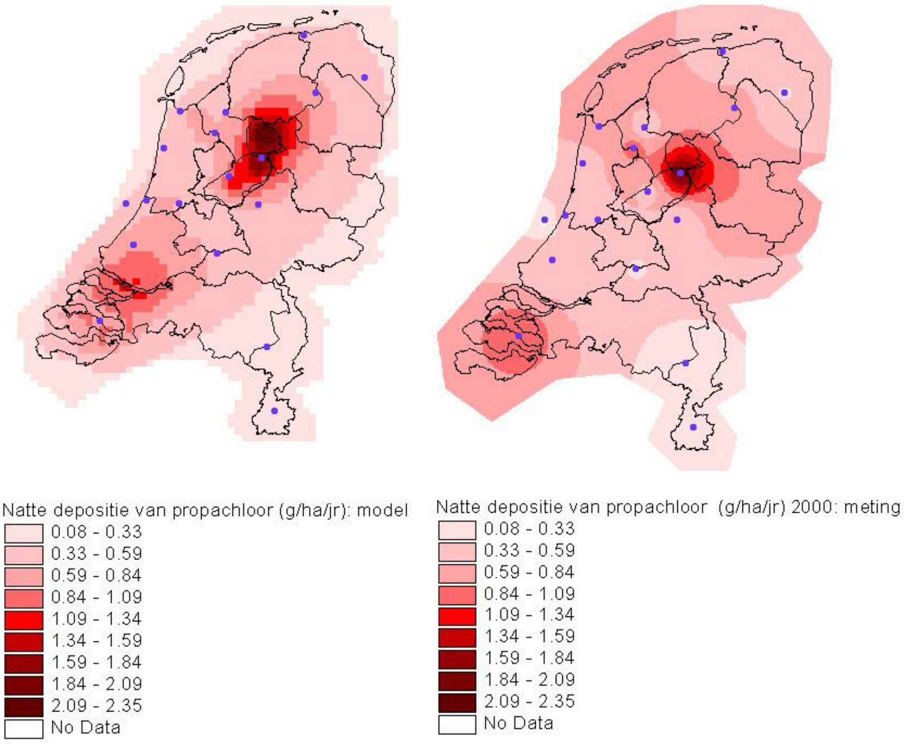


Figure 24 *Spatial distribution of the modelled (model) and measured (meting) wet deposition (Natte depositie) of propachlor (g/ha/yr) for the year 2000.*

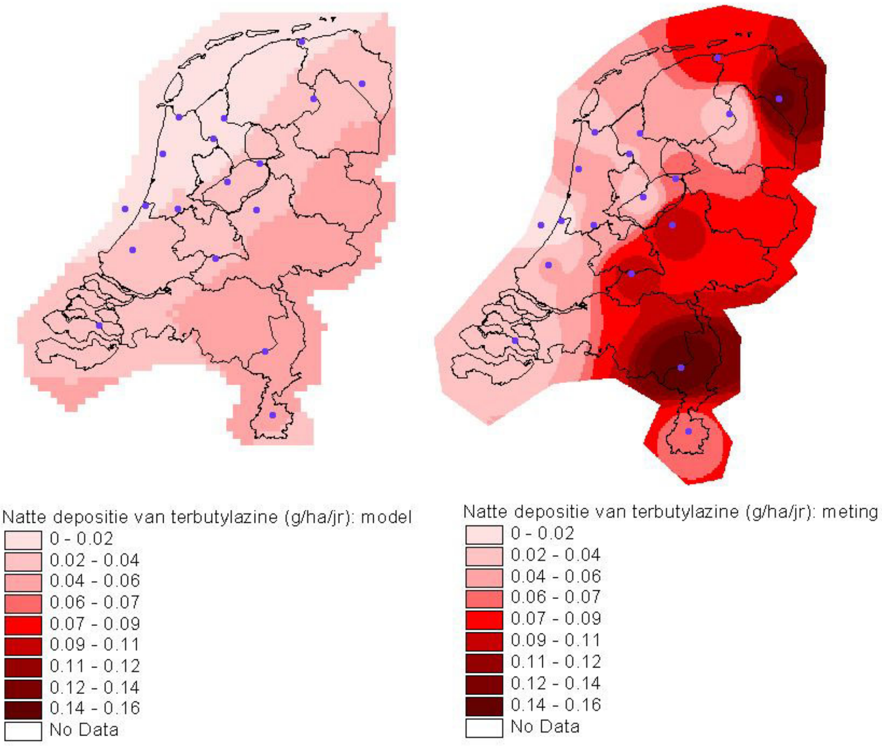


Figure 25 *Spatial distribution of the modelled (model) and measured (meting) wet deposition (Natte depositie) of terbutylazine (g/ha/yr) for the year 2001.*

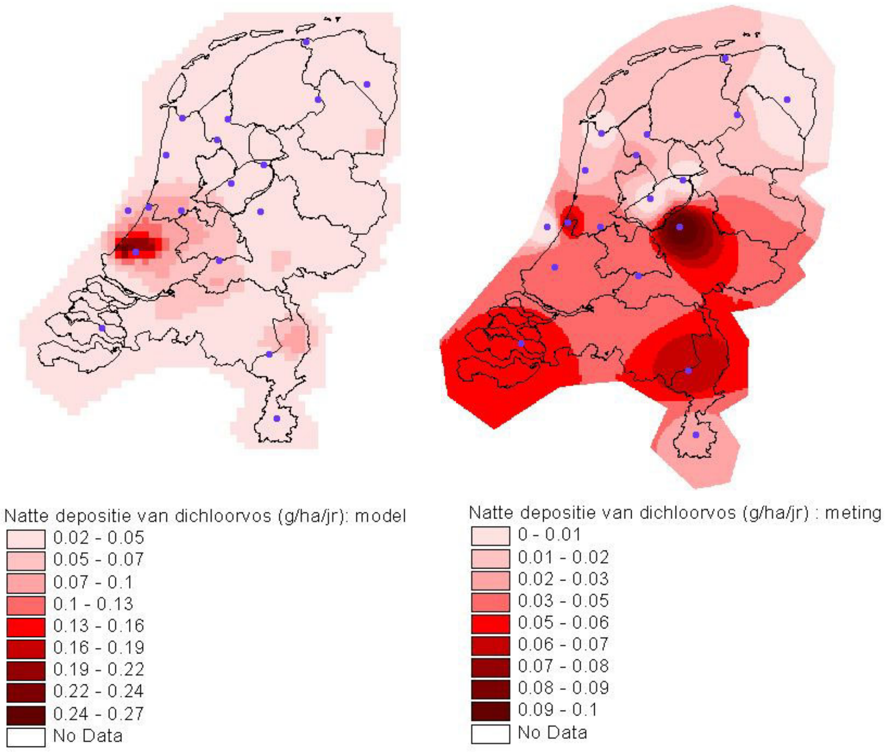


Figure 26 *Spatial distribution of the modelled (model) and measured (meting) wet deposition (Natte depositie) of dichlorvos (g/ha/yr) for the year 2000.*

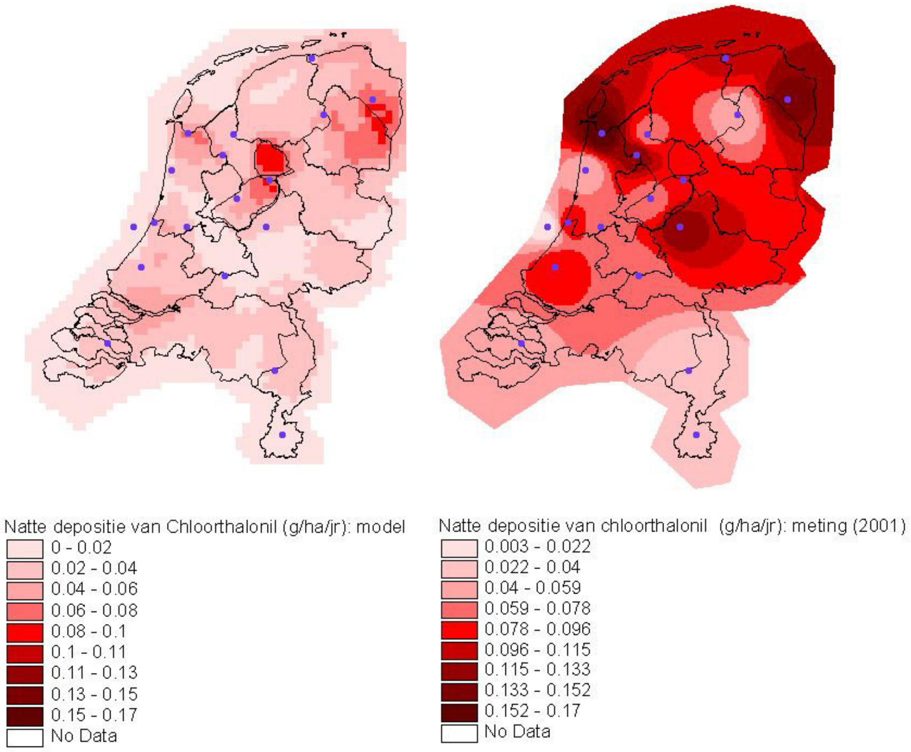


Figure 27 *Spatial distribution of the modelled (model) and measured (meting) wet deposition (Natte depositie) of chlorothalonil(g/ha/yr) for the year 2001.*

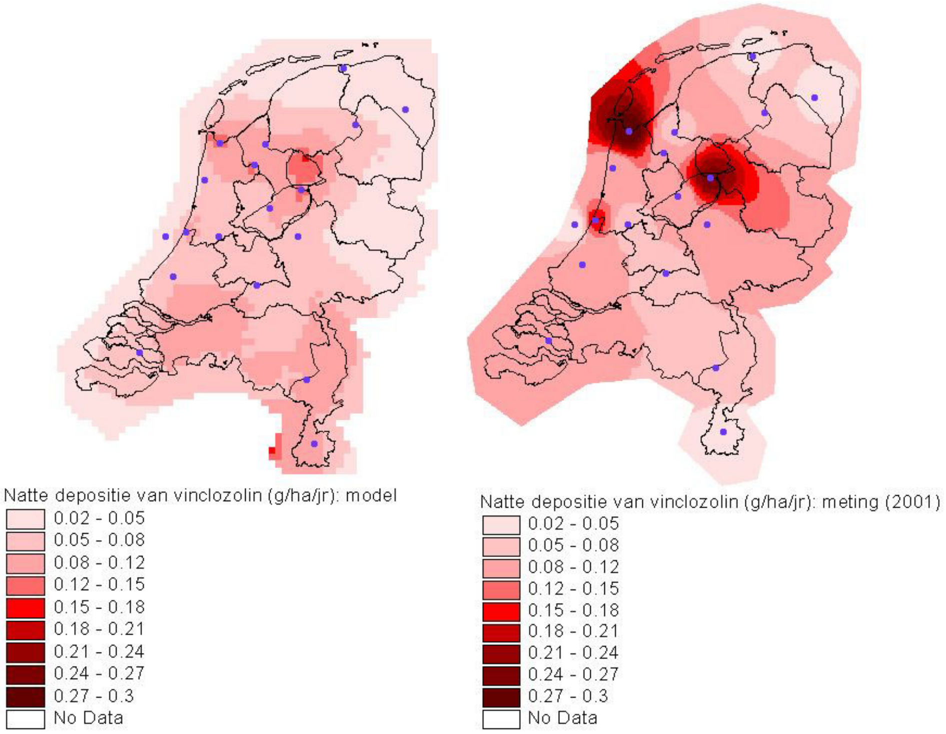


Figure 28 Spatial distribution of the modelled (model) and measured (meting) wet deposition (Natte depositie) of vinclozolin (g/ha/yr) for the year 2001.

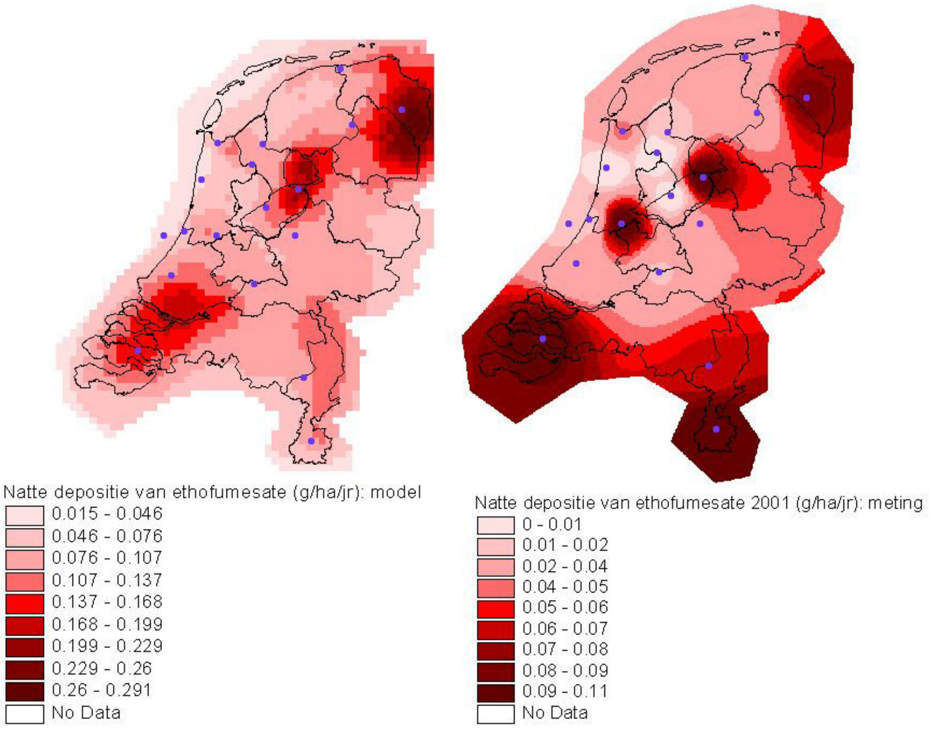


Figure29 Spatial distribution of the modelled (model) and measured (meting) wet deposition (Natte depositie) of ethofumesate (g/ha/yr) for the year 2001.

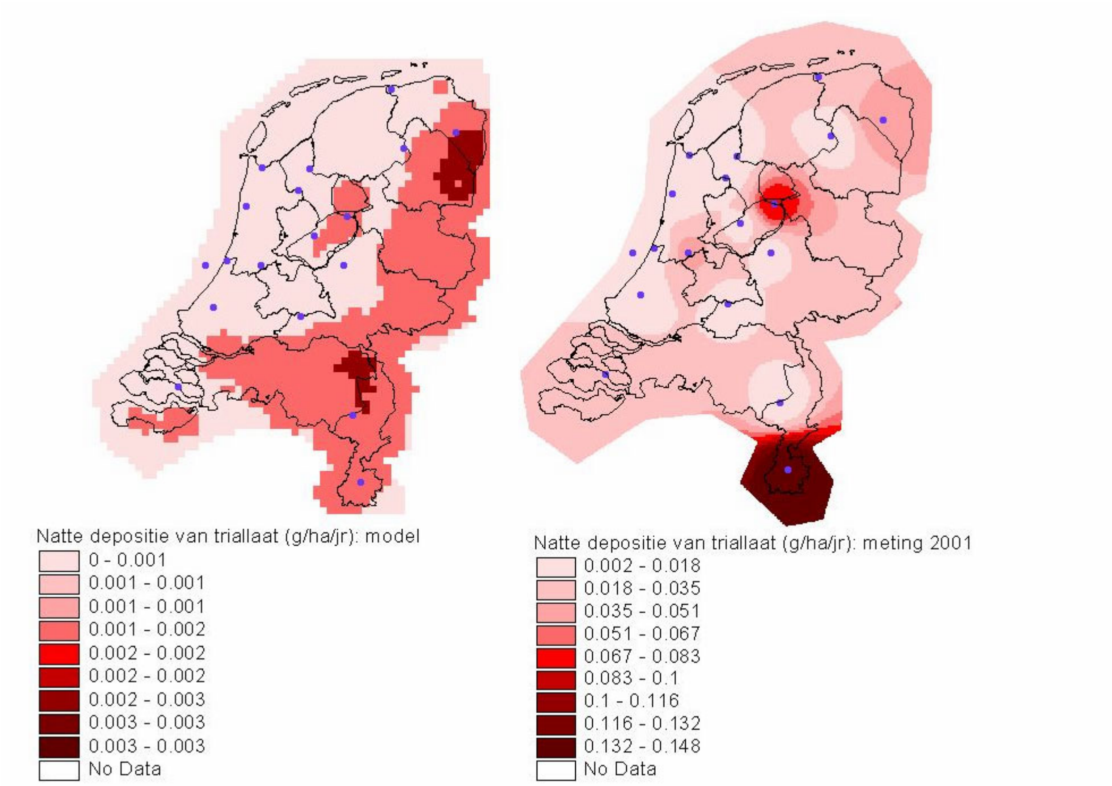


Figure 30 Spatial distribution of the modelled (model) and measured (meting) wet deposition (Natte depositie) of triallate (g/ha/yr) for the year 2001.

These figures show that the correlation between measured and modelled wet deposition and the order of magnitude of the modelled wet deposition for these pesticides is fairly good, except for *triallate*. The correlation for this herbicide in the year 2001 is reasonable (Figure 30), but the order of magnitude of the modelled deposition is poor. The emission is probably strongly underestimated. The quality of the measurements of *triallate* in the year 2000 is probably low in view of the poor correlation between measurement and model for that year.

3.4.2 Contributions of emissions of the different provinces to depositions in water board districts

The transport and deposition model OPS can be used to determine the correlation between sources (emission of substances) and deposition; the source of certain deposition can be calculated. Calculation of transport and deposition of the emission per source, or the emission per province, with the model enables calculation of the contribution of that source or province. In the context of this study, the following calculations have been made for a limited number of substances:
1) the deposition in a water control area, and
2) the contribution of the emissions from the different provinces in such an area.

The distribution of the emissions of a substance over the country can be used to estimate the deposition to an area of that substance, as well as the proportion of emissions outside that area. The contribution of emissions in the different provinces of the Netherlands to the input in water control areas has been calculated for four pesticides and two PAHs. Figures 31 – 34 show these contributions for the pesticides *procymidon, fluazinam, dichlorvos* and *chlorothalonil* and for the PAH *benzo(a)pyrene*. In view of the correlation between the model calculations and measurements (Table 10), the model is capable to produce reasonable to good calculations of the spatial distribution of these substances over the country. Besides the contributions, the average input per area is also shown. Possible over- or underestimates of the measured deposition values made by the model should, however, be taken into account in the interpretation of the figures. Figure 32, e.g., shows that the average deposition of ‘waterschap Hunze and de AA’ with *fluazinam* is about 3.3 g per ha, whereas the average measured deposition of this substance was not higher than 0.45 g per ha at the monitoring point with the highest value in the Netherlands. The model underestimated the deposition for *dichlorvos* and *chlorothalonil*. Modelling of the atmospheric deposition of *procymidon* showed good agreement with the measurements. The average measured total deposition of *procymidon* in Noord-Holland was 0.08 gram per hectare per year.
Figure 31 shows that a large part of this deposition originates from the own province and that the total input to water quality control areas there ranges from 0.04 to 0.07 gram per hectare per year.
Generally, deposition depends very strongly on the position of an area in relation to source areas. *Dichlorvos*, mainly used in the Westland, causes high depositions in the control area of de Hoogheemraadschappen Schieland and Delfland. There is the same correlation for *procymidon*, which has an important use in the area of Hoogheemraadschap Uitwaterende Sluizen. Substances used a lot at a national scale, such as *fluazinam* and *chlorothalonil,* contribute to the deposition in all control areas.
Recently, Bleeker and Duyzer (2002) conducted an extensive study into the atmospheric deposition onto surface water in the control area of the Zuiveringsschap Hollandse Eilanden en Waarden (ZHEW). This showed that the deposition of the different pesticides used in the area did for about 25-30% originate from emissions from the area itself.

The contribution of emissions in the different provinces in the Netherlands to the deposition at each of the monitoring locations, and which part of the deposition originated from the surrounding countries, has also been calculated. These data are given in Annex VII.

