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Effects of atmospheric deposition of pesticides on terrestrial organisms in the Netherlands

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Abstract

At present there is much focus on the atmospheric dispersal of pesticides. However, there is very little known about the effects of atmospheric deposition, especially in terrestrial ecosystems. In the study described here, a start has been made to clarify the possible effects on terrestrial organisms using available data. To realise this aim a number of assumptions were necessary, which lead to uncertainties in the results. Keeping this in mind, however, we could conclude that the Maximum Permissible Concentration (MPC) for soil is exceeded for 3 out of 60 compounds with a known MPC. The so-called ‘Negligible Risk Level’ for soil is exceeded for 20 out of 60 compounds. The Negligible Risk Level (NRL) is exceeded for a number of compounds for all species groups studied (soil organisms, flying insects, birds and plants. For these species groups, the MPC is exceeded especially for plants (for 11 herbicides). Further research and refinement is needed in order to assess the risks, following from the exceeding of NRL and MPC. Data are lacking though for estimating effects of fungicides on fungi. The conclusions refer to mid-range effects. Closer to the treated areas, higher depositions may be expected - one of the aspects for further research, along with the role of the transport of airborne pesticides from abroad.

Contents

Samenvatting

In deze studie is met behulp van bestaande gegevens een schatting gemaakt van de effecten van atmosferische depositie van bestrijdingsmiddelen op terrestrische organismen. Hierbij is voor het schatten van de depositie gebruik gemaakt van berekeningen van het RIVM en van door TNO verrichte metingen. Er is uitgegaan van de *worst case* situatie dat alle middelen op één dag neerkomen. Daarentegen is de depositie gemiddeld over Nederland genomen. Lokaal, dichterbij de landbouwpercelen, kunnen dus hogere deposities voorkomen. Bij de analyse zijn alleen in Nederland toegelaten middelen betrokken. De aanvoer van deze middelen uit het buitenland is slechts zeer ten dele meegenomen. De aanvoer van niet in Nederland toegelaten middelen is buiten beschouwing gelaten, evenals het eventuele illegale gebruik, met als belangrijkste reden het ontbreken