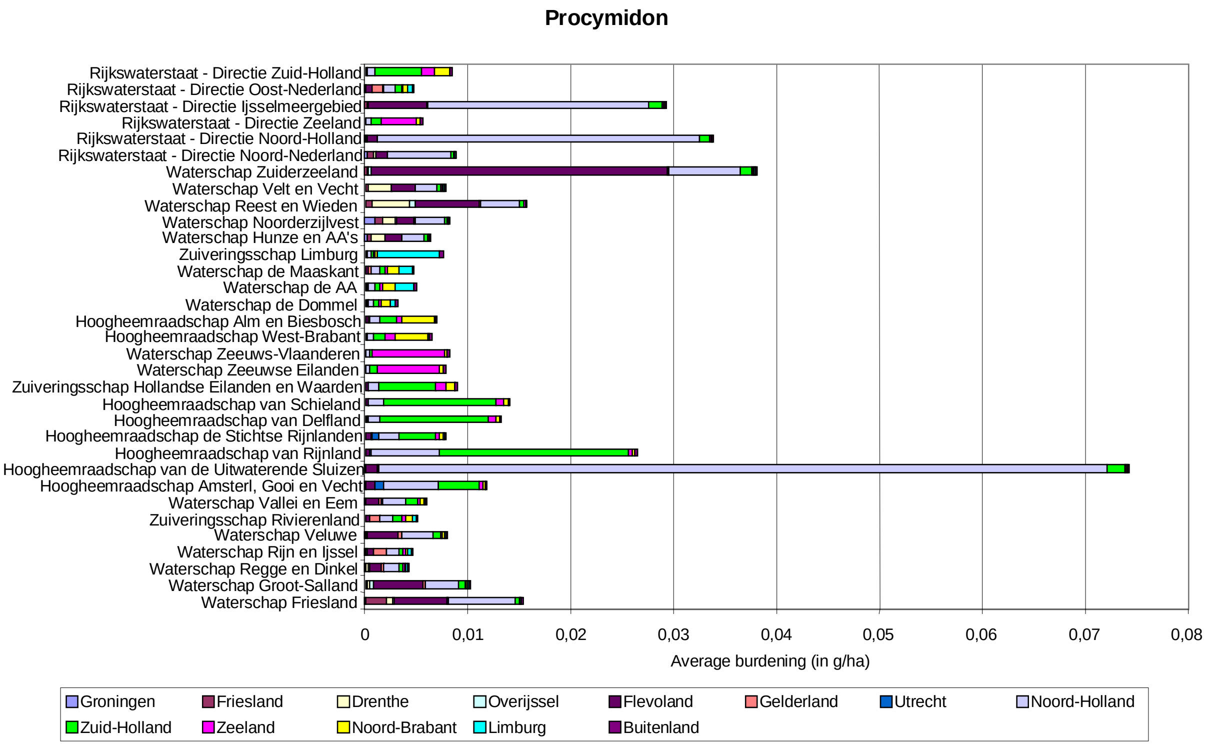


Figure 31 *Contributions of procymidon emissions per province to the average deposition with procymidon in the water quality control areas (g/ha).*

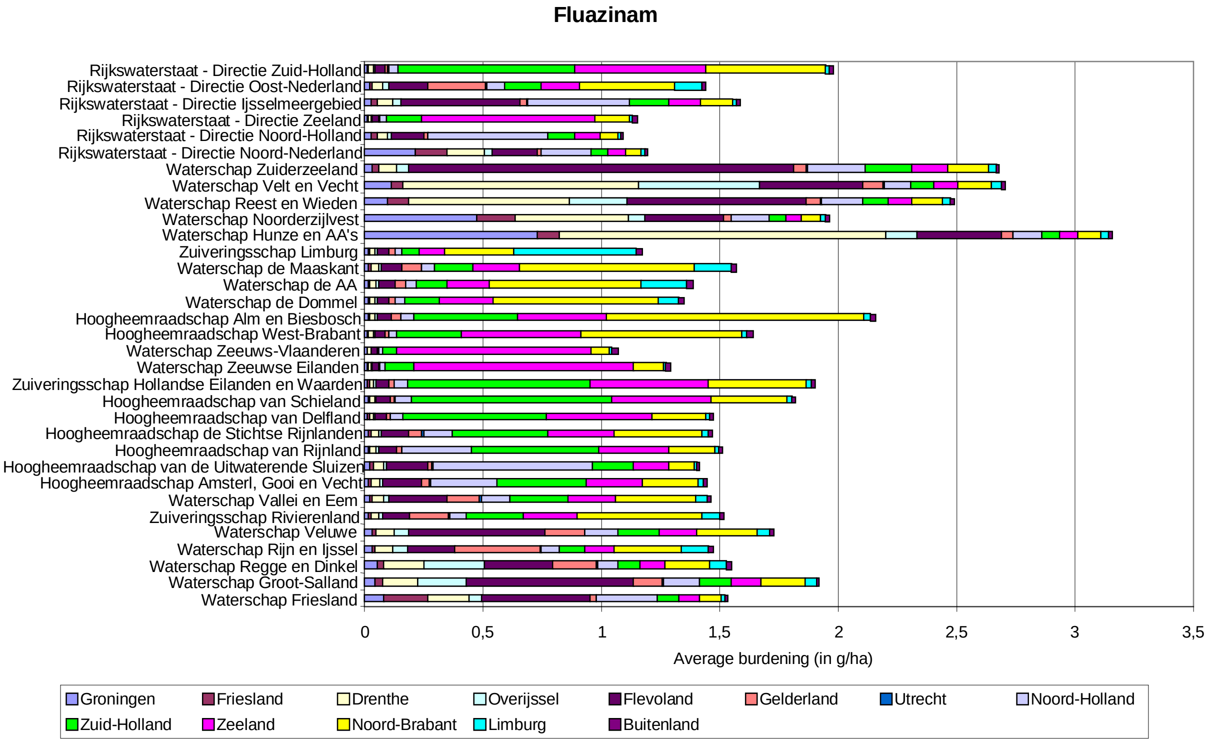


Figure 32 *Contributions of fluazinamemissions per province to the average deposition in the water quality control areas (g/ha).*

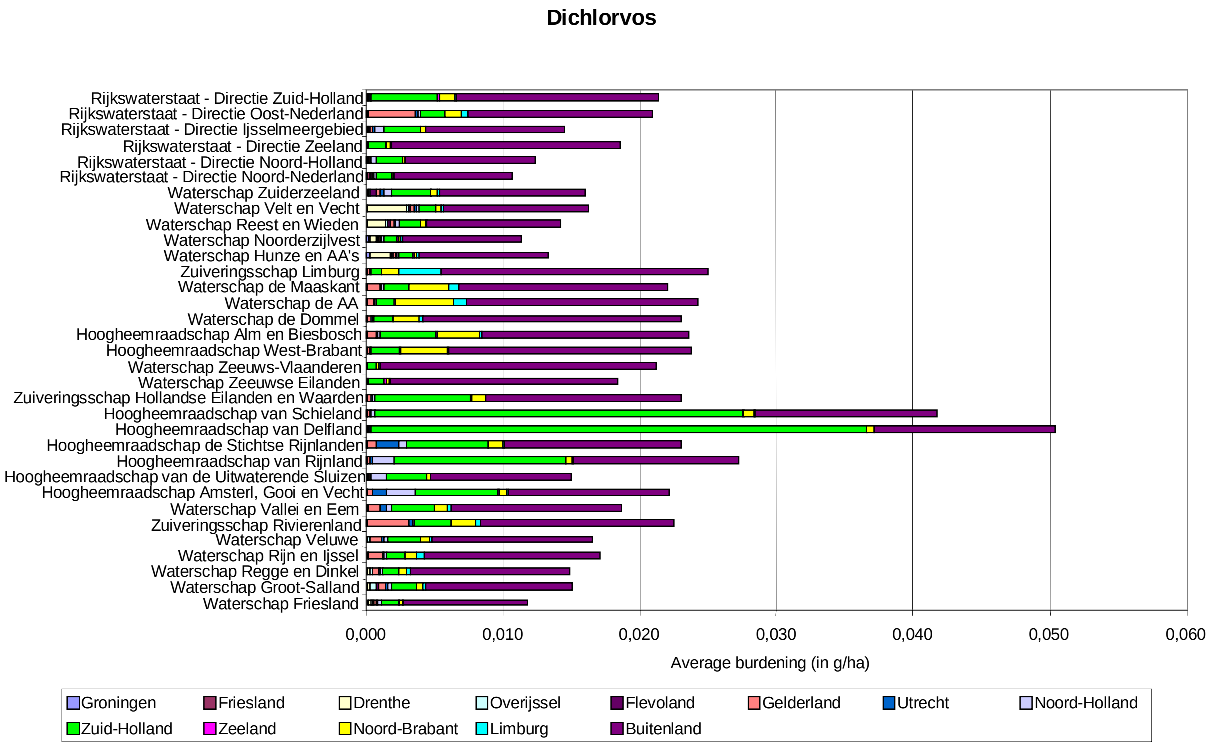


Figure 33 Contributions of dichlorvos emissions per province to the average deposition in the water quality control areas (g/ha).

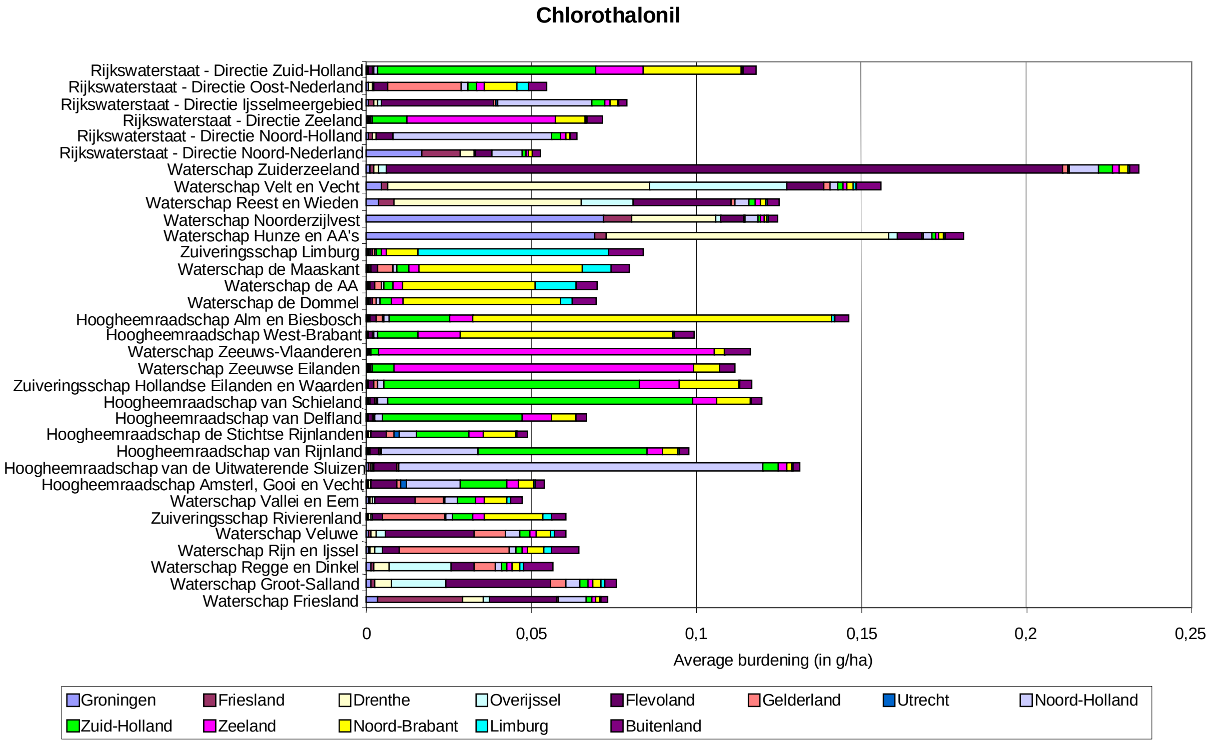


Figure 34 Contributions of chlorothalonil emissions per province to the average deposition in the water quality control areas (g/ha).

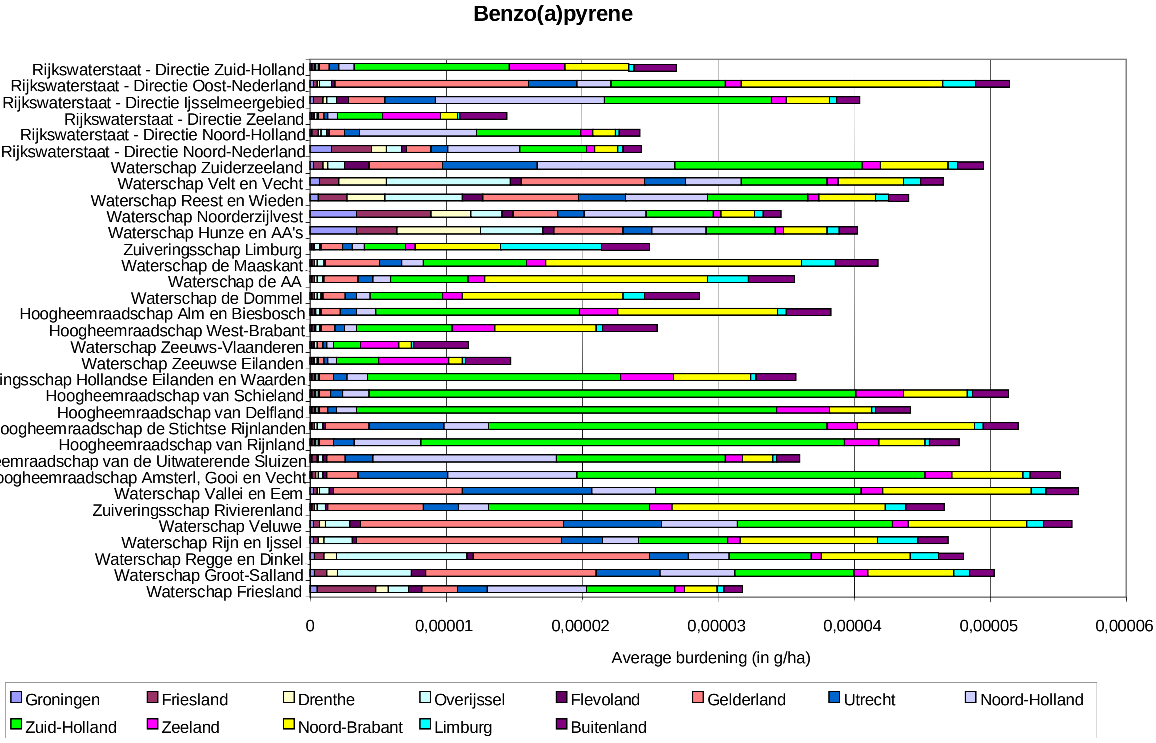


Figure 35 *Contributions of benzo(a)pyrene emissions per province and from surrounding countries to the average deposition in the water quality control areas (g/ha).*

3.4.3 Effect of atmospheric deposition of pesticides on the concentrations in surface water; comparison with criterion values

Substances are subjected to various processes after getting into the surface water via atmospheric deposition. These processes, including dilution and degradation, finally lead to the concentration becoming lower than the concentration in the atmospheric deposition. The concentration in the aqueous phase can be calculated by means of simulation models. Different models have been described in the literature. The *Waterbox* model (Bakker and Van den Hout, 1993) has been used in this study. This box model calculates the fate of substances in water while taking the following processes into account:
- adsorption of the substance to suspended sludge and sediment
- diffusion of a substance between water and sediment
- degradation of the substance in water and sediment.

The model calculates the concentration in the situation that complete equilibrium has been reached between all input and output processes. This concentration is calculated for different standard waters such as ditches, pools, rivers and lakes. Because dilution is lowest for the aquatic system *pool*, the highest concentration is calculated for this system. The concentration calculated for *pools* is therefore in this study used as indicator of possible problems. The concentration in the other aquatic systems remains much lower. As an illustration, the concentration in a large lake is presented in Table 11 as well. The table presents the concentration as result of the atmospheric deposition calculated for a number compounds. Input by other sources such as drift has not been taken into account.

Calculations were based on the annual average atmospheric deposition on surface water in the Netherlands. The calculations have been made for a number of pesticides that were found in surface water in 2001.

Table 11 Concentrations in a pool and in a large lake (in ng/l and as percentage of the MTR) as result of atmospheric deposition only, calculated with the Waterbox model. The relationship between the highest deposition found in the Netherlands at a certain location and the average deposition for the Netherlands that has been used, are also given. The isoproturon concentration has only been determined in samples of one location. The triphenyltin has been determined at four locations.

Substance	**MTR (ng/l)**	**Concentration in a pool (ng/l)**	**Concentration in a large lake (ng/l)**	**Concentration in a pool (% of MTR)**	**Concentration in a large lake (% of MTR)**	**Relationship between average deposition in Nl and highest value (factor)**	**Location with highest value**
atrazine	2900	0.8	0.1	0.03	0.00004	2.3	WY
simazine	140	0.3	0.05	0.2	0.0003	6.0	GP
terbutylazine	190	11.7	0.7	6	0.0039	4.4	GP
chlorfenvinfos	2	0.08	0.02	4	0.01	12.6	EG
dichlorvos	0.7	0.01	0.002	1	0.003	6.7	GP
isoproturon	320	40	4.8	13	0.01	1	
lindane	920	1.0	0.2	0.1	0.0002	3.1	WY
MCPA	2000	0.6	0.1	0.03	0.00005	2.0	YE
mecoprop		0.6	0.1	0.03		2.1	YE
propachlor	1300	34	3.6	3	0.003	5.6	KE
triphenyltin (fentin hydroxide)	5	0.7	0.1	14	0.03	1.9	VE
benzo[a]pyrene	500	0.2	0.1	0.04	0.02	1.5	KE/LE
fluorantene	500	18	8	3.5	1.5	12	EG

Widely differing values are found in the literature for the half-life values of *isoproturon, propachlor* and *simazine* in surface water. The calculations have been carried out with the highest half-life value. The MTR in surface water was not exceeded for any of the studied pesticides as result of the contribution of atmospheric deposition.

For *isoproturon, propachlor* and *terbutylazine,* concentrations in a pool were calculated of some (tens) ng/litre: 13, 3 and 6%, respectively, of the MTR of those substances.

The atmospheric deposition of the organophosphorous compounds *dichlorvos* and *chlorfenvinfos* leads to low calculated concentrations in surface water. Even in the most sensitive system (pool), the concentration is below the MTR. For *dichlorvos* and *chlorfenvinfos,* however, the target value for surface water is exceeded.

The table also shows the correlation between the annual average deposition used here and the highest annual average deposition that was found in the country, at a

monitoring station. This shows that the concentration of *terbutylazine, chlorfenvinvos, dichlorvos, propachlor* and *triphenyltin* could in some source areas as result of atmospheric deposition still rise to values between five and fifty per cent of the MTR value. Atmospheric deposition can, together with other sources, easily lead to exceedance of criterion values for these substances.
Besides the MTR level, the NR level is also important. This value is usually 100 times lower than the MTR value. The table shows that for a number of substances such as *terbutylazine, chlorfenvinfos, isoproturon, propachlor* and *triphenyltin*, the NR concentration in different types of surface water can be exceeded by the contribution of atmospheric deposition alone.

These calculated values are equilibrium concentrations as result of the annual load by deposition. The concentrations in surface water, however, may soon after a high load, e.g., in the month in which the pesticide is used, temporarily be much higher than the equilibrium concentration calculated with this model. The model that has been used is not suitable to calculate this increased concentration.
It should also be kept in mind, however, that the objective of study presented here was to obtain a nation-wide picture. This is the reason why all monitoring stations were chosen in such a way that they were located far from large sources. The deposition in the direct vicinity of these sources is probably still considerably higher. This could therefore easier lead to exceedance of criterion values. An example where the concentration at one km distance from a source is fifty times higher than at ten km distance from the source is elaborated in Annex V. It was also found that up to 20% of the emitted amounts can deposit within a radius of the first km from the source.

More information about the deposition to surface water in the vicinity of the source is given by Baas and Bakker (1996), who make realistic estimates of the deposition in the vicinity of greenhouse horticulture and the resulting concentration in surface water, soil and air. Depending on the replacement rate of the surface water, the concentration in surface water can rise to 500 ng/l at short distance from the greenhouse area.

4. Discussion and conclusions

4.1 Introduction

Model calculations and measurements can be used to study to what extent atmospheric deposition of (eco)toxic substances can burden surface water and terrestrial ecosystems. Model calculations are often a first step to establish whether the contribution is indeed significant, and who is responsible for it. Measurements form the second step to investigate whether a problem does really exist. Over the past fifteen years the IWAD followed the line from preliminary calculations of the deposition to a complete monitoring network with the objective to derive the deposition from measurements. Over the last three years, the first systematic measurements of the deposition in the Netherlands have been carried out in the framework of IWAD. The collected monitoring data enable proper evaluation of the quality of model calculations. In this chapter we attempt to place the currently available results in a broader context, to identify white spots, and to indicate possibilities for improvement. The possibilities for monitoring atmospheric deposition in the future and for reducing atmospheric deposition in the future are discussed as well.

4.2 The results of the measurements

The measured atmospheric deposition can be evaluated in different ways. On the onehand, it can be investigated whether effects of the observed *levels* are to be expected. This more concerns the effects at the short term than those at the long term. On the other hand, the *load* in kg/year is an important variable for water quality control bodies. Both sides of the monitoring results will be discussed.

Levels
The observed concentration levels have been compared with the MTR values for surface water in view of the fact that criteria for precipitation or air are lacking. The concentration of seventeen different pesticides and different PAHs did one or more times exceed the MTR level. The concentration of twenty two different pesticides and a number PAHs exceeded the drinking water criterion of 100 ng/litre. It is not clear to what extent this input of the surface water does actually lead to effects. The average concentration of substances in surface water is usually lower than in precipitation as result of various processes such as degradation and adsorption to sediment.
Calculations with a simple computer model for some substances, which simulates processes in water, show that the average deposition levels will not soon lead to continuous exceedance of the MTR level in surface water. The situation is different for a number pesticides, of which the total annual dose is sometimes applied in

relatively short periods. The peak load found in (source) areas during some months, could, however, lead to exceedance of criteria in some aquatic systems[1].
The concentration as result of atmospheric deposition can become higher at locations close to large sources or source areas. This was already shown in the study by Baas and Bakker (1996) into the effect of emissions from greenhouse horticulture. As stated above, the current study is aimed at the *national* scale. All monitoring locations are situated at the largest possible distance from sources. The concentration may be much higher closer to sources. In an elaborated example (Annex V), the concentration at one km distance from a source was fifty times higher that at ten km distance from the source.

Comparison with the input by other sources
On a national scale, the input of pesticides to surface water is similar to the input by drift and lateral leaching. The measurements show that on average 0.1 % of the amount of used active substance gets into the surface water by atmospheric deposition (see Annex IX), while the input via the other routes that are mentioned is 0.22%. For some substances, such as *propachlor* and *vinclozolin*, deposition to water, with about one per cent of the use, can even be regarded as high.
The estimated input to soil is very uncertain. The deposition on soil derived from the measurements would roughly equal 0.2 tot 0.3% of the use.
There are, however, important differences between the effect of atmospheric input and, e.g., drift. The atmosphere is only source for remote nature areas (terrestrial and non-terrestrial)! This means that possible effects in these areas are exclusively caused by atmospheric deposition from sources further away. This does in some cases even concern sources in surrounding countries. Another difference is that the input by drift lands on a relatively small water surface close to the source. The atmospheric deposition as derived from the measurements, is deposited on the total water surface of the Netherlands. Drift leads to high concentrations in a small area around the source. The large-scale atmospheric deposition measured here leads to a, relatively low, concentration in all waters in the Netherlands. The total amount of material that reaches the surface water in the Netherlands, however, is of the same order of magnitude for both routes.
Table 12 and Table 13 present a review of the deposition of a number of substances and substance groups to the IJsselmeer and to the Netherlands surface water. These data originate from the Annual Reports of the Hoofdinspectie Milieuhygiëne (Main Inspectorate Environmental Hygiene) (Koch et al., 2001, Van Harmelen et al., 2001).
For all substances, the measured input by atmospheric deposition is of the same order of magnitude as the direct input and the input via effluents. For *HCB*, *PCBs* and *pentachlorophenol*, the atmospheric route is by far the most important route.

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[1] The ALTERRA research bureau is on the basis of the monitoring data presented here, in cooperation with RIVM, investigating to what extent atmospheric deposition leads to effects in surface water. The results of this study become available at the end of the year 2002.

Table 12 Input to the IJsselmeer area from different sources in the year 1998 in kg/ year (Koch et al., 2001 indicated by [1)]) compared with measured deposition.

	Direct[1)] (1998)	**Atmospheric deposition[1)]**	**Effluents[1)]**	**Atmospheric deposition (measurements)**	
				2000	**2001**
Benzo[a]pyrene	81.9	124	0.16	48	18
Fluorantene	219	2120	0.936	560	419
PAHs (6 of Borneff)	407	2250	1.35	880	558
HCB	0.00024	-	-	2	1
PCB (sum of 10)	0	-	0.00000635	8.1	5.5
PCP	0.00143	-	0.698	26	23

Table 13 Input to surface water of the Netherlands from different sources in the year 1998 in kg year (van Harmelen et al., 2001 [2)]) compared with the measured deposition to the Netherlands in total (area 5421 km^2, this is without the Waddenzee).

	Direct[2)] (1999)	**Atmospheric deposition[2)]**	**Effluents[2)]**	**Atmospheric deposition (measurements)**	
				2000	**2001**
Benzo[a]pyrene	417	273	15.6	116	58
Fluorantene	1700	2920	89.8	1671	1378
PAHs (6 of Borneff)	2890	3200	187	2426	1836
HCB	0.266	-	0.154	4	4
PCB (sum of 10)	0	-	0.00075	30	13
PCP	1.26	-	63.3	74	54

Part of the deposition on soil can also reach the surface water via leaching or run-off. Estimates of the contribution of this *indirect deposition* via leaching and runoff from soil are relatively uncertain. The contribution of this indirect deposition has not been taken into account in the estimates presented here. The contribution of indirect deposition is according to a first estimate (for the water management area of ZHEW) of the same order of magnitude as direct atmospheric deposition. This means that the observed deposition could be doubled by the contribution of indirect deposition.

Comparison with other estimates

Calculations of the input to surface water by atmospheric deposition have also been carried out in the context of the recently conducted evaluation of the MJP-G (Multi-Year Crop Protection Plan) in which calculations were carried out for all substances used in the Netherlands. Moreover, the emissions were according to a deviating procedure spread over the country. Averaged over all substances, 0.01% of the used amount of active substance would get into the surface water of the Netherlands via atmospheric deposition. This is a much lower value than now found in the measurements presented here. The physical-chemical properties used in the MJP-G calculations are possibly not representative of the product package in

the current study. This probably explains the difference between the current results and the results from the MJP-G evaluation.
In the estimates given above from the reports of the Main Inspectorate for Environmental Hygiene, the Emission Monitor presents deposition estimates for some substances. The estimated atmospheric deposition on the surface water of the Netherlands presented there is usually a factor three to four higher than the deposition derived from the measurements for some PAHs.

Developments in the deposition in recent years and the authorisation policy
For many substances, the now measured deposition seems to be considerably lower than the deposition estimated 10 years ago. The deposition derived from the measurements is usually lower than the atmospheric deposition on the Netherlands calculated with dispersion models for the year 1990. For PAHs and PCBs, the current deposition is a factor four and a factor two, respectively, lower than estimated for the year 1990. The depositions of *endosulfan*, *lindane* and *trifluralin*, however, differ by a factor 10 to 50. Also striking are the low values for the deposition of *parathion-ethyl*, *mevinphos, pentachlorophenol, simazine, bentazon* and *atrazine*.
Duyzer and Vonk (2001) extensively deal with these differences. One of the causes of this difference is the strongly reduced use of precisely those substances that received a lot of attention in the first study. The use of a number of these substances has, as result of policy measures, decreased. Other substances are now used instead. The MJP-G evaluation also mentions a decrease of the total volume of active substance over the period under consideration. It is not quite clear to what extent this decrease is also indicative for the substances studied here. Duyzer and Vonk (2001) already illustrated that this decrease is also relevant for substances such as *azinphos-methyl, bentazon, parathion, lindane* and *simazine*. The authorisation policy clearly also has a strong influence on the presence of substances in precipitation. The observed decrease in the concentration of *dichlorvos* in the precipitation collected in Zuid Holland is an illustration of this. This decrease since 1990 runs parallel with the decrease in the presence of this substance in surface water (Duyzer et al., 2001).
Despite this trend, unauthorised products are still frequently found. These are substances such as *atrazine, endosulfan, propachlor, trifluralin* and *mevinphos*. All these substances were in 2001 found in precipitation. For *atrazine*, which is no longer authorised in the Netherlands from the year 2000*, trifluralin* and *lindane,* sources from surrounding countries also seem to make an important contribution.

4.3 Comparison with model calculations

For twenty substances, which were regularly found, calculations have also been carried out with the OPS model on the basis of estimates of the emission to air. For over 40% of these substances, the calculated and the measured wet deposition differed less than a factor two and there was a good correlation between measured and calculated deposition. This indicates that the estimates that have been made of the volume of the emission and the distribution over the country and the calculations of the dispersion through the atmosphere are reasonable. For some substances (almost 40%), the distribution over the country was predicted properly but the emission or transformation processes in the air were either overestimated or underestimated. This resulted in a strong overestimation or underestimation of the average deposition. The agreement between measured and calculated wet deposition differed up to a factor ten for *dichlobenil*, *DNOC* and *triallate*. The conclusion seems justified that, in principle, a good estimate can be obtained using the model, i.e. within a factor two, of the concentration and deposition of substances, for which it is in any case necessary that good emission estimates are available.

The calculated deposition of *fluorantene* (a gaseous PAH) and of *benzo[a]pyrene* did also differ considerably from the measured deposition. The model underestimated the measured deposition of *benzo[a]pyrene* by a factor three. The model overestimates the wet deposition of *fluorantene* by a factor two.
Calculations of the input to surface water by atmospheric deposition have been carried out in the context of the recently conducted evaluation of the MJP-G,
in which calculations were carried out for all substances used in the Netherlands. Moreover, the emissions were according to a deviating procedure spread over the country. Averaged over all substances, 0.01% of the used amount of active substance would get into the surface water of the Netherlands via atmospheric deposition. This is a factor of about ten lower than now found in the measurements presented here. The physical-chemical properties used in the MJP-G calculations are possibly not representative of the product package in the current study. This probably explains the difference between the current results and the results from the MJP-G evaluation.

Source of the deposition
Model calculations make it possible to indicate which sources contribute to which extent to the deposition. Based on the detailed database of emissions that was available for the current study, a calculation has been made of the contribution of the various source areas in the Netherlands to the deposition. An analysis of the contribution of different source areas to the deposition in different water management areas shows that this contribution is strongly associated with the situation of the source in relation to the water quality control area in question. Roughly, two patterns can be recognised. The highest deposition of *dichlorvos*, a substance used in greenhouses, is found in Delfland, an area with many greenhouses. For the Netherlands as a whole, the emission from this area is the most important source and leads to exceedances of the MTR for surface water in precipitation. The deposition of *chlorothalonil*, a substance used a lot in potatoes, shows a completely different pattern over the country. Also for this substance, the deposition is highest in the specific potato areas in Groningen and Zeeland. In most cases, however, the largest contribution comes from the own province.
The good fit between the calculated and measured concentration of *atrazine* is another indication that the deposition of this substance, not authorised in the Nether-

lands, is caused by emissions from surrounding countries. In the calculations it was assumed that the substance is not used in the Netherlands.

4.4 Recommendation for follow-up

This section deals with the need and objective of follow-up research and the possibilities for incorporation of the research results into the policy aimed at improvement of the quality of surface water.

4.4.1 Recommendations for further research

The deposition values derived here originate from measurements at eighteen stations spread over the country. This yields a representative picture of the deposition on the Netherlands. The use of measurements makes the quality of the deposition estimates relatively high in comparison with the estimates, made in the past, on the basis of model calculations. The uncertainty in the calculated deposition to surface water was estimated at a factor 5-10. Deposition has in this study now for the first time also been determined on the basis of measurements. A comparison between measurements and model calculations shows that the model can often estimate wet deposition within a factor two. This seems to justify the conclusion that the current methods enable a good determination of the deposition. Improvement of the quality can, in as far as the quality of the measurements is concerned still be reached by:

– Improvement of the sensitivity of the chemical analysis
  An improved accuracy can still be reached for a number of substances by increasing the sensitivity of the chemical analysis. For most substances, however, the accuracy is sufficient. The sensitivity for polar pesticides is too low.
– Increasing the knowledge about dry deposition
  An uncertain parameter is the value of the dry deposition rate to soil, vegetation and surface water. Dry deposition is now derived from measurements of the concentration in air and an estimate of the deposition rate. The uncertainty in this deposition rate is fairly large and there is a need for validation of the estimates. The attention for the input to terrestrial ecosystems from the atmosphere is, except for acidifying substances, very limited. This is one of the reasons for the fact that little research has been carried out into the underlying processes and that the knowledge is very limited.
– Increasing the knowledge about indirect deposition
  The significance of indirect deposition (runoff etc) has not been included in this study. The uncertainty in the significance of this process is very large while the contribution is possibly high. Depending on various factors, the runoff of deposition from hardened surfaces could be large.

- Improvement of the estimate of the deposition on the North Sea
  Deposition on the North Sea is now estimated on the basis of measurements close to the Dutch coast. This means that the course in concentration from coast to more open sea has not been taken into account. Improvement of these estimates is possible by conducting measurements at relevant locations. An extensive model study could also increase insight.

Apart from this, the quality of the model calculations of the deposition of pesticides can be increased by improvements in:

- Data about the emission of pesticides in surrounding countries. The estimates used here are mainly based on data about Dutch agriculture. Good data about the use in surrounding countries are lacking.

- The quality of physical-chemical parameters such as the Henry constant and vapour pressure. The uncertainty about the vapour pressure is especially large for substances with a low volatility. The knowledge about the transformation rate of substances in the atmosphere is very limited. This leads to large uncertainties about the contribution of the input by substances from sources from surrounding countries.

- As mentioned above, there is a need for knowledge about the process of dry deposition and indirect deposition.

A very large amount of data have been collected in the study presented here. The analysis of the data is very time-consuming and was aimed at main points. There is certainly room for a more detailed, scientific analysis of the monitoring data. Model calculations could of course play a role in an extensive analysis. A further analysis of the cause of the differences between measured and calculated depositions, as observed for some substances, could be a starting point for this activity.

4.4.2 Future monitoring of atmospheric deposition of organic compounds

The volume of atmospheric deposition as diffuse source for surface water as established in this study seems so large that it may be expected that the need for data about the significance of this process will continue to exist. The comparison between the results of model calculations and measurements also seem to justify the conclusion that suitable models are available. It also seems to be technically quite well possible to determine the deposition at a national scale by measurements. The costs of the monitoring programme do not seem to be unreasonable.

Measurements as well as model calculations are therefore available for the development of a strategy aimed at monitoring deposition in the years ahead. The concrete objective and the possible costs in relation to available budgets are in particular relevant for the development of such a strategy. A combination of model calculations and measurements is the most likely option. A monitoring programme could then be aimed at mapping the distribution of the deposition over the country and at following of the developments in the range of pesticides. Model calculations could be used for regionally mapping the deposition in relation to emissions and its developments.

TNO developed a strategy aimed at the information requirement of water quality control bodies in a study that was recently carried out for RIZA (Duyzer *et al.*, 2002). Three lines can be distinguished, also for the information requirement at the national scale:

- Model calculations for the Netherlands as a whole
  It is proposed to carry out calculations with a dispersion model in a formal route on the basis of data about the emission to air. These data can be obtained on the basis of the Emission Registration, in which estimates of the emissions of pesticides could be included. The results of the calculations can also be included in the Emission Registration. The advantage of this formal approach is that the quality of the data is guaranteed and maintained. A resolution of the data on a 5 by 5 km scale enables each water quality control body to collect data.

- Measurement of the deposition at a national scale
  This route is in particular important for pesticides. Because the product range is continually changing, measurements are required to observe and monitor developments. Measurements can also play an important role in monitoring the effects of policy and for monitoring the use within and outside the Netherlands. Illegal use could also be detected. For obtaining a national picture, the efforts could be restricted to some monitoring stations. A good picture of the large differences in deposition over the Netherlands could probably be obtained by spreading five stations according to a dice configuration. This would enable mapping of the large differences between North/South/East and West, for which it is important that these stations monitor a wide range of substances with a high sensitivity as well as regular adaptation of the range to the developments. The input to specific regions could be estimated on the basis of model calculations based on detailed emission estimates. Model calculations can in any case yield a reasonable estimate.

- Measurement of deposition at a regional scale
  Single monitoring stations can be set up in case data are required on a regional scale, for which a specific product package can be specified. Connection with the national monitoring activities enables comparison of the results.

Other substances

The now completed research was especially aimed at organic compounds with the emphasis on persistent compounds and pesticides. Research by Baart et al. (1995) already showed that input from the atmosphere can also be important for other substances. Moreover, there are recent indications that the atmospheric deposition of zinc is higher than assumed earlier (Smolders, 2002). The input of organic compounds to surface water, not included in the current study, could also be important. Hamers (2002) demonstrated by means of bio-assays that precipitation can have oestrogenic potential. This potential could be related to the presence of *organochlor* pesticides. The significance of these research results is still subject of further study.

Relatively little is also known about the significance of atmospheric deposition of nutrients to surface water. The effect of atmospheric deposition of ammonia on remote waters such as pools has meanwhile been documented in detail (Heij and Schneider, 1991). It is suggested in a recently reported study that the input into estuaries and coastal waters of nutrients from the atmosphere may have a significant effect (Paerl, 2002). An influence on known effects such as algal bloom and ultimately even a loss of biodiversity has been suggested. It is for the time being not clear to what extent the results of this North American study are also significant for Northern Europe.

It seems meaningful to conduct - in the near future- an inventorial study into the significance of the deposition of metals, oestrogenically active compounds and nutrients for the quality of surface water.

4.4.3 Atmospheric deposition in the authorisation policy

The pesticides load of surface water in the Netherlands via atmospheric deposition appears to be of the same order of magnitude as the load via drift and lateral leaching. The contribution of indirect deposition can result in a considerable further increase of the input.

An important difference, however, is that all input by drift takes place on a relatively small water area whereas atmospheric deposition takes place all over the Netherlands. This means that input by drift will sooner lead to high concentrations and criterion exceedance. Atmospheric deposition causes a relatively low input into of large areas. This means that the average level will not easily lead to criterion exceedances in those areas. Measurements, however, show at the same time that in concentration areas, in periods in which the substances are used, deposition can be up to a factor ten higher. Although this has not been investigated in the current study, it is clear that still closer to the source, within some kilometres from sources or source areas, concentration and deposition could still be much higher.

Atmospheric deposition thus seems to be an important diffuse source for surface water. High loads in certain periods in source areas probably lead to exceedance of MTR levels. In view of this situation it seems meaningful to investigate how atmospheric deposition can be brought into the authorisation policy. There seem to be good possibilities to make the now available knowledge suitable as instrument in the authorisation evaluation.

A pilot study into the possibilities to include atmospheric deposition in the authorisation policy seems meaningful. It could be investigated to what extent physical-chemical parameters do in practice affect the transport over medium-range distances and which other aspects are relevant here. The study could be aimed at the medium-range distance from the more or less direct vicinity of the source to hundreds of kilometres. Deposition in the direct vicinity of the source has not been investigated in the current study. Deposition in the direct vicinity is probably much higher than at the current monitoring stations. The chance of exceedance of criterion concentrations and effects in the direct vicinity of sources, within some kilometres, is much higher than at a national scale. Furthermore, it seems useful to make calculations for a number of realistic situations.

Restriction of atmospheric deposition

The question is to what extent the deposition of crop protection products could be restricted. An extensive discussion of the possibilities is outside the scope of this study. A study into the possibilities has already been carried out by CLM (Centrum voor Landbouw en Milieu; Centre for Agriculture and Environment, Boland and Leendertse, 1999). Here, we therefore restrict ourselves to some remarks of a general nature. A first important fact is that deposition is virtually proportional to the emission to air. Furthermore, the model calculations show that the deposition in the different water quality areas can often be related to emissions in the area (within the province). This means that restrictions in the use and the emission in the Netherlands do directly lead to a reduction in deposition. Choosing substances with a shorter residence time in air, e.g. caused by a higher transformation rate, does also lead to a reduction of the deposition. The residence time in air could therefore be taken into account in the authorisation of pesticides. This, however, is a complex matter for which a large number of basic data are lacking. Moreover, the effect of a high transformation rate (i.e. a short residence time in air) will be rather restricted in the direct vicinity of the source, at a scale of tens of kilometres, where deposition is highest. The advantage of substances with a high transformation rate is more relevant at larger distances from the source.

Sources in surrounding countries play a significant role in as far as a number of substances are concerned that are no longer authorised in the Netherlands. The Dutch policy has no influence on the contribution of these sources. Here, the international authorisation policy would have to play a role.

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Date upon which, or period in which, the research took place:

January 1999 - October 2002

Signed:	Authorised by:
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Annex I Deposition rate

The dry deposition rate of substances strongly depends on meteorological conditions and the surface on which this deposition takes place. Exchange to the surface from the atmosphere first takes place via turbulent movements, and the exchange close to the surface is regulated by molecular diffusion through a laminar layer on top of this surface. This process is limiting in the exchange to the surface, and is different for water and soil.

Deposition to surface water

The solubility of a substance is an important factor for deposition on water. Substances with a high solubility deposit easily on water surfaces. Absorption in water is then virtually only limited by the resistance in the air layer above that water. Deposition rates are then of the order of 0.4 cm per second. A poor solubility of substances limits the absorption in water and decreases the deposition rate. A substance such as chlorothalonil has a poor solubility; about 0.6 milligram of the substance can dissolve per litre water. The Henry coefficient (H) of that substance, which can be approached by the ratio between the vapour pressure (P_l) of that substance and its solubility (S), is high. The calculated deposition rate (V_d) of chlorothalonil to the water surface is in the order of 0.1 cm per second. This deposition rate has been calculated according to Liss and Slater (1974):

$$Flux = V_d\,(C_l - \frac{H}{RT} C_w)$$

where

$$V_d = \frac{1}{\frac{1}{k_a} + \frac{H}{RT}\frac{1}{k_l}}$$

where k_a is the exchange coefficient to air, k_l the exchange coefficient to water, R the specific gas constant and T the air temperature in degrees Kelvin.

Re-emission

The equation shows how the Liss and Slater model takes the concentration of a substance in surface water (C_w) into account. In some cases, when the concentration in the surface water is high, the direction of the flux may invert and (re-)emission from the water surface may even occur. This may concern substances that got into the surface water via precipitation or substances that got into the water in a different way.

To investigate the significance of this process, samples of surface water, grass and soil were in December 2000 taken in the context of this study; these were analysed by means of the same analytical method as used for the precipitation samples

(Beeltje, 2002). The deposition rate to water surfaces for those substances has been adjusted for the concentration in water (Table I.1). This shows that, at the found concentrations in surface water, a number of substances will probably sooner evaporate from the surface water than that they will be absorbed by dry deposition. A good assessment of the significance of this adjustment for the Netherlands as a whole is only possible on the basis of good insight into the concentration of substances in surface water. This insight is currently not available. The table illustrates only that for *2,4 D, atrazine* and *hexachlorobenzene* this effect may play a role. The data on the other substances are lacking.

Table I.1 Deposition rate (in cm/s) of a number of substances calculated on the basis of a negligible concentration of the substance in surface water (V_{d1}) and on the basis of the measured concentration (V_{d2}).

Substance	V_{d1}	V_{d2}
2,4-D	0.45	-210.74
atrazine	0.46	-5.16
azinphos-methyl	0.43	0.40
bentazon	0.45	0.40
dichlobenil	0.49	0.12
hexachlorobenzene	0.37	-51.29
MCPA	0.46	0.10
mecoprop	0.45	0.32
PCP	0.46	0.45
triallate	0.40	0.40

Deposition rate to soil and vegetation

The deposition rate to soil for the various substances has been calculated with the *SimpleBox* model (Van der Meent, 1993). This model calculates the partitioning of those substances over the components air, water, soil, and sediment. A substance may also occur partially adsorbed to aerosols and partially in the gaseous phase in the atmosphere; this depends on factors such as vapour pressure of a substance and actual temperature. The deposition rate of the aerosol-bound substances differs from the substance occurring in the gaseous phase. This is why we use the concept effective deposition rate, in which adsorption of a substance to particles has been taken into account. Important substance properties in this model are:

- K_{ow} octanol -water distribution coefficient: measure of the affinity of a substance for fat and organic material
- solubility in water
- vapour pressure
- degradation rate of the substance in air, water, soil, and sediment.

Dry and wet deposition have been calculated for those substances that were frequently found in precipitation and air samples. The deposition rate of these substances and a number of physical substance properties are presented in Table I.2.

Table I.2 Dry deposition rate of pesticides to water and soil, calculated according to Liss and Slater (1974) and with the SIMPLEBOX model, respectively.

Substance name	MW	$\log K_{ow}$	P	S	Trans-formation in air	Vd water	Vd soil
	g/mol		Pa	mg/L	%/hr	cm/s	cm/s
atrazine	215.7	2.6	0.00004	30	57	0.46	0.244
bentazon	240	2.3	4.6E-04	500	24	0.48	0.279
chlorpropham	214	3.4	1.3E-03	89	15	0.46	0.068
chlorpyriphosmethyl	322.5	4.7	5.6E-03	4	23	0.44	0.005
chlorothalonil	265.9	3.1	8.1E-03	1	3.7	0.12	0.004
dichlobenil	172.0	2.7	7.3E-02	18	0.07	0.52	0.006
dichlorvos	221	1.9	1.6E+00	10000	0.12	0.51	0.014
dimethoate	229.3	0.7	1.1E-03	25000	31	0.49	0.303
DNOC	198	2.4	8.7E-03	130	0.08	0.51	0.025
ethofumesate	286	2.7	8.6E-05	80	21	0.46	0.145
fluazinam	465	5.7	5.1E-06	0.4	0.03	0.40	0.006
hexachlorobenzene	283.8	5.7	1.5E-03	0	0.01	0.39	0.025
PCP	266.3	5.1	5.0E-02	80	0.21	0.48	0.007
pentachlorobenzene	250.3	5.2	1.3E-01	1	0.02	0.12	0.001
pirimiphos-methyl	305	1.2	1.5E-02	305	62	0.45	0.022
procymidon	284.1	3.1	1.8E-02	5	3	0.48	0.005
propachlor	212	1.8	3.1E-02	613	8.7	0.47	0.030
terbuthylazine	229.7	3.2	1.5E-04	9	3.7	0.49	0.060
tolclofosmethyl	301.1	4.6	5.7E-02	0	24	0.12	0.001
triallate	304.7	4.3	1.6E-02	4	13	0.42	0.004
trifluralin	335.5	5.3	1.4E-02	1	9.	0.44	0.003
vinclozolin	286.1	3.0	1.6E-05	1000	12.86	0.48	0.134

Annex II Concentrations in precipitation and in air

The pesticide, PAH and PCB concentrations as found in precipitation and air samples have been summarised in this Annex. The annual average concentration over all samples at all monitoring locations, the percentage of the samples in which the substance was found, and the average concentration of only those samples in which the substances was found, are presented. The data for the years 2000 and 2001 are presented in separate columns.

Concentrations in air

Table II.1 Presence of pesticides in air samples in the Netherlands for the years 2000 and 2001. The average of the samples in which the substance was found (- analysis for this substance was no longer carried out, 0: analysis for the substance was carried out but the substance was not found) is given besides the average concentration over all samples .

Substance	**Found in air 2000 (%)**	**Found in air 2001 (%)**	**Average concentration 2000 (ng/m^3)**	**Average concentration 2001 (ng/m^3)**	**Average concentration > zero 2000 (ng/m^3)**	**Average concentration > zero 2001 (ng/m^3)**
2,4-D	0	0	0.00	0.00	0.00	0.00
aldrin	0	-	0.00	-	0.00	-
atrazine	3	1	0.001	0.0002	0.05	0.02
bentazon	1	11	0.002	0.01	0.15	0.13
bitertanol	3	-	0.13	-	4.43	-
captan	3	12	0.02	0.14	0.76	1.19
chlorfenvinfos	2	1	0.002	0.001	0.08	0.09
chlorpropham	48	46	0.85	0.41	1.76	0.91
chlorpyriphos-methyl	19	14	0.002	0.001	0.01	0.01
chlorothalonil[1]	51	31	0.38	0.24	0.74	0.77
chloridazon (pyrazon)	2	-	0.003	-	0.17	-
deltamethrin	0	-	0	-	0.00	-
demeton-S-methyl	11	-	0.02	-	0.18	-
diazinon	1	0	0.001	0.00	0.06	0.00
dichlobenil	99	96	1.57	0.99	1.59	1.04
dichlorvos	1	6	0.021	0.02	2.37	0.41
dicofol	0	-	0.00	-	0.00	-
dimethoate	2	6	0.001	0.01	0.03	0.11
disulfoton	0	-	0.00	-	0.00	-
DNOC	28	37	0.37	0.35	1.33	1.0
endosulfan I	1	2	0.003	0.01	0.21	0.29
endosulfan II	0.4	1	0.002	0.002	0.45	0.25
endrin	6	-	0.01	-	0.15	-
epoxiconazole	6	-	0.005	-	0.08	-
ethofumesate[1]	8	25	0.001	0.06	0.01	0.24
parathion-ethyl	0.4	1	0.000	0.0004	0.01	0.04
fenitrothion	0	-	0.00	-	0.00	-

Substance	Found in air 2000 (%)	Found in air 2001 (%)	Average concentra-tion 2000 (ng/m^3)	Average concentra-tion 2001 (ng/m^3)	Average concentra-tion > zero 2000 (ng/m^3)	Average concentra-tion > zero 2001 (ng/m^3)
fenthion	0	-	0.00	-	0.00	-
fluazinam	28	23	0.04	0.03	0.13	0.14
fluroxypyr[2]	11	13	0.05	0.04	0.42	0.29
phosphamidon	0	-	0.00	-	0.00	-
heptachlor	0	0.4	0.00	0.002	0.00	0.47
heptachlor epoxide	51	-	0.01	-	0.01	-
heptenophos	0	-	0.00	-	0.00	-
hexachlor-1.3-butadiene	6	-	0.002	-	0.04	-
hexachlorobenzene	61	30	0.003	0.003	0.004	0.01
isoxaflutole	6	-	0.02	-	0.28	-
kresoxim-methyl[1]	19	15	0.01	0.01	0.06	0.06
lindane	12	3	0.04	0.01	0.34	0.36
malathion	0.4	-	0.000	-	0.03	-
MCPA	3	1	0.001	0.002	0.05	0.21
mecoprop	1	2	0.000	0.003	0.01	0.12
metamitron	0	-	0.00	-	0.00	-
methiocarb	0	1	0.00	0.001	0.00	0.1
methomyl	0	-	0.00	-	0.00	-
parathion-methyl	0	-	0.00	-	0.00	-
metolachlor	6	4	0.02	0.003	0.30	0.07
mevinphos	8	4	0.004	0.01	0.06	0.15
o.p'-DDD[1]	5	2	0.01	0.0004	0.12	0.02
o.p'-DDE	6	-	0.004	-	0.07	-
p.p'-DDT[1]	0	0 (0.5)	0.00	0 (0.0002)	0.00	0 (0.04)
PCP	84	74	0.06	0.05	0.08	0.06
pentachlorobenzene[2]	8	21	0.002	0.001	0.03	0.01
pirimicarb	4	2	0.001	0.001	0.02	0.05
pirimiphos-methyl	-	12	-	0.94	-	7.89
prochloraz	0.4	0.5	0.000	0.001	0.01	0.18
procymidon	40	12	0.03	0.01	0.08	0.10
propachlor	30	21	0.40	0.18	1.33	0.87
propoxur	0.4	0	0.001	0.00	0.13	0.00
pyrazophos	8	2	0.01	0.001	0.09	0.04
simazine	3	2	0.01	0.01	0.23	0.28
telodrin	9	-	0.001	-	0.02	-
terbutylazine[1]	3	8	0.001	0.057	0.03	0.75
tetrabromobisphenol A[1]	41	31	0.0001	0.002	0.0001	0.002
tolclofos-methyl	16	18	0.08	0.06	0.50	0.36
triadimenol	0	-	0.00	-	0.00	-
triallate	75	58	0.4	0.21	0.58	0.36
triazophos	3	-	0.002	-	0.07	-
trifluralin	82	91	0.17	0.25	0.21	0.27
vinclozolin[2]	59	42	0.11	0.23	0.19	0.55

[1] Substances determined at four locations in 2000 and at all locations in 2001.

- Substances no longer determined in the year 2001.

Table II.2 Presence of PAHs in air samples in the Netherlands for the years 2000 and 2001. The average of only those samples in which the substance was found is given besides the average concentration over all samples.

Substance	**Found in air 2000 (%)**	**Found in air 2001 (%)**	**Average concentration 2000 (ng/m^3)**	**Average concentration 2001 (ng/m^3)**	**Average concentration > zero 2000 (ng/m^3)**	**Average concentration > zero 2001 (ng/m^3)**
acenaphtene	100	96	2.44	1.65	2.44	1.73
acenaphtylene	59	52	0.14	0.12	0.23	0.24
anthracene	95	64	0.31	0.10	0.33	0.16
benzo[a]anthracene	78	32	0.03	0.01	0.04	0.03
benzo[a]pyrene	84	74	0.04	0.03	0.05	0.04
benzo[b]fluorantene	90	91	0.12	0.12	0.13	0.13
benzo[g.h.i]perylene	94	86	0.04	0.04	0.04	0.05
benzo[k]fluorantene	90	92	0.07	0.08	0.08	0.09
chrysene	80	34	0.14	0.04	0.17	0.11
dibenz[a.h]-anthracene	75	42	0.004	0.01	0.01	0.01
fenantrene	100	98	6.14	5.33	6.14	5.46
fluorantene	98	98	1.62	1.46	1.65	1.49
fluorene	100	97	3.86	3.00	3.86	3.10
indeno[1.2.3-cd]pyrene	88	76	0.03	0.04	0.04	0.06
naphthalene	100	99	12.05	11.62	12.05	11.75
pyrene	95	93	0.77	0.71	0.81	0.76

Table II.3 Presence of PCBs in air samples in the Netherlands for the years 2000 and 2001. The average of only those samples in which the substance was found is given besides the average concentration over all samples (0: analysis for the substance was carried out, but was not found).

Substance	**Found in air 2000 (%)**	**Found in air 2001 (%)**	**Average concentration 2000 (ng/m^3)**	**Average concentration 2001 (ng/m^3)**	**Average concentration > zero 2000 (ng/m^3)**	**Average concentration > zero 2001 (ng/m^3)**
PCB-101	51	27	0.0024	0.004	0.005	0.014
PCB-118	4	6	0.0001	0.0003	0.002	0.005
PCB-138	18	3	0.0004	0.0001	0.002	0.004
PCB-153	28	3	0.0005	0.00003	0.002	0.001
PCB-180	11	0	0.0003	0	0.003	0.000
PCB-20	18	16	0.0070	0.004	0.038	0.023
PCB-28	18	9	0.0052	0.002	0.028	0.022
PCB-35	2	3	0.0004	0.001	0.023	0.029
PCB-52	37	29	0.0047	0.006	0.013	0.021
PCB-8	32	12	0.0044	0.003	0.013	0.021

Concentrations in precipitation

Table II.4 *Presence of pesticides in precipitation samples in the Netherlands for the years 2000 and 2001. The average of only those samples in which the substance was found (- analysis for this substance was no longer carried out, 0: analysis for the substance was carried out but the substance was not found) is given besides the average concentration over all samples .*

Substance	**Found in precipitation 2000 (%)**	**Found in precipitation 2001 (%)**	**Average concentration 2000 (ng/l)**	**Average concentration 2001 (ng/l)**	**Average concentration > zero 2000 (ng/l)**	**Average concentration > zero 2001 (ng/l)**
2.4-D	9	31	0.8	1.9	8.9	6.2
aldrin	0	-	0.0	-	0.0	-
atrazine	23	25	12.1	16.5	52.4	65.9
bentazon	5	11	0.4	0.6	7.5	5.4
bitertanol	0	-	0.0	-	0.0	-
captan	12	5	4.8	9.0	41.0	171.4
chlorfenvinfos	1	1	0.1	0.2	9.4	26.5
chlorpropham	90	85	116.7	59.6	130.3	70.4
chlorpyriphos-methyl	43	38	0.4	0.2	0.9	0.4
chlorothalonil[1]	69	54	20.0	14.8	28.9	27.5
chloridazon (pyrazon)	2	-	0.4	-	23.0	-
deltamethrin	0	-	0.0	-	0.0	-
demeton-S-methyl	0	-	0.0	-	0.0	-
diazinon	3	4	0.5	0.3	18.0	7.2
dichlobenil	99	89	15.9	10.6	16.0	12.0
dichlorvos	21	29	3.8	3.4	18.4	12.0
dicofol	0	-	0.0	-	0.0	-
dimethoate	4	0.4	2.8	0.1	70.7	18.9
disulfoton	0	-	0.0	-	0.0	-
DNOC	99	98	1061.1	462.6	1075.1	470.9
endosulfan I	0.4	0.4	0.1	0.2	20.2	49.0
endosulfan II	5	0.4	1.5	0.0	31.6	9.0
endrin	0	-	0.0	-	0.0	-
epoxiconazole	11	-	3.7	-	34.9	-
ethofumesate[1]	18	21	6.0	9.3	33.2	43.9
parathion-ethyl	2	0	0.2	0.0	7.5	0.0
fenitrothion	1	-	0.0	-	3.5	-
fenthion	3	-	0.0	-	1.0	-
fluazinam	45	35	8.9	15.1	19.8	43.6
fluroxypyr[1]	13	0 (2)	4.5	0 (0.2)	35.2	0 (9.7)
phosphamidon	0	-	0.0	-	0.0	-
heptachlor	0	0	0.0	0.0	0.0	0.0
heptachlor epoxide	0	-	0.0	-	0.0	-
heptenophos	0	-	0.0	-	0.0	-
hexachlor-1.3-butadiene	3	-	0.0	-	0.3	-
hexachlorobenzene	97	71	0.4	0.3	0.4	0.4
isoxaflutole	3	-	0.4	-	14.2	-
kresoxim-methyl[1]	31	44	6.3	8.3	20.3	18.8

Substance	Found in precipi-tation 2000 (%)	Found in precipi-tation 2001 (%)	Average concentra-tion 2000 (ng/l)	Average concentra-tion 2001 (ng/l)	Average concentra-tion > zero 2000 (ng/l)	Average concentra-tion > zero 2001 (ng/l)
lindane	27	31	14.3	10.9	53.1	35.4
malathion	0.4	-	0.0	-	2.7	-
MCPA	64	58	9.7	6.1	15.1	10.5
mecoprop	47	36	7.3	5.6	15.7	15.9
metamitron	5	-	0.3	-	5.5	-
methiocarb	2	10	0.0	3.2	1.7	32.8
methomyl	0	-	0.0	-	0.0	-
parathion-methyl	1	-	0.0	-	2.5	-
metolachlor	24	18	8.8	2.8	36.9	15.2
mevinphos	2	4	0.2	0.1	9.3	3.4
o.p'-DDD[1]	46	10	3.2	0.28	7.0	3.0
o.p'-DDE	0	-	0.0	-	0.0	-
p.p'-DDT[1]	0	0 (1)	0.0	0 (0.1)	0.0	0 (10.8)
PCP	99	97	7.9	4.4	8.0	4.5
pentachlorobenzene[1]	82	79	0.2	0.2	0.3	0.3
pirimicarb	1	5	0.1	0.3	6.6	5.5
pirimiphos-methyl	-	4	-	10.3	-	293.9
prochloraz	1	1	1.2	1.1	88.4	86.4
procymidon	67	56	8.5	4.0	12.6	7.1
propachlor	50	50	103.9	68.4	205.9	138.0
propoxur	14	11	1.4	0.6	10.1	5.8
pyrazophos	3	0	1.5	0.0	50.4	0.0
simazine	3	1	0.8	0.5	29.8	40.7
telodrin	0	-	0.0	-	0.0	-
terbutylazine[1]	36	39	9.9	16.0	27.5	41.7
tetrabromobisphenol A[1]	41	69	0.2	2.9	0.4	4.1
tolclofos-methyl	8	9	2.2	2.2	28.7	25.2
triadimenol	0	-	0.0	-	0.0	-
triallate	77	58	20.8	4.4	26.8	7.5
triazophos	0	-	0.0	-	0.0	-
trifluralin	73	76	2.2	3.7	3.0	4.8
vinchlozolin[1]	85	75	17.2	15.6	20.4	20.8

[1] Substances determined at four locations in 2000 and at all locations in 2001.

\- Substances no longer determined in the year 2001.

Table II.5 The presence of PAHs in precipitation samples in the Netherlands for the years 2000 and 2001. The average of only those samples in which the substance was found is given besides the average concentration over all samples.

Substance	**Found in precipitation 2000 (%)**	**Found in precipitation 2001 (%)**	**Average concentration 2000 (ng/l)**	**Average concentration 2001 (ng/l)**	**Average concentration > zero 2000 (ng/l)**	**Average concentration > zero 2001 (ng/l)**
acenaphtene	96	95	8.4	5.2	8.7	5.5
acenaphtylene	26	48	2.9	3.0	11.2	6.4
anthracene	100	94	9.3	6.3	9.3	6.7
benzo[a]anthracene	99	97	11.6	9.7	11.7	10.0
benzo[a]pyrene	91	94	19.4	12.7	21.3	13.4
benzo[b]fluorantene	100	99	39.3	25.3	39.3	25.6
benzo[g.h.i]perylene	97	97	16.8	11.2	17.3	11.6
benzo[k]fluorantene	100	99	25.0	18.7	25.0	18.9
chrysene	100	100	35.3	26.2	35.3	26.2
dibenz[a.h]-anthracene	74	91	2.9	3.4	3.9	3.7
fenantrene	100	100	109.6	78.5	109.6	78.5
fluorantene	100	100	93.8	77.6	93.8	77.6
fluorene	98	99	17.5	12.2	17.8	12.4
indeno[1.2.3-cd]pyrene	92	95	15.7	10.6	17.1	11.1
naphthalene	100	100	60.3	40.5	60.3	40.5
pyrene	99	98	80.4	48.4	81.1	49.5

Table II.6 The presence of PCBs in precipitation samples in the Netherlands for the years 2000 and 2001. The average of only those samples in which the substance was found is given besides the average concentration over all samples.

Substance	**Found in precipitation 2000 (%)**	**Found in precipitation 2001 (%)**	**Average concentration 2000 (ng/l)**	**Average concentration 2001 (ng/l)**	**Average concentration > zero 2000 (ng/l)**	**Average concentration > zero 2001 (ng/l)**
PCB-101	58	19	1.4	0.2	2.3	1.2
PCB-118[1]	39	15	0.4	0.3	1.1	24.6
PCB-138	50	17	0.5	0.2	1.0	1.4
PCB-153	73	45	0.6	0.3	0.8	0.8
PCB-180	14	6	0.2	0.1	1.4	1.9
PCB-20	47	55	1.0	0.6	2.0	1.1
PCB-28	39	22	1.3	0.4	3.3	2.0
PCB-35	4	3	0.1	0.1	2.3	2.8
PCB-52	34	28	2.2	0.7	6.6	2.3
PCB-8	11	7	0.3	0.1	2.6	1.4

[1] The average concentration of PCB-118 was higher (3.6 ng/l), as result of a peak in the concentrations of 298 ng/l at station Ketelhaven, in December 2001. Because this is probably an outlier, this value has not been taken into account. The concentration at the Lelystad station was also increased, but to a lesser extent (10 ng/l).

Annex III Accuracy of the estimated atmospheric load

Accuracy of the estimated deposition
As stated before, it is not simple to determine representative variables from monitoring data. Some substances are not found above the limit of detection during part of the year, but they are found during the rest of the year (see Annex II). Examples of substances with such a concentration course are the herbicides *propachlor* and the fungicide *fluazinam.* In case the concentration of a substance is so low that it cannot be detected with the used measuring method, the substance can still be present in a sample. In case the concentration in this sample would be included as '0' in the averaging of the concentrations, this may possibly result in an underestimation of the calculated average concentration, and of the burdening by deposition derived from this. The significance of this possible artefact has been studied, where it has been assumed that the concentration has a value that equals one third of the reported limit of detection instead of the reported concentration *'0'*. The difference between the thus calculated average deposition or concentration indicates the magnitude of the effect of the sensitivity of the method. For some substances, which are found throughout the year, the effect of the limit of detection on the calculated deposition is small. Examples of such substances are *DNOC* and *dichlobenil*. For other substances the uncertainty is so large, that no reliable conclusion can be drawn. This applies for *aldrin, bitertanol, demeton-S-methyl, diazinon, endrin, fenthion, heptachlorepoxide, heptenophos, hexachlorbutadiene, malathion, o,p'-DDE, p,p-DDT, prochloraz, telodrin, triademol, triazophos*. Table III.1 shows the range found for all substances as affected by the limit of detection.

Table III.1 gives an estimate of the annual dry deposition of pesticides to soil and water for the years 2000 and 2001. Dry deposition as proportion of total deposition is given as well. Deposition of substances that are difficult to detect in precipitation or air could be underestimated in case these substances occur in concentrations that are lower than the limit of detection. To get an impression of the possible underestimation of the deposition, an upper deposition limit has been calculated by applying one third of the limit of detection in case the substances were not detected in samples. This deposition is presented in the column 'to' of the table.

Table III.1 *Annual average range in wet and total deposition of pesticides on Dutch inland waters, for the years 2000 and 2001 (kg/yr), calculated from an average over all monitoring locations.*

	Wet deposition				**Total deposition on Dutch surface water (kg/yr)**			
	2000		**2001**		**2000**		**2001**	
Substance	**from**	**to**	**from**	**to**	**from**	**to**	**from**	**to**
2.4-D	1.2	5.5	3.7	5.3	1.2	7.5	3.7	6.7
aldrin	0.0	19.7	-	-	0.0	27.0	-	-
atrazine	23.9	30.7	25.9	30.1	24.5	35.1	26.0	33.3
	-	-	-	-	-	-	-	-
bentazon	1.0	8.2	1.6	5.4	1.8	12.6	7.6	13.5
bitertanol	0.0	8.3	-	-	44.6	55.6	-	-
captan	9.3	37.0	12.8	129.2	17.8	59.8	66.8	243.2
chlorfenvinphos	0.3	24.9	0.2	6.5	0.9	35.3	0.7	9.7
chlorpropham	240.2	241.2	140.2	141.3	566.5	569.9	298.7	302.2
chlorpyriphos-methyl	0.7	2.5	0.4	2.2	1.5	4.1	0.7	3.1
chlorothalonil[1]	33.3	34.9	22.8	24.1	72.7	74.5	39.3	40.9
chloridazon (pyrazon)	0.6	21.2	-	-	0.6	30.9	-	-
	-	-	-	-	-	-	-	-
deltamethrin	0.0	15.6	-	-	0.0	21.6	-	-
demeton-S-methyl	0.0	9.2	-	-	7.4	19.7	-	-
diazinon	1.1	33.0	0.9	21.9	1.4	46.4	0.9	31.9
dichlobenil	30.9	30.9	23.2	23.5	704.2	704.3	448.0	448.4
dichlorvos	9.8	40.2	10.1	29.2	18.7	67.0	20.0	51.8
dicofol	0.0	54.4	-	-	0.0	73.5	-	-
dimethoate	6.1	55.0	0.2	77.8	6.4	78.7	2.8	119.9
disulfoton	0.0	16.8	-	-	0.0	23.1	-	-
DNOC	2254.8	2258.2	1087.3	1089.0	2412.3	2503.9	1237.4	1282.2
endosulfan I	0.1	71.8	0.6	45.8	1.0	97.9	2.2	65.4
endosulfan II	2.9	75.0	0.1	30.4	3.5	102.8	0.8	41.5
endrin	0.0	29.4	-	-	3.0	42.9	-	-
epoxiconazole	5.1	16.7	-	-	6.8	22.9	-	-
ethofumesate[2]	13.3	19.3	13.7	19.1	13.7	21.4	34.6	42.0
parathion-ethyl	0.3	10.0	0.0	7.1	0.3	14.2	0.1	9.9
fenitrothion	0.1	18.8	-	-	0	26.5	-	-
fenthion	0.1	1.3	-	-	0	1.7	-	-
fluazinam	20.0	39.5	43.1	73.0	31.8	60.8	54.5	98.2
fluroxypyr[1]	8.4	21.5	0.7	3.1	24.4	42.3	9.3	12.3
fosfamidon	0.0	43.1	-	-	0.0	60.4	-	-
heptachlor	0.0	22.2	0.0	12.2	0.0	31.2	0.7	18.6
heptachlor epoxide	0.0	3.2	-	-	2.3	6.0	-	-
heptenophos	0.0	26.4	-	-	0.0	36.3	-	-
hexachlor-1.3-butadiene	0.0	1.0	-	-	0.8	2.2	-	-
hexachlorbenzene	0.9	0.9	0.6	1.0	1.7	2.0	1.6	2.2
isoxaflutole	0.9	56.3	-	-	6.6	82.2	-	-
kresoxim-methyl[1]	10.3	13.9	13.3	15.3	14.8	20.0	16.3	19.4
lindane	24.1	53.4	18.3	39.1	39.8	84.8	22.6	57.6
malathion	0.0	29.3	-	-	0.1	41.2	-	-
MCPA	15.6	17.5	11.7	12.8	16.1	20.4	12.4	15.0

	Wet deposition				Total deposition on Dutch surface water (kg/yr)			
	2000		2001		2000		2001	
Substance	**from**	**to**	**from**	**to**	**from**	**to**	**from**	**to**
mecoprop	12.0	30.9	11.3	24.1	12.1	46.0	12.3	35.6
metamitron	1.2	8.4	-	-	1.2	11.4	-	-
	-	-	-	-	-	-	-	-
methiocarb	0.1	4.6	6.6	10.9	0.1	6.7	6.9	13.3
methomyl	0.0	175.2	-	-	0.0	254.0	-	-
parathion-methyl	0.1	23.5	-	-	0.1	33.0	-	-
metolachlor	17.8	21.3	4.3	7.4	24.2	29.5	5.4	10.2
mevinphos	0.4	18.2	0.3	11.3	2.2	28.1	3.0	19.9
o.p'-DDD2	6.2	7.7	1.1	2.6	8.5	10.9	1.2	3.4
o.p'-DDE	0.0	4.6	-	-	0.0	4.6	-	-
p.p'-DDT2	-	-	0.3	4.2	-	-	0.4	6.3
PCP	13.9	14.0	9.2	9.3	39.5	39.8	28.1	28.3
pentachlorbenzene[1]	0.4	0.5	0.5	0.8	0.6	0.7	0.6	0.9
pirimicarb	0.2	42.2	0.9	7.7	0.5	59.4	1.2	11.2
pirimiphos-methyl	0.0	49.9	17.0	19.6	3.9	72.4	370.6	373.9
prochloraz	2.1	33.2	2.6	26.2	2.1	45.6	2.8	34.9
procymidon	13.1	15.4	8.4	9.8	25.6	29.4	13.2	15.9
propachlor	161.0	169.0	127.8	132.7	320.8	333.7	199.8	209.0
propoxur	2.3	13.5	1.5	8.9	2.5	18.8	1.5	12.6
pyrazophos	2.2	12.0	0.0	7.0	4.5	18.0	0.3	9.9
simazine	1.4	24.7	1.1	19.7	3.9	38.5	3.6	31.7
telodrin	0.0	6.2	-	-	0.5	8.8	-	-
terbutylazine[1]	19.2	20.1	18.7	20.0	19.5	21.0	29.8	31.6
Tetrabromobisphenol A	0.0	0.0	7.1	7.4	0.1	0.1	7.8	8.2
tolclofos-methyl	7.8	21.3	5.8	14.5	15.7	30.7	12.2	21.9
triadimenol	0.0	11.7	-	-	0.0	15.7	-	-
triallate	54.6	56.4	8.2	9.3	207.3	210.0	82.0	84.0
triazophos	0.0	22.8	-	-	0.7	31.8	-	-
trifluralin	5.3	6.0	9.1	9.5	58.6	59.5	86.1	86.6
vinchlozolin[1]	31.3	31.6	30.1	31.9	76.5	77.4	109.6	112.7

[1] Substances determined at four locations in 2000 and at all locations in 2001.

\- Substances no longer determined in the year 2001.

Table III.2 Annual average range in wet deposition of PAHs for the years 2000 and 2001 ($\mu g/m^2/yr$).

Substance	Wet deposition 2000 (μg/m^2/yr)		Wet deposition 2001 (μg/m^2/yr)	
	from	to	from	to
acenaphtene	6.2	6.2	4.4	4.4
acenaphtylene	2.9	3.6	2.8	3.3
anthracene	8.5	8.5	5.7	5.8
benzo[a]anthracene	9.4	9.4	8.1	8.1
benzo[a]pyrene	14.2	14.3	9.3	9.3
benzo[b]fluorantene	31.8	31.8	20.7	20.7
benzo[g.h.i]perylene	13.8	13.8	9.3	9.3
benzo[k]fluorantene	19.3	19.3	15.2	15.2
chrysene	28.1	28.1	21.7	21.7
dibenz[a.h]-anthracene	2.2	2.2	3.0	3.0
fenantrene	89.5	89.5	67.8	67.8
fluorantene	78.3	78.3	72.2	72.2
fluorene	14.0	14.0	10.2	10.2
indeno[1.2.3-cd]pyrene	12.5	12.5	8.8	8.8
naphtalene	44.7	44.7	33.3	33.3
pyrene	58.2	58.2	45.4	45.4

Table III.3 Proportion of dry deposition in the total deposition to soil and water for the years 2000 and 2001.

Pesticides	Fraction dry deposition to water 2000	Fraction dry deposition to water 2001	Fraction dry deposition to soil 2000	Fraction dry deposition to soil 2001
2.4-D	0.00	0.00	0.00	0.00
atrazine	0.02	0.00	0.01	0.00
bentazon	0.45	0.78	0.34	0.69
captan	0.47	0.63	0.02	0.03
chlorfenvinphos	0.71	0.68	0.29	0.26
chlorpropham	0.58	0.53	0.18	0.15
chlorpyriphos-methyl	0.51	0.43	0.01	0.01
chlorothalonil	0.66	0.42	0.09	0.02
diazinon	0.22	0.00	0.01	0.00
dichlobenil	0.96	0.95	0.20	0.18
dichlorvos	0.47	0.49	0.02	0.03
dimethoate	0.03	0.93	0.02	0.89
DNOC	0.07	0.12	0.00	0.01
endosulfan I	0.87	0.76	0.42	0.26
endosulfan II	0.18	0.86	0.02	0.41
ethofumesate	0.03	0.61	0.01	0.35
ethyl-parathion	0.06	1.00	0.00	1.00
fluazinam	0.37	0.21	0.01	0.00
fluroxypyr	0.68	0.92	0.00	0.25
heptachlor	-	1.00	-	1.00
hexachlorobenzene	0.50	0.61	0.00	0.00
kresoxim-methyl	0.31	0.18	0.02	0.01

Pesticides	Fraction dry deposition to water 2000	Fraction dry deposition to water 2001	Fraction dry deposition to soil 2000	Fraction dry deposition to soil 2001
lindane	0.39	0.19	0.01	0.00
MCPA	0.03	0.07	0.03	0.06
mecoprop	0.00	0.09	0.00	0.06
methiocarb	0.00	0.05	0.00	0.00
metolachlor	0.26	0.17	0.11	0.07
mevinvos	0.82	0.87	0.00	0.85
o.p'-DDD	0.29	0.04	0.00	0.00
p.p'-DDT	-	0.20	-	0.00
PCP	0.64	0.64	0.03	0.03
pentachlorbenzene	0.34	0.20	0.01	0.00
pirimicarb	0.49	0.27	0.39	0.19
pirimiphos-methyl	1.00	0.95	1.00	0.53
prochloraz	0.01	0.12	0.00	0.00
procymidon	0.47	0.35	0.01	0.01
propachlor	0.49	0.36	0.06	0.04
propoxur	0.09	0.00	0.00	0.00
pyrazophos	0.54	1.00	0.00	1.00
simazine	0.63	0.71	0.32	0.40
terbutylazine	0.02	0.37	0.00	0.00
tetrabromobisphenol A	0.11	0.09	0.00	0.01
tolclofos-methyl	0.50	0.52	0.01	0.01
triallate	0.73	0.90	0.03	0.09
trifluralin	0.91	0.90	0.08	0.07
vinclozolin	0.68	0.72	0.46	0.44

PAH/PCB	Fraction dry deposition to water 2000	Fraction dry deposition to water 2001	Fraction dry deposition to soil 2000	Fraction dry deposition to soil 2001
acenaphtene	0.98	0.98	0.74	0.77
acenaphtylene	0.86	0.88	0.27	0.30
anthracene	0.89	0.76	0.25	0.11
benzo_a_anthracene	0.30	0.18	0.04	0.02
benzo_a_pyrene	0.33	0.41	-0.24	-0.38
benzo_b_fluorantene	0.38	0.52	0.00	0.00
benzo_g.h.i_perylene	0.30	0.47	0.41	0.58
benzo_k_fluorantene	0.40	0.51	0.38	0.48
chrysene	0.41	0.26	-0.79	-0.28
dibenz_a.h_-anthracene	0.21	0.27	0.01	0.02
fenantrene	0.90	0.93	0.36	0.46
fluorantene	0.75	0.81	0.44	0.50
fluorene	0.97	0.98	0.66	0.72
indeno_1.2.3-cd_pyrene	0.30	0.48	0.09	0.18
naphtalene	0.94	0.96	0.06	0.09
pyrene	0.67	0.77	0.08	0.12

PAH/PCB	**Fraction dry deposition to water 2000**	**Fraction dry deposition to water 2001**	**Fraction dry deposition to soil 2000**	**Fraction dry deposition to soil 2001**
PCB				
PCB-101	0.06	0.40	-0.002	-0.016
PCB-118	0.02	0.03	0.000	-0.001
PCB-138	0.03	0.01	-0.001	0.000
PCB-153	0.03	0.00	-0.001	0.000
PCB-180	0.03	0.00	-0.001	0.000
PCB-20	0.20	0.17	-0.007	-0.005
PCB-28	0.10	0.11	-0.003	-0.003
PCB-35	0.13	0.27	-0.004	-0.009
PCB-52	0.07	0.24	-0.002	-0.008
PCB-8	0.30	0.48	-0.012	-0.023

This table shows that dry deposition may represent a considerable part of total deposition. Above water surfaces, dry deposition of easily soluble substances has a large part in the total deposition.

Table III.4 Annual average wet deposition of PCB's for the years 2000 and 2001 (ug/m^2/jr).

Substance	**Wet deposition 2000 (µg/m^2/yr)**		**Wet deposition 2001 (µg/m^2/yr)**	
	from	**to**	**from**	**to**
PCB-101	0.9	1.0	0.2	0.4
PCB-118	0.3	0.4	4.4	4.5
PCB-138	0.3	0.4	0.2	0.4
PCB-153	0.4	0.4	0.3	0.4
PCB-180	0.1	0.3	0.1	0.3
PCB-20	0.6	0.9	0.5	0.7
PCB-28	0.9	1.5	0.4	1.2
PCB-35	0.1	0.6	0.1	0.6
PCB-52	1.4	1.6	0.6	0.8
PCB-8	0.2	0.9	0.1	0.9

Table III.5 Total deposition on the Netherlands (soil) and on the Netherlands surface waterr (kg/yr). The variation over the Nethelands is indicated by presenting the national minimum and the national maximum. The area used for the Netherlands is 36 783 km^2; the area used for surface water is 2 790 km^2. This is without IJsselmeer, Waddenzee, and Ooster- and Westerschelde. An Italic '0' indicates that the substance was not found.

	To Dutch soil				**To Dutch surface water**			
Total deposition (kg/yr)	**National minimum 2000**	**National maximum 2000**	**National minimum 2001**	**National maximum 2001**	**National minimum 2000**	**National maximum 2000**	**National minimum 2001**	**National maximum 2001**
2.4-D	0	3	1	7	0	35	10	95
atrazine	12	47	9	58	163	622	119	766
bentazon	0	9	0	18	0	87	0	199
captan	0	67	0	208	0	206	0	1688
chlorfenvinphos	0	6	0	8	0	47	0	38
chlorpropham	14	2738	40	1389	153	17805	470	8547
chlorpyriphos-methyl	0	5	0	2	0	28	-	-
chloridazon (pyrazon)	0	4	-	-	0	52	-	-
chlorothalonil	8	74	2	102	3	684	13	651
diazinon	0	6	0	8	0	84	0	111
dichlobenil	97	1454	171	827	159	1117	117	739
dichlorvos	0	173	0	129	0	382	0	349
dimethoate	0	32	0	13	0	422	0	126
DNOC	763	2997	345	1571	8433	38351	3694	17970
endosulfan I	0	8	0	11	0	33	0	129
endosulfan II	0	14	0	8	0	190	0	26
epoxiconazole	0	7	-	-	0	88	-	-
ethofumesate	0	20	5	115	0	266	33	764
parathion-ethyl	0	2	0	1	0	30	0	0
fenitrothion	0	1	-	-	0	10	-	-
fenthion	0	0.2	-	-	0	3	-	-
fluazinam	7	83	5	184	72	636	1	1653
fluroxypyr	0	22	0	22	0	286	0	82
hexachlor-1.3-butad.	0	0.04	-	-	0	0	-	-
hexachlorbenzene	1	5	0	7	5	35	4	22
isoxaflutole	0	3	-	-	0	35	-	-
kresoxim-methyl	0	20	3	28	0	202	-	-
lindane	11	103	0	53	143	539	0	598
malathion	0	1	-	-	0	12	-	-
MCPA	1	30	5	26	12	401	72	344
mecoprop	2	26	3	25	28	343	34	300
metamitron	0	2	-	-	0	24	-	-
methiocarb	0	1	0	26	0	13	0	337
parathion-methyl	0	1	-	-	0	13	-	-
metolachlor	5	143	1	25	68	1283	7	244
mevinphos	0	8	0	12	0	80	0	118
o.p'-DDD	0	12	0	4	0	158	0	50
p.p'-DDT	0	0	0	3	0	0	0	45
PCP	21	48	13	62	128	228	74	195
pentachlorbenzene	0	1	0	2	0	8	3	13
pirimicarb	0	3	0	5	0	35	0	60
pirimiphos-methyl	0	15	99	1172	0	10	-	-
prochloraz	0	18	0	23	0	237	0	303
procymidon	3	83	1	61	25	416	12	369
propachlor	52	1536	28	1089	207	8253	366	7171
propoxur	0	9	0	5	0	118	0	64
pyrazophos	0	32	0	0	0	425	0	0
simazine	0	20	0	21	0	206	0	148
terbutylazine	0	39	1	129	0	504	12	585
tetrabromobisphen. A	0	0	1	26	0	0	10	336
tolclofos-methyl	0	138	0	93	0	921	-	-
triallate	100	464	16	433	177	1882	10	603
trifluralin	27	89	38	153	28	150	44	305
vinclozolin	6	143	16	507	25	1067	124	2640

Spatial distribution of total deposition

Minimum, maximum and average deposition in gram per hectare per year have been calculated from the total deposition to surface water for all eighteen locations. The location with the maximum dpeosition has also been determined. These data are presented in Table III.6.

Table III.6 Total deposition on Dutch surface water (gram per hectare per year). The variation over the Netherlands is indicated by presenting the national minimum and the national maximum.

Netherlands surface water	**2000 min g/ha/yr**	**2000 max g/ha/yr**	**2000 average g/ha/yr**	**Location max**	**2001 min g/ha/yr**	**2001 max g/ha/yr**	**2001 average g/ha/yr**	**Location max 2001**
2.4-D	*0.000*	0.009	0.004	KE	0.003	0.026	0.013	AP
atrazine	0.044	0.169	0.086	WY	0.032	0.208	0.090	YE
bentazon	*0.000*	0.032	0.006	ZI	*0.000*	0.063	0.025	KE
captan	*0.000*	0.241	0.055	WY	*0.000*	0.746	0.231	ST
chlorfenvinphos	*0.000*	0.021	0.003	EG	*0.000*	0.029	0.002	EG
chlorprophfam	0.051	9.814	1.965	AP	0.143	4.980	1.031	ZI
chlorpyriphos-methyl	0.001	0.017	0.005	ZI	0.000	0.007	0.002	KE
chloridazon (pyrazon)	*0.000*	0.014	0.002	YE	-	-	-	-
chlorothalonil	0.027	0.266	0.150	VE	0.007	0.365	0.136	AP
diazinon	*0.000*	0.023	0.004	AL	*0.000*	0.030	0.003	AN
dichlobenil	0.348	5.210	2.423	YE	0.612	2.963	1.547	KE
dichlorvos	*0.000*	0.620	0.065	GP	*0.000*	0.462	0.069	DE
dimethoate	*0.000*	0.115	0.022	AN	*0.000*	0.046	0.010	WY
DNOC	2.736	10.743	8.463	SP	1.238	5.632	4.272	DE
endosulfan I	*0.000*	0.030	0.004	WY	*0.000*	0.038	0.008	SP
endosulfan II	*0.000*	0.052	0.012	GP	*0.000*	0.028	0.003	VE
epoxiconazole	*0.000*	0.024	0.011	GP	-	-	-	-
ethofumesate	*0.000*	0.073	0.029	GP	0.019	0.413	0.119	WY
parathion-ethyl	*0.000*	0.008	0.001	AN	*0.000*	0.005	0.000	NO
fenitrothion	*0.000*	0.003	0.000	NO	-	-	-	-
fenthion	*0.000*	0.001	0.000	DE	-	-	-	-
fluazinam	0.025	0.296	0.111	YE	0.016	0.660	0.188	KE
fluroxypyr	*0.000*	0.078	0.017	GP	*0.000*	0.081	0.032	AL
hexachlor-1.3-butadiene	*0.000*	0.000	0.000	AP	-	-	-	-
hexachlorbenzene	0.003	0.019	0.006	AP	0.001	0.024	0.005	AN
isoxaflutole	*0.000*	0.009	0.002	DE	-	-	-	-
kresoxim-methyl	*0.000*	0.070	0.030	AP	0.012	0.101	0.055	WY
lindane	0.039	0.370	0.139	WY	*0.000*	0.189	0.078	AL
malathion	*0.000*	0.003	0.000	SP	-	-	-	-
MCPA	0.003	0.109	0.056	YE	0.020	0.093	0.043	AP
mecoprop	0.008	0.093	0.042	YE	0.009	0.090	0.043	WY
metamitron	*0.000*	0.007	0.002	DE	-	-	-	-
methiocarb	*0.000*	0.002	0.000	AL	*0.000*	0.092	0.024	BE
parathion-methyl	*0.000*	0.004	0.000	SP	-	-	-	-
metolachlor	0.019	0.514	0.084	WY	0.002	0.089	0.019	WY
mevinphos	*0.000*	0.028	0.007	SP	*0.000*	0.044	0.010	LE
o.p'-DDD	*0.000*	0.043	0.013	GP	*0.000*	0.014	0.004	NO

Netherlands surface water	**2000 min**	**2000 max**	**2000 average**	**Loca-tion**	**2001 min**	**2001 max**	**2001 average**	**Loca-tion max 2001**
	g/ha/yr	**g/ha/yr**	**g/ha/yr**	**max**	**g/ha/yr**	**g/ha/yr**	**g/ha/yr**	
p.p'-DDT	*0.000*	*0.000*	*0.000*	-	*0.000*	0.012	0.001	KE
PCP	0.075	0.171	0.137	ST	0.047	0.222	0.097	AN
pentachlorbenzene	0.000	0.005	0.001	AP	0.001	0.006	0.002	AN
pirimicarb	*0.000*	0.009	0.001	SP	*0.000*	0.016	0.003	AN
pirimiphos-methyl	*0.000*	0.053	0.008	AP	0.354	4.202	1.279	LE
prochloraz	*0.000*	0.065	0.007	NO	*0.000*	0.082	0.009	AN
procymidon	0.012	0.298	0.089	ZI	0.003	0.218	0.046	ZI
propachlor	0.185	5.507	1.112	KE	0.099	3.902	0.690	KE
propoxur	*0.000*	0.032	0.008	AN	*0.000*	0.017	0.005	DE
pyrazophos	*0.000*	0.116	0.008	EG	*0.000*	*0.000*	*0.000*	VE
simazine	*0.000*	0.073	0.014	GP	*0.000*	0.074	0.012	AP
terbutylazine	*0.000*	0.140	0.041	GP	0.003	0.461	0.103	GP
tetrabromobisphenol A	*0.000*	0.000	0.000	DE	0.003	0.092	0.025	ZI
tolclofos-methyl	*0.000*	0.494	0.054	ZI	*0.000*	0.332	0.042	AP
triallate	0.359	1.664	0.716	KE	0.057	1.552	0.283	WY
trifluralin	0.096	0.320	0.201	ZI	0.138	0.549	0.297	YE
vinclozolin	0.023	0.513	0.157	AP	0.057	1.817	0.378	KE

The location with the maximum deposition in both monitoring years is only the same for a number of pesticides: *chlorfenvinphos* (EG), *metolachlor* (GP), *procymidon* (ZI), *propachlor* (KE) and *terbutylazine* (GP). This is partly caused by the fact that some pesticides were in 2000 only measured at four locations and in 2001 at eighteen. The meteorological influence or a change in use of pesticides may be another explanation.

Annex IV Processing methods used

Interpolation of spatial data

Method for spatial presentation of the deposition data at the monitoring stations
Spatial presentation of the deposition results at the monitoring stations over the Netherlands requires interpolation for locations between the monitoring stations, and extrapolation for locations that are not situated between monitoring stations. Linear interpolation with the GIS programme ArcView was used earlier. This sometimes leads to strange values in areas outside the monitoring stations as result of interpolation artefacts. The *fluazinam* values at the 18 monitoring locations have, with an OPS-modelled concentration field, been extracted to find a more suitable interpolation method. It has been attempted with different interpolation techniques to resolve the best possible model field with those eighteen point values, where the OPS model has been taken as a sort of reference because it generally gives a good description of the distribution of substances over the country in case good emission data bases are available. The following interpolation methods have been used:

- the Kriging method, with different models for modelling semi-variogrammes (linear, exponential, circular etc.),
- interpolation, weighted with the inversion of the distance between the monitoring stations (IDW),
- the Nearest Neighbour method, in which a number of neighbouring stations has an effect on the concentration.

A distance scale, over which the effect of the monitoring points reaches, needs to be set for all methods, either by giving a number of neighbouring stations or by giving a fixed distance as radius. As stated earlier, the different monitoring points will be representative at a scale of some kilometres to tens of kilometres, because the monitoring points are not situated in the direct vicinity of emission sources of pesticides. The influence of the monitoring stations should therefore be chosen in that order of magnitude. It was found that a good description of the semi-variogrammes, which are for the Kriging method determined from the monitoring values of the different stations, with one of the semi-variogramme models was only possible for few substances. The spatial relationship between the mutual stations as often poor. For those substances that are transported to the Netherlands from surrounding countries, there was sufficient coherence and Kriging interpolation works well. For *fluazinam*, in this example, with the most important sources within the Netherlands, the result with Kriging with linear model, with 50 kilometre radius, was reasonable. Disadvantages of Kriging were that maximum values were levelled off and that minimum values were filled, resulting in a very “smoothened” picture. This was not so much the case with the IDW method. The best result was obtained with the IDW method with a radius of 50 kilometres. A larger scale leads to artefacts, in particular for points that are not situated between monitoring stations. A disadvantage then still is that not the total country is covered. Inverted distance interpolation, with 50 kilometre radius, has thus been used for the pictures presented here.

Averaging methods

Determination of representative variables from the available monitoring data is often not simple because the concentration of some substances does not frequently and often only little exceed the limit of detection. Attention was paid to this in the determination of the concentration of substances in the samples that were taken. The limit of detection was estimated for each series of samples, but the concentration can also be determined below this limit of detection. This can be understood as follows:

The GC-MS method determines the identity of substances on the basis of retention time and further identification of the substances by means of mass spectrometry. In practice, the ratio between the intensity of two masses in the mass spectrum is always determined and compared with the ratio in a standard solution of that substance (see Duyzer et al., 1999). In case of successful identification of the substance, the concentration of the substance is calculated on the basis of calibration by means of the standard solution. A ‘0’ value is reported for this substance in case the identification is not successful. For each sample and for each substance, a rough estimate has been made of the concentration below which identification is not possible. This makes it possible to get an impression of the concentration of the substance, which might be present, without detecting the substance in the sample. This ‘detection limit’ strongly depends on the amount of interfering substances in the sample and therefore varies per sample. This means that it is possible to detect a substance in one sample in concentrations well below the limit of detection that would apply for a different, more polluted, sample. This means that the limit of detection (calculated as three times the noise level) is not much more than an indication of the quality of the **quantitative** analysis.

This results in the following three types of analytical results being used:

1. A concentration in g/l in case of successful identification; in the tables indicated as the percentage in which the substance was *detected.*
2. A concentration *‘0’* is reported in case identification of the substance is not possible.
3. An estimate of the limit of detection, calculated as three times the noise.

The limit of detection is in principle only used to gain insight into the quality of the measuring result.

For pesticides there is the additional aspect that substances are not used throughout the year. A particular herbicide may only be used during three months in spring. The substance is quite well measurable during three months and not during the rest of the year. The substance can even exceed the MTR values during those three months. This results in a very low calculated annual average and does then not give a good picture of the presence of the substance.

As an illustration Figure 9 presents the DNOC concentration in precipitation samples. This substance is found throughout the year. Derivation of representative variables is relatively simple because a reasonably normal distribution is found. It is much more difficult to derive a representative variable for a substance such as *propachlor* (Figure 4 and 5). The number of times that this substance can be detected is much lower than the number of times that *DNOC* can be detected. In addition, the presence shows a log-normal distribution. There are many samples in which the substance can hardly be detected.

It is not beforehand clear how in this case the data are best presented. To overcome this problem, this report presents the number of times that the substance is found besides the average values, and any criterion exceedances.

Finally, the annual average deposition has also been calculated for most substances.

Alternative averaging methods

To restrict the sensitivity of summarising statistics, such as the mathematical average, for outliers and to obtain a more representative central value for the measured concentration ranges, alternatives have been sought for the mathematical average of the concentration, where the statistical distribution of those substances is taken into account. It is, e.g., assumed that the concentrations of substances below the lower limit of detection of the method shows a log-normal distribution. Such a distribution can arise because many substances are present at levels close to the “limit of detection” of the analytical method, and are consequently difficult to determine. The data are on that side limited by the analytical method. The mathematical average is then strongly influenced by a small number of high values. This means that a data set is statistically distorted (more than half of the values from the measuring range are then lower than the average). It should be noted that concentrations of substances below the level of the limit of detection, which could, however, be identified via mass spectrometry and gas chromatography, have been reported and have not been replaced by a “< limit of detection” sign. They are incorporated in the mathematical average of the concentration. Duyzer and Vonk (2001) extensively deal with this point. Percentiles then probably give a better picture of the measuring range, where the median, the 50-percentile, indicates the middle of the measuring range, when all measurements are listed in order of rank.

A different method to estimate the average of distorted measuring ranges is to plot the logarithm of the cumulative percentage of the total mass of a measuring range of concentrations against the concentration of the measurements of that range on log-probability paper. This showed for a number of substances that these did not show a log-normal distribution. An average could therefore not be estimated via log-probability plots.

Annex V Atmospheric deposition in the vicinity of the source

The objective of the study presented here was to obtain an overview of the atmospheric deposition into the Netherlands. Although deposition close to the source was not subject of the current study, this process does of course also occur in the vicinity of the source. At a *local* scale, close to sources, deposition can even be much higher. This is illustrated in Figure V.1 where the concentration of a substance in the vicinity of a source is given. This is an example for which calculations have been carried out with a standard emission source with a strength of 0.01 g/s. It is clear that the concentration in air decreases with the distance to the source. Deposition is directly proportional to the concentration in air and therefore decreases just as rapidly. The concentration at ten kilometres distance from the source, e.g., is already 50 times as low as at a distance of one kilometre. This is a completely different scale than that of the current monitoring network. The current monitoring network is aimed at obtaining picture on a national scale. The monitoring locations have therefore been selected in that way, far away from large sources. For an order of magnitude: eighteen monitoring stations in the whole of the Netherlands (almost 40 000 km^2) makes the stations representative at a scale of almost 50 by 50 km. This means that deposition in the direct vicinity of sources will be much higher than the national average. The results from the current study are more representative of the national average. The chance that this process results in high concentrations in surface water which may possibly lead to effects is therefore also higher.
It is also noteworthy that, in Figure V.1 almost 20% of the emitted amount of material deposits within one kilometre from the source. The substance chosen in the example given here is a substance with a moderately high solubility. The concentration course may be somewhat different for substances with a lower solubility, which may result in a smaller proportion that deposits. The effect on the concentration course, however, is fairly small because the concentration course over distance is mainly determined by dilution. The deposited fraction depends much more on the physical-chemical properties. The example given here is roughly the maximum. Substances with a lower solubility deposit less at a small distance from the source.

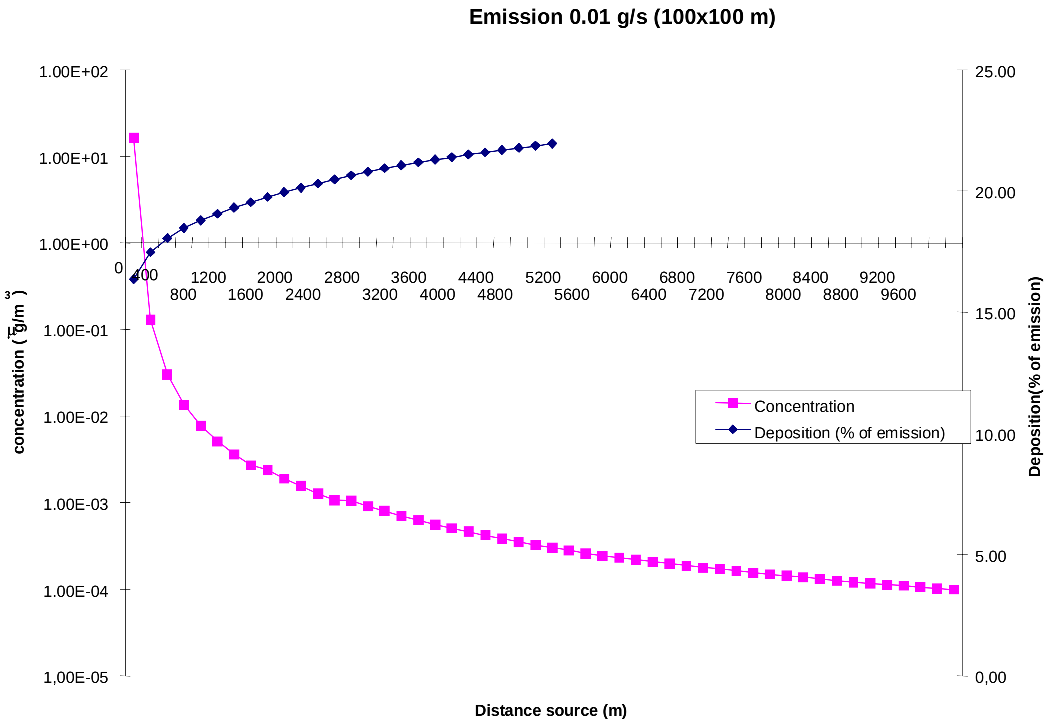

Figure V.1 *Concentration(µg/m^3) in air of an example substance (sulphur dioxide) and the fraction of the emitted substance which deposits, as function of the distance to the source, for a field of 100 by 100 m at a source strength of 0.01 g/s.*

Annex VI Concentrations and depositions in Noord-Holland

1.1 Introduction

Measurements have been carried out at additional monitoring stations in the province Noord-Holland during the course of the project. The results of the measurements at these stations are discussed separately in this Annex.

1.2 Results

1.2.1 Concentrations and criteria

Tables VI.1 – VI.6 present the results of the measurements in the province at the locations Andijk, Anna Paulowna, Aalsmeer and Egmond.
The exceedance of the drinking water criterion is high for *chlorpropham* at the monitoring locations in Noord-Holland in comparison with the other monitoring locations in the Netherlands. The average concentration reaches a value of six times the drinking water criterion, while the average for the Netherlands as a whole (including Noord-Holland) reaches a factor four. The percentage of the samples in which the criterion is exceeded is two times as high as the percentage for the Netherlands as a whole (including Noord-Holland). *Pyrazophos*, *tolcofos-methyl* en *chlorothalonil* do in Noord-Holland only exceed the drinking water criterion in precipitation (Table VI.7).
The *chlorothalonil* concentration was in 2000 only measured at the monitoring location Anna Paulowna where it exceeded the MTR for surface water in 90% of the precipitation samples. The maximum exceedance was a factor 32! In 2001, the MTR for *chlorothalonil* was exceeded in 25% of the samples, which corresponds with the national average. Also in 2001, the maximum exceedance was found in Anna Paulowna. In Noord Holland. *pyrazophos* and *chlorfenvinphos* did only exceed the MTR in precipitation.

Table VI.1 Presence of pesticides in air samples in Noord-Holland for the years 2000 and 2001. The percentage of the samples in which the substance was detected is given besides the average concentration over all samples (- analysis for this substance was no longer carried out; 0: analysis for this substance was carried out but the substance was not detected).

Substance	**Detected in air 2000 (%)**	**Detected in air 2001 (%)**	**Average concentration 2000 (ng/m^3)**	**Average concentration 2001 (ng/m^3)**
2,4-D	0	0	0.000	0.000
aldrin	0	-	0.000	-
atrazine	4	2	0.003	0.000
bentazon	0	6	0.000	0.009
bitertanol	0	-	0.000	-
captan	0	2	0.000	0.012
chlorfenvinphos	2	2	0.002	0.004
chlorpropham	52	56	1.940	0.714
chlorpyriphos-methyl	21	12	0.003	0.001
chlorothalonil[1]	56	34	0.229	0.217
chloridazon (pyrazon)	0	-	0.000	-
deltamethrin	0	-	0.000	-
demeton-S-methyl	33	-	0.060	-
diazinon	2	0	0.002	0.000
dichlobenil	98	98	1.565	1.071
dichlorvos	0	6	0.000	0.016
dicofol	0	-	0.000	-
dimethoate	0	2	0.000	0.002
disulfoton	0	-	0.000	-
DNOC	21	28	0.299	0.302
endosulfan I	0	0	0.000	0.000
endosulfan II	2	0	0.009	0.000
endrin	11	-	0.023	-
epoxiconazole	0	-	0.000	-
ethofumesate[1]	22	22	0.003	0.040
parathion-ethyl	0	0	0.000	0.000
fenitrothion	0	-	0.000	-
fenthion	0	-	0.000	-
fluazinam	25	20	0.029	0.015
fluroxypyr[2]	0	12	0.000	0.036
phosfamidon	0	-	0.000	-
heptachlor	0	0	0.000	0.000
heptachlor epoxide	44	-	0.004	-
heptenophos	0	-	0.000	-
hexachlor-1.3-butadiene	11	-	0.009	-
hexachlorbenzene	42	32	0.006	0.010
isoxaflutole	0	-	0.000	-
kresoxim-methyl[1]	33	12	0.017	0.006
lindane	12	2	0.056	0.003
malathion	0	-	0.000	-
MCPA	2	0	0.000	0.000
mecoprop	2	4	0.000	0.002
metamitron	0	-	0.000	-

Substance	**Detected in air 2000 (%)**	**Detected in air 2001 (%)**	**Average concentration 2000 (ng/m^3)**	**Average concentration 2001 (ng/m^3)**
methiocarb	0	4	0.000	0.001
methomyl	0	-	0.000	-
parathion-methyl	0	-	0.000	-
metolachlor	0	2	0.000	0.000
mevinphos	2	4	0.001	0.006
o.p'-DDD[1]	11	0	0.016	0.000
o.p'-DDE	11	-	0.007	-
p.p'-DDT[1]		-		-
PCP	79	74	0.055	0.067
pentachlorbenzene[2]	33	26	0.010	0.004
pirimicarb	0	0	0.000	0.000
pirimiphos-methyl	19	10	0.019	0.796
prochloraz	0	0	0.000	0.000
procymidon	44	22	0.055	0.025
propachlor	40	28	0.436	0.158
propoxur	2	0	0.002	0.000
pyrazophos	4	0	0.003	0.000
simazin	8	4	0.019	0.014
telodrin	11	-	0.003	-
terbutylazine[1]	0	6	0.000	0.018
tetrabromobisphenol A[1]	44	22	0.000	0.005
tolclofos-methyl	33	30	0.161	0.247
triadimenol	0	-	0.000	-
triallate	75	60	0.316	0.131
triazophos	0	-	0.000	-
trifluralin	81	92	0.193	0.328
vinclozolin[1]	89	36	0.321	0.171

1) Substances analysed for at four locations in 2000 and at all locations in 2001.

\- Substances no longer analysed for in the year 2001.

Table VI.2 Presence of pesticides in precipitation samples in Noord-Holland for the years 2000 and 2001. The percentage of the samples in which the substance was detected is given besides the average concentration over all samples (- analysis for this substance was no longer carried out; 0: analysis for this substance was carried out but the substance was not detected).

Substance	**Detected in precipitation 2000 (%)**	**Detected in precipitation 2001 (%)**	**Average concentration 2000 (ng/l)**	**Average concentration 2001 (ng/l)**
2,4-D	10	33	0.9	2.0
aldrin	0	-	*0.0*	-
atrazine	21	23	10.1	13.8
bentazon	2	12	0.3	0.3
bitertanol	0	-	*0.0*	-
captan	11	8	4.9	22.7
chlorfenvinphos	4	4	0.5	1.0
chlorpropham	96	87	250.8	102.2
chlorpyriphos-methyl	52	38	0.7	0.2
chlorothalonil[1]	100	60	52.6	17.0
chloridazon (pyrazon)	0	-	*0.0*	-
deltamethrin	0	-	*0.0*	-
demeton-S-methyl	0	-	*0.0*	-
diazinon	4	12	0.8	1.0
dichlobenil	100	88	17.8	13.7
dichlorvos	23	27	3.2	3.3
dicofol	0	-	*0.0*	-
dimethoate	8	0	8.4	*0.0*
disulfoton	0	-	*0.0*	-
DNOC	100	96	1168.4	451.7
endosulfan I	0	0	*0.0*	*0.0*
endosulfan II	2	2	0.5	0.2
endrin	0	-	*0.0*	-
epoxiconazole	0	-	*0.0*	-
ethofumesate[1]	40	12	5.1	9.1
parathion-ethyl	2	0	0.5	*0.0*
fenitrothion	0	-	*0.0*	-
fenthion	0	-	*0.0*	-
fluazinam	48	40	6.3	15.7
fluroxypyr[2]	40	4	12.9	0.2
phosfamidon	0	-	*0.0*	-
heptachlor	0	0	*0.0*	*0.0*
heptachlor epoxide	0	-	*0.0*	-
heptenophos	0	-	*0.0*	-
hexachlor-1.3-butadiene	0	-	*0.0*	-
hexachlorbenzene	94	79	0.5	0.5
isoxaflutole	0	-	*0.0*	-
kresoxim-methyl[1]	50	44	13.4	8.7
lindane	31	31	16.8	15.1
malathion	0	-	*0.0*	-
MCPA	63	52	10.9	7.8

Substance	**Detected in precipitation 2000 (%)**	**Detected in precipitation 2001 (%)**	**Average concentration 2000 (ng/l)**	**Average concentration 2001 (ng/l)**
mecoprop	42	31	7.4	5.5
metamitron	10	-	0.5	-
methiocarb	7	12	0.1	2.8
methomyl	0	-	*0.0*	-
parathion-methyl	0	-	*0.0*	-
metolachlor	21	15	5.7	2.0
mevinphos	2	8	0.3	0.2
o.p'-DDD[1]	10	2	0.1	0.0
o.p'-DDE	0	-	*0.0*	-
p.p'-DDT[1]		-		-
PCP	98	88	9.4	5.0
pentachlorbenzene[2]	90	81	0.4	0.3
pirimicarb	0	6	*0.0*	0.3
pirimiphos-methyl	0	8	*0.0*	10.2
prochloraz	2	2	1.1	2.9
procymidon	81	69	17.0	6.8
propachlor	54	52	126.4	62.0
propoxur	21	12	2.6	0.7
pyrazophos	4	0	6.2	*0.0*
simazin	2	4	0.5	1.3
telodrin	0	-	*0.0*	-
terbutylazine[1]	40	29	3.1	10.0
tetrabromobisphenol A[1]	50	71	0.0	4.1
tolclofos-methyl	19	29	4.7	8.3
triadimenol	0	-	*0.0*	-
triallate	79	50	23.0	3.2
triazophos	0	-	*0.0*	-
trifluralin	73	79	2.6	4.4
vinclozolin[1]	100	83	45.7	18.9

Table VI.3 Presence of PAHs in air samples in Noord-Holland for the years 2000 and 2001. The percentage of the samples in which the substance was detected is given besides the average concentration over all samples.

Substance	**Detected in air 2000 (%)**	**Detected in air 2001 (%)**	**Average concentration 2000 (ng/m^3)**	**Average concentration 2001 (ng/m^3)**
acenaphtene	100	97	2.293	1.747
acenaphtylene	50	55	0.083	0.124
anthracene	90	61	0.137	0.087
benzo[a]anthracene	73	29	0.027	0.006
benzo[a]pyrene	73	71	0.036	0.031
benzo[b]fluorantene	83	89	0.080	0.139
benzo[g,h,i]perylene	88	82	0.033	0.050
benzo[k]fluorantene	83	92	0.052	0.095
chrysene	73	32	0.104	0.016
dibenz[a.h]-anthracene	63	50	0.004	0.008
fenantrene	100	97	6.522	5.841
fluorantene	96	97	1.867	1.822
fluorene	100	97	3.591	2.855
indeno[1,2,3-cd]pyrene	79	76	0.028	0.047
naphtalene	100	100	12.769	12.994
pyrene	92	92	0.775	0.728

Table VI.4 Presence of PCBs in air samples in Noord-Holland for the years 2000 and 2001. The percentage of the samples in which the substance was detected is given besides the average concentration over all samples (0: analysis for this substance was carried out but the substance was not detected).

Substance	**Detected in air 2000 (%)**	**Detected in air 2001 (%)**	**Average concentration 2000 (ng/m^3)**	**Average concentration 2001 (ng/m^3)**
PCB-101	38	24	0.004	0.003
PCB-118	0	5	0.0004	*0.000*
PCB-138	15	3	0.0002	0.001
PCB-153	25	5	0.0001	0.001
PCB-180	12	0	*0.000*	0.0003
PCB-20	19	18	0.007	0.010
PCB-28	17	18	0.004	0.009
PCB-35	2	8	0.002	0.001
PCB-52	19	26	0.007	0.003
PCB-8	21	21	0.005	0.003

Table VI.5 Presence of PAHs in precipitation samples in Noord-Holland for the years 2000 and 2001. The percentage of the samples in which the substance was detected is given besides the average concentration over all samples.

Substance	**Detected in precipitation 2000 (%)**	**Detected in precipitation 2001 (%)**	**Average concentration 2000 (ng/l)**	**Average concentration 2001 (ng/l)**
acenaphtene	98	95	7.9	6.0
acenaphtylene	31	50	3.0	4.1
anthracene	100	95	8.5	7.1
benzo[a]anthracene	100	100	11.8	11.6
benzo[a]pyrene	88	98	18.2	16.5
benzo[b]fluorantene	100	100	39.1	30.1
benzo[g,h,i]perylene	98	95	18.3	13.7
benzo[k]fluorantene	100	100	23.0	22.8
chrysene	100	100	34.0	31.2
dibenz[a.h]-anthracene	79	88	2.9	3.7
fenantrene	100	100	96.4	95.3
fluorantene	100	100	94.2	93.6
fluorene	100	100	16.3	14.5
indeno[1,2,3-cd]pyrene	92	95	16.0	12.5
naphtalene	100	100	73.3	41.0
pyrene	100	98	52.1	58.1

Table VI.6 Presence of PCBs in precipitation samples in Noord-Holland for the years 2000 and 2001. The percentage of the samples in which the substance was detected is given besides the average concentration over all samples

Substance	**Detected in precipitation 2000 (%)**	**Detected in precipitation 2001 (%)**	**Average concentration 2000 (ng/l)**	**Average concentration 2001 (ng/l)**
PCB-101	50	15	1.4	0.1
PCB-118	33	3	0.4	0.02
PCB-138	46	8	0.4	0.2
PCB-153	73	33	0.5	0.2
PCB-180	13	5	0.1	0.2
PCB-20	42	45	0.9	0.6
PCB-28	37	5	1.3	0.1
PCB-35	6	5	0.1	0.1
PCB-52	31	23	2.0	0.4
PCB-8	10	0	0.1	0.0

Table VI.7 Substances that exceeded the drinking water criterion in precipitation (100 ng/l) at the monitoring locations in Noord-Holland in 2000 and 2001. The percentage samples that exceed the criterion are presented as well as the average concentration in those samples.

Substance	% samples > drinking water criterion 2000	% samples > drinking water criterion 2001	Average concentration of sample above the drinking water criterion 2000	Average concentration of sample above the drinking water criterion 2001
Fungicides				
captan	0	2	0	999
chlorothalonil	10	4	325	190
fluazinam	0	4	0	143
prochloraz	0	2	0	152
procymidon	2	0	245	0
pyrazophos	2	0	320	0
tolclofos-methyl	0	2	0	105
vinclozolin	2	4	174	146
Herbicides				
atrazine	0	6	0	118
chlorpropham	37	33	638	252
dichlobenil	2	2	259	167
ethofumesate	0	2	0	323
propachlor	29	21	407	252
terbutylazine	0	2	0	156
Insecticides				
dimethoate	6	0	146	0
lindane	6	2	117	341
pirimiphos-methyl	0	8	0	132
Other				
DNOC	100	96	1168	470
PCP	2	0	165	0

Table VI.8 Exceedance of the MTR in precipitation samples at the monitoring locations in Noord-Holland 2000 and 2001. The percentages of the month samples that exceed the MTR, the maximum MTR exceedance and the area of use are presented.

Substance	**Area of use**	**MTR (ng/l)**	**% detected in precipitation 2000**	**Concentration in precipitation 2000 (ng/l)**	**% > MTR 2000**	**% detected in precipitation 2001**	**Concentration in precipitation 2001 (ng/l)**	**% > MTR 2001**
chlorothalonil	F	10	100	52.6	90	60	17.0	25
captan	F	110	11	4.9	0	8	22.7	2
pyrazophos	F	40	4	6.2	2	0	0.0	0
propachlor	H	1300	54	126.4	2	52	62.0	0
chlorfenvinphos	I	2	4	0.5	4	4	1.0	4
chlorpyriphos-methyl	I	3	52	0.7	6	38	0.2	0
dichlorvos	I	0.7	23	3.2	23	27	3.3	27
parathion-ethyl	I	2	2	0.5	2	-	-	-
mevinphos	I	2	2	0.3	2	8	0.2	6
propoxur	I	10	21	2.6	4	12	0.7	2
methiocarb	I and B	16	7	0.1	0	12	2.8	8
pirimiphos-methyl	I/A	2	0	0.0	0	8	10.2	8
benzo[a]anthracene	PAK	30	100	11.8	4	100	11.6	0
fenantrene	PAK	300	100	96.4	4	100	95.3	3
fluorantene	PAK	500	100	94.2	2	100	93.6	3

1.2.2 Loads of atmospheric deposition in the province Noord-Holland

The total atmospheric deposition of pesticides on the surface water (g/ha/yr) in the year 2000 and 2001 are presented in Table VI.9 and Table VI.10. Besides the average deposition over Noord-Holland and the deposition at the four monitoring locations in Noord-Holland, the national average deposition over the Netherlands is presented for comparison. Surface water burdening at the monitoring stations in Noord Holland by *chlorpropham, chlorothaloni,. chlorpyriphos-methy,. procymidon, tolcofos-methyl* and *vinclozolin* is higher than the national average for the Netherlands. These substances are mainly used in flower bulb growing or potato growing.

In 2001, the maximum deposition was found at a monitoring location in the province for one third of the substances that was analysed for. This is slightly more than would be expected in view of the distribution of the monitoring locations. Four of the total of 18 monitoring locations are situated in the province. This means that in case of a proportional distribution of the maximum values over the country only 4/18 (22%) of the maximum values would be found in the province. The slight increase of this percentage is probably caused by the accidental situation of the monitoring locations in relation to sources and the composition of the analytical package.

Table VI.9 Total atmospheric deposition of pesticides on surface water (g/ha/yr) in the year 2000. The average deposition over the Netherlands, over Noord-Holland and the deposition at the four monitoring locations in Noord-Holland are presented. The values for the substances ***printed bold*** *are reliable. The reliability of the other values is low (see also text). The Italic "0" means that the substance was not detected.*

Total deposition on surface water (g/ha/yr) 2000	**Nether-lands average**	**Average Noord-Holland**	**Aalsmeer**	**Andijk**	**Anna Paulowna**	**Egmond**	**Location maximum Nether-lands**
2,4-D	0.004	0.005	0.003	0.008	0.004	0.003	KE
atrazine	0.09	0.07	0.05	0.08	0.07	0.08	WY
bentazon	0.01	0.003	*0.00*	0.01	*0.00*	*0.00*	ZI
captan	0.06	0.05	0.05	0.05	0.05	0.03	WY
chlorfenvinphos	0.003	0.006	*0.00*	0.002	*0.00*	0.02	EG
chlorpropham	1.99	4.43	0.53	2.93	9.81	4.43	AP
chlorpyriphos-methyl	0.005	0.008	0.001	0.004	0.018	0.010	ZI
chlorothalonil	0.29	0.32	-	-	0.32	-	VE
diazinon	0.005	0.01	0.03	*0.00*	*0.00*	*0.00*	AL
dichlobenil	2.45	2.51	2.25	2.39	4.01	1.39	YE
dichlorvos	0.06	0.03	0.05	0.03	0.01	0.02	GP
dimethoate	0.02	0.07	0.08	0.11	*0.00*	0.07	AN
DNOC	8.56	9.08	10.09	8.88	8.79	8.54	SP
endosulfan II	0.01	0.01	0.02	*0.00*	*0.00*	0.04	GP
epoxiconazole	0.02	*0.00*	-	-	*0.00*	-	GP
ethofumesate	0.04	0.04	-	-	0.04	-	GP
fluazinam	0.11	0.08	0.17	0.08	0.05	0.03	YE
fluroxypyr	0.07	0.10	-	-	0.10	-	GP
heptachlor epoxide	0.007	0.005	-	-	0.005	-	GP
hexachlorbenzene	0.01	0.01	0.003	0.02	0.02	0.01	AP
kresoxim-methyl	0.04	0.09	-	-	0.09	-	AP
lindane	0.14	0.18	0.22	0.28	0.08	0.12	WY
MCPA	0.06	0.06	0.07	0.05	0.07	0.06	YE
mecoprop	0.04	0.04	0.04	0.04	0.03	0.04	YE
methiocarb	0.0002	0.001	0.004	*0.000*	*0.000*	*0.000*	AL
metolachlor	0.08	0.04	0.02	0.07	0.03	0.05	WY
mevinvos	0.007	0.003	*0.000*	*0.000*	0.007	0.005	SP
o.p'-DDD	0.03	0.02	-	-	0.02	-	GP
PCP	0.14	0.13	0.13	0.13	0.13	0.13	ST
pentachlorbenzene	0.002	0.01	-	-	0.01	-	AP
pirimicarb	0.002	*0.00*	*0.00*	*0.00*	*0.00*	*0.00*	SP
pirimiphos-methyl	0.016	0.024	0.003	0.002	0.083	0.007	AP
prochloraz	0.01	0.01	*0.00*	0.02	*0.00*	*0.00*	NO
procymidon	0.09	0.16	0.06	0.15	0.19	0.26	ZI
propachlor	1.12	1.30	0.76	2.01	1.78	0.64	KE
propoxur	0.01	0.02	0.03	0.03	0.003	0.002	AN
pyrazophos	0.02	0.03	*0.00*	*0.00*	0.00	0.13	EG
simazine	0.01	0.03	0.07	0.03	0.02	*0.00*	AL
terbutylazine	0.05	0.02	-	-	0.02	-	GP
tetrabromobisphenol A	0.0001	0.0001	-	-	0.0001	-	DE
tolclofos-methyl	0.05	0.12	0.00	0.03	0.37	0.06	ZI
triallate	0.72	0.63	0.56	0.67	0.66	0.61	KE
trifluralin	0.21	0.24	0.28	0.21	0.25	0.20	ZI
vinclozolin	0.28	0.71	-	-	0.71	-	AP

Table VI.10 Total atmospheric deposition of pesticides on surface water (g/ha/yr) in the year 2001. The average deposition over the Netherlands, over Noord-Holland and the deposition at the four monitoring locations in Noord-Holland are presented. The values for the substances **printed bold** *are reliable. The reliability of the other values is low (see also text). The Italic "0" means that the substance was not detected.*

Total deposition on surface water (g/ha/yr) 2001	Nether-lands average	Average Noord-Holland	Aalsmeer	Andijk	Anna Paulowna	Egmond	Location maximum Nether-lands
2,4-D	0.01	0.01	0.01	0.01	0.03	0.01	AP
atrazine	0.09	0.07	0.11	0.06	0.05	0.06	YE
bentazon	0.03	0.02	0.002	0.020	0.008	0.037	KE
captan	0.31	0.36	1.25	*0.00*	0.12	0.08	AL
chlorfenvinphos	0.002	0.01	*0.00*	*0.00*	0.01	0.03	EG
chlorpropham	1.04	1.78	0.39	1.55	2.71	2.45	ZI
chlorpyriphos-methyl	0.002	0.003	0.001	0.002	0.006	0.002	KE
chlorothalonil	0.14	0.18	0.11	0.17	0.36	0.09	AP
diazinon	0.003	0.01	0.01	0.03	0.01	*0.00*	AN
dichlobenil	1.59	1.75	1.81	2.06	2.05	1.07	KE
dichlorvos	0.07	0.05	0.08	0.10	0.03	0.01	DE
dimethoate	0.01	0.003	*0.00*	0.01	*0.00*	*0.00*	WY
DNOC	4.33	4.05	3.74	4.14	4.33	4.00	DE
endosulfan II	0.003	0.002	*0.00*	*0.00*	0.01	*0.00*	VE
epoxiconazole	-	-	-	-	-	-	-
ethofumesate	0.12	0.09	0.12	0.11	0.12	0.02	WY
fluazinam	0.19	0.14	0.08	0.13	0.25	0.08	KE
fluroxypyr	0.03	0.05	0.08	*0.00*	0.08	0.04	AL
heptachlor epoxide	-	-	-	-	-	-	-
hexachlorbenzene	0.01	0.02	0.01	0.02	0.01	0.01	AN
kresoxim-methyl	0.06	0.06	0.03	0.06	0.10	0.04	WY
lindane	0.08	0.09	0.25	0.03	0.05	0.04	AL
MCPA	0.05	0.06	0.08	0.04	0.09	0.03	AP
mecoprop	0.04	0.04	0.04	0.04	0.07	0.03	WY
methiocarb	0.02	0.03	0.02	0.02	0.07	0.02	BE
metolachlor	0.02	0.02	0.06	0.002	0.008	0.004	WY
mevinvos	0.01	0.01	0.02	0.002	0.02	*0.00*	LE
o.p'-DDD	0.004	0.0002	0.001	*0.00*	*0.00*	*0.00*	KE
PCP	0.10	0.14	0.17	0.22	0.08	0.09	AN
pentachlorbenzene	0.002	0.004	0.004	0.006	0.005	0.002	AN
pirimicarb	0.004	0.004	*0.000*	0.016	0.001	*0.000*	AN
pirimiphos-methyl	1.26	1.10	1.58	0.71	0.89	1.23	LE
prochloraz	0.01	0.02	*0.00*	0.08	*0.00*	*0.00*	AN
procymidon	0.04	0.08	0.02	0.06	0.10	0.13	ZI
propachlor	0.69	0.63	0.62	0.58	1.00	0.30	KE
propoxur	0.01	0.01	0.011	0.011	0.001	*0.000*	DE
pyrazophos	0.001	*0.00*	*0.00*	*0.00*	*0.00*	*0.00*	VE
simazine	0.01	0.03	*0.00*	0.02	0.07	0.03	AP
terbutylazine	0.10	0.07	0.06	0.04	0.04	0.15	GP
tetrabromobisphenol A	0.03	0.04	0.04	0.07	0.01	0.04	ZI
tolclofos-methyl	0.04	0.16	0.02	0.21	0.33	0.08	AP
triallate	0.29	0.18	0.15	0.14	0.23	0.21	WY
trifluralin	0.30	0.40	0.27	0.38	0.46	0.48	YE
vinclozolin	0.38	0.38	0.10	0.33	0.85	0.25	KE

Table VI.11 Atmospheric deposition of PAHs and PCBs on the surface water of the Netherlands and Noord Holland in the year 2000. The values for the substances ***printed bold*** *are reliable. The reliability of the other values is low.*

Atmospheric deposition	**Netherlands average**	**Average Noord-Holland**	**Aalsmeer**	**Andijk**	**Anna Paulowna**	**Egmond**	**Location maximum Netherlands**
PAHs							
acenaphtene	2.51	2.27	2.77	1.56	1.62	3.14	LA
acenaphtylene	0.19	0.12	0.20	0.08	0.07	0.12	NO
anthracene	0.64	0.26	0.29	0.18	0.28	0.27	KE
benzo[a]anthracene	0.13	0.14	0.14	0.11	0.11	0.20	EG
benzo[a]pyrene	0.21	0.19	0.24	0.14	0.14	0.25	KE
benzo[b]fluorantene	0.49	0.45	0.42	0.39	0.33	0.67	KE
benzo[g,h,i]perylene	0.19	0.20	0.19	0.18	0.16	0.28	EG
benzo[k]fluorantene	0.32	0.27	0.25	0.21	0.21	0.41	KE
chrysene	0.47	0.45	0.43	0.39	0.34	0.63	NO
dibenz[a.h]anthracene	0.03	0.03	0.03	0.03	0.02	0.04	SP
fenantrene	8.65	9.08	10.39	7.14	7.69	11.10	NO
fluorantene	3.08	3.45	3.36	2.96	2.50	4.97	EG
fluorene	4.49	4.15	4.75	3.58	3.32	4.95	NO
indeno[1,2,3-cd]pyrene	0.17	0.18	0.15	0.16	0.14	0.26	EG
naphtalene	7.04	7.21	10.37	5.49	5.65	7.32	AL
pyrene	1.73	1.63	1.77	1.33	1.30	2.13	NO
PCBs							
PCB-101	0.009	0.010	0.006	0.007	0.019	0.009	AP
PCB-118	0.003	0.003	0.001	0.002	0.005	0.002	AP
PCB-138	0.003	0.003	0.001	0.002	0.005	0.003	NO
PCB-153	0.004	0.003	0.001	0.002	0.004	0.005	SP
PCB-180	0.002	0.001	0.000	0.000	0.000	0.001	KE
PCB-20	0.008	0.008	0.004	0.006	0.008	0.012	NO
PCB-28	0.009	0.010	0.009	0.009	0.014	0.010	NO
PCB-35	0.001	0.001	0.001	0.001	*0.000*	0.002	EG
PCB-52	0.015	0.013	0.009	0.008	0.024	0.012	LA
PCB-8	0.003	0.002	0.002	0.002	0.003	0.001	NO

Table VI.12 Atmospheric deposition of PAHs and PCBs on the surface water of the Netherlands and Noord Holland in the year 2001. The values for the substances ***printed bold*** *are reliable. The reliability of the other values is low.*

Atmospheric deposition	**Nether-lands average**	**Average Noord-Holland**	**Aalsmeer**	**Andijk**	**Anna Paulowna**	**Egmond**	**Location maximum Nether-lands**
PAH							
acenaphtene	1.60	1.74	1.94	1.06	1.73	2.24	EG
acenaphtylene	0.16	0.17	0.34	0.13	0.12	0.07	NO
anthracene	0.18	0.16	0.16	0.09	0.20	0.19	NO
benzo[a]anthracene	0.08	0.08	0.08	0.07	0.07	0.09	EG
benzo[a]pyrene	0.12	0.13	0.16	0.11	0.13	0.11	AL
benzo[b]fluorantene	0.33	0.36	0.48	0.31	0.32	0.33	AL
benzo[g,h,i]perylene	0.13	0.15	0.20	0.13	0.14	0.13	AL
benzo[k]fluorantene	0.23	0.27	0.35	0.22	0.25	0.25	AL
chrysene	0.23	0.21	0.21	0.19	0.20	0.25	EG
dibenz[a.h]anthracene	0.03	0.04	0.03	0.04	0.05	0.03	AP
fenantrene	7.38	8.06	9.52	5.51	7.94	9.28	NO
fluorantene	2.68	3.27	3.54	1.96	3.21	4.39	EG
fluorene	3.44	3.31	4.00	2.53	2.96	3.75	NO
indeno[1,2,3-cd]pyrene	0.13	0.14	0.18	0.12	0.12	0.13	AL
naphtalene	6.34	7.09	11.29	5.24	5.82	6.01	AL
pyrene	1.44	1.51	1.56	0.90	1.64	1.93	NO
PCB							
PCB-101	0.002	0.002	0.001	0.003	0.001	0.001	LA
PCB-118	0.005	0.000	0.000	0.000	0.000	0.000	LE
PCB-138	0.002	0.001	0.001	0.004	0.001	0.000	KE
PCB-153	0.003	0.002	0.002	0.003	0.002	0.000	KE
PCB-180	0.001	0.001	0.001	0.005	0.000	0.000	AN
PCB-20	0.006	0.005	0.009	0.005	0.003	0.004	LA
PCB-28	0.006	0.003	0.008	0.002	0.000	0.001	LA
PCB-35	0.001	0.001	0.002	0.001	0.002	0.000	AL
PCB-52	0.006	0.004	0.003	0.006	0.004	0.002	KE
PCB-8	0.002	0.001	0.001	0.001	0.001	0.001	NO

Annex VII Contribution of the different provinces and surrounding countries to the depositions at the monitoring locations

Figure VII.1 - Figure VII.5 show these contributions for the pesticides *procymidon*, *fluazinam, dichlorvos* and *chlorothalonil* and for the PAH b*enzo(a)pyrene*. The figures show that the largest part of the deposition on the monitoring locations originates from the own province. Except for *dichlorvos* and *benzo(a)pyrene*, the contribution from surrounding countries on the deposition at the monitoring locations is small.

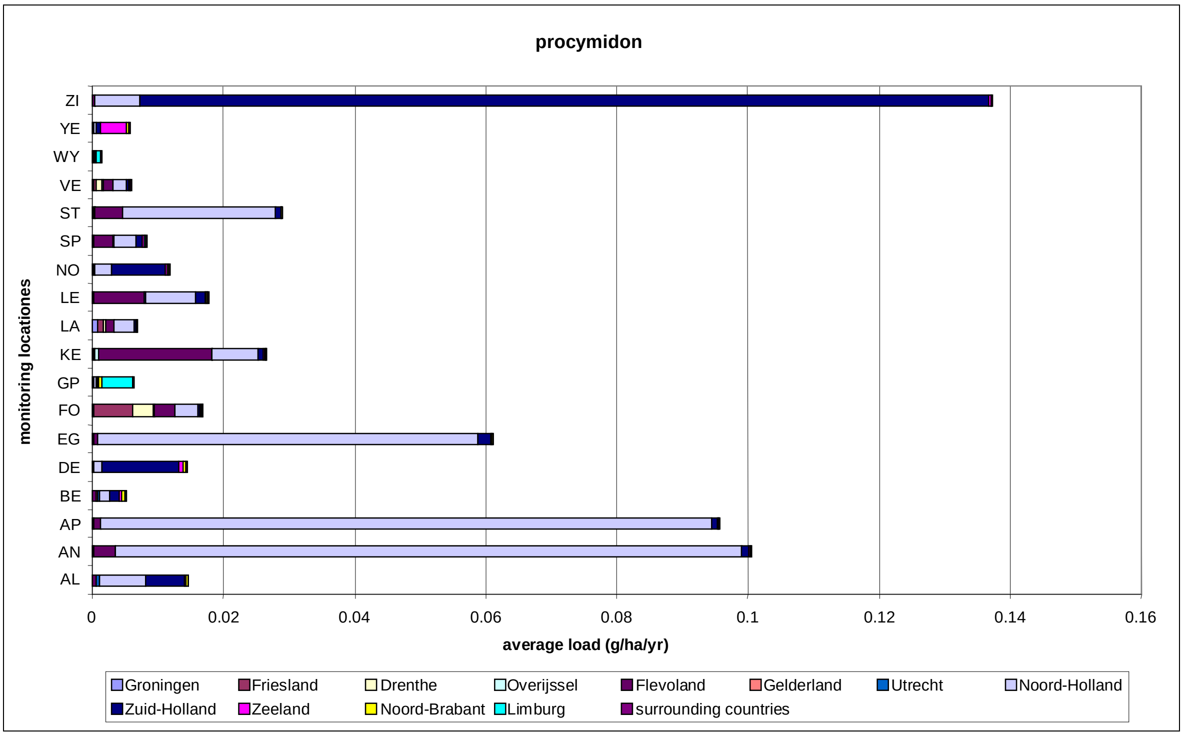

Figure VII.1 Contributions of emissions of procymidon per province and from surrounding countries to the average load at the monitoring locations (g/ha).

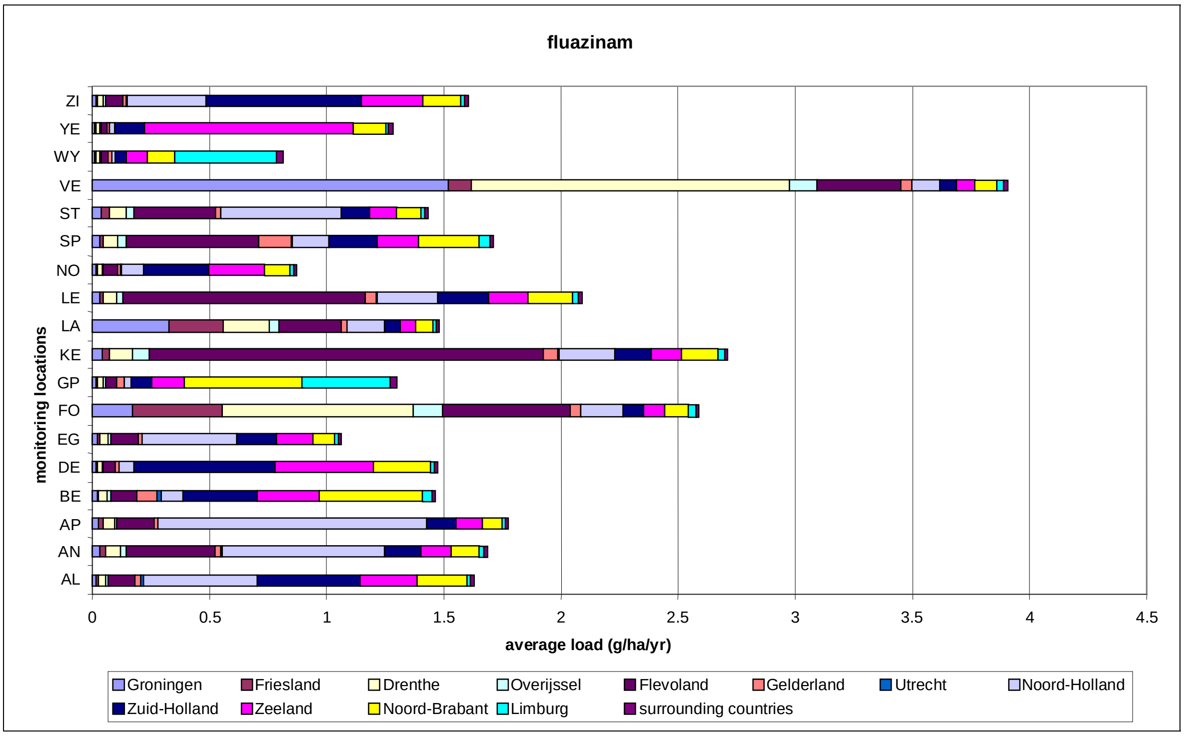

Figure VII.2 Contributions of emissions of fluazinam per province and from surrounding countries to the average load at the monitoring locations (g/ha).

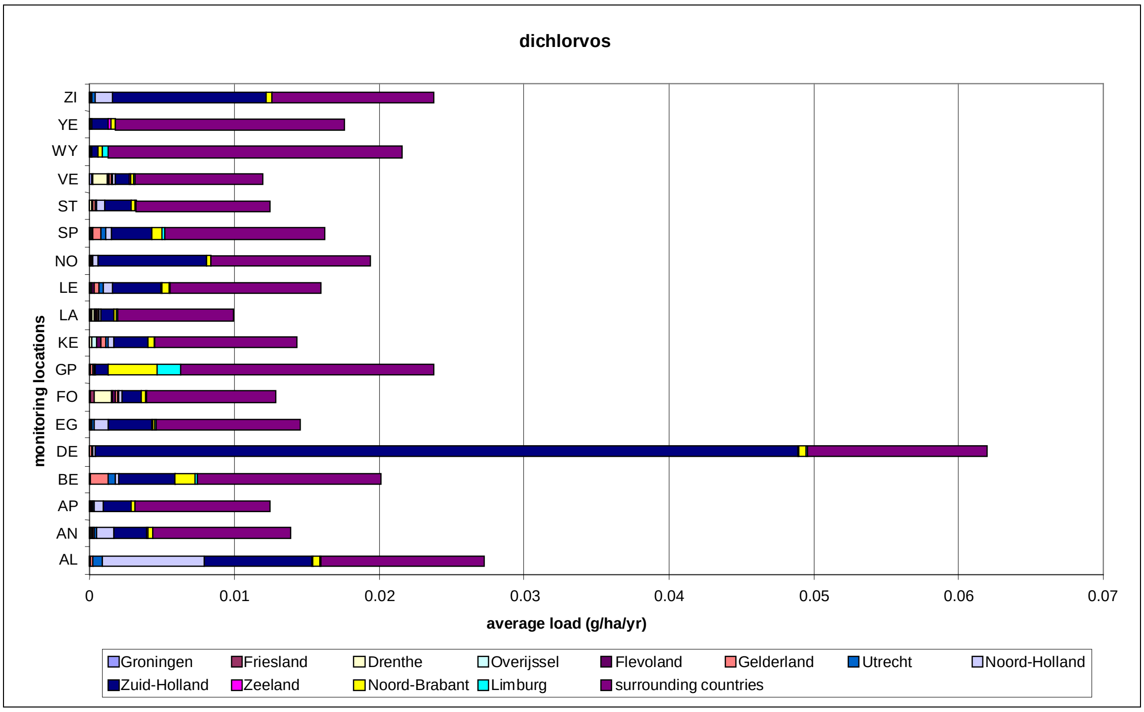

Figure VII.3 Contributions of emissions of dichlorvos per province and from surrounding countries to the average load at the monitoring locations (g/ha).

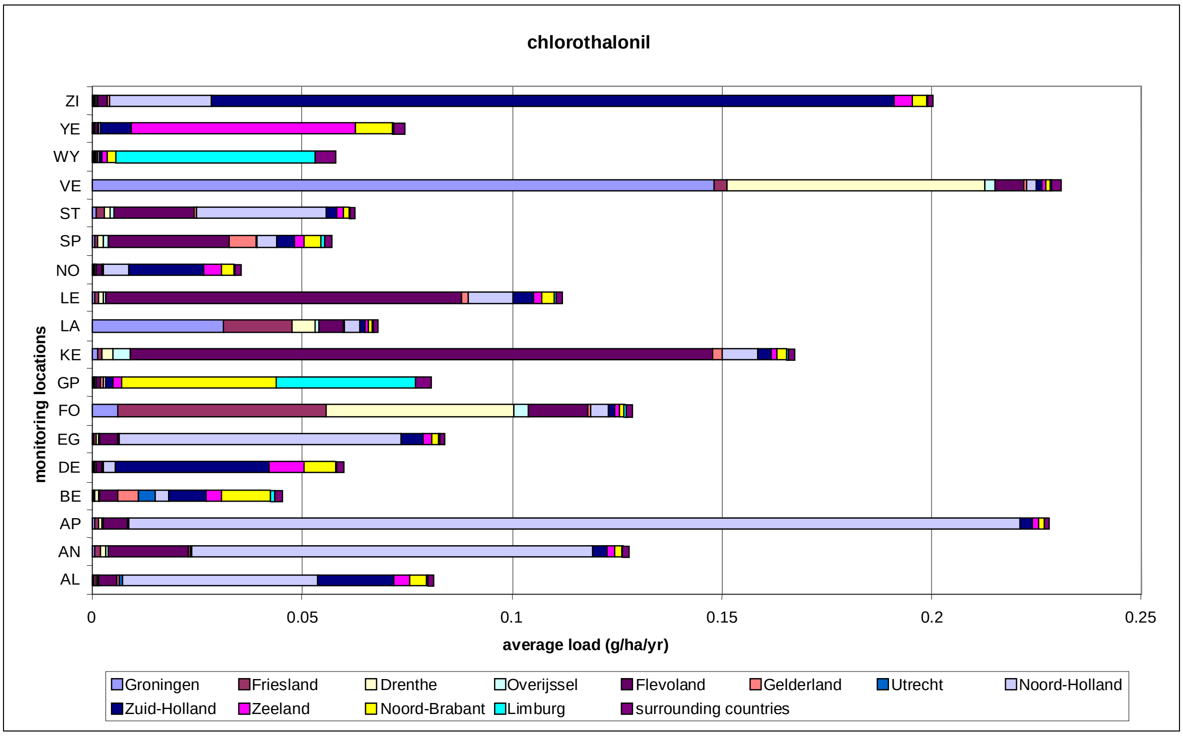

Figure VII.4 Contributions of emissions of chlorothalonil per province and from surrounding countries to the average load at the monitoring locations (g/ha).

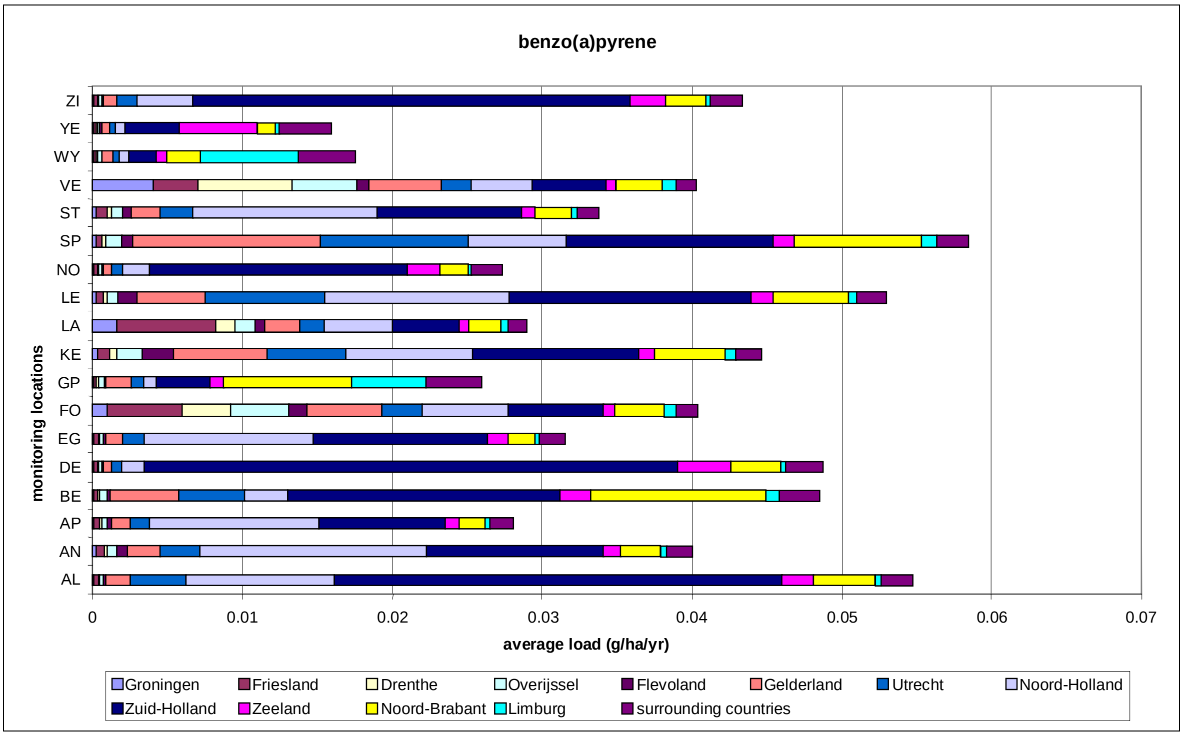

Figure VII.5 Contributions of emissions of benzo(a)pyrene per province and from surrounding countries to the average load at the monitoring locations (g/ha).

Annex VIII Concentration of some polar pesticides in precipitation

Table VIII.1 Concentration of some polar pesticiden in precipitation of a limited number of locations. Given are: substance name, the date on which sampling was stopped (which was started four weeks earlier), and the concentration of the substance in ng/l.

Substance	Date	Andijk	Delft	Fochteloer-veen	Wynands-rade
isoproturon	29-03-00	34	37	28	130
	25-04-00	41	60	no sample	no sample
	24-05-00	< 20	< 20	< 20	22
	21-06-00	< 20	< 20	28	130
	16-08-00	< 10	< 10	< 10	< 10
	13-09-00	< 10	< 10	< 10	< 10
diuron	29-3-00	< 15	< 15	< 15	< 15
	25-4-00	< 15	32	no sample	no sample
	24-5-00	25	< 15	< 15	24
	21-6-00	< 15	18	< 15	< 20
	16-8-00	< 7	< 7	< 7	< 7
	13-9-00	< 7	< 7	< 7	< 7
metoxuron	16-08-00	7	14	< 10	< 10
metabromuron	24-05-00	< 20	< 20	< 20	51
	21-06-00	51	< 20	< 20	< 25
pencycuron	24-05-00	47	27		
carbendazim	29-03-00	< 25			
	21-06-00	< 45			
	16-08-00	< 40			
	13-09-00	< 40			
chloridazon	25-04-00	31			
metamitron	24-05-00	60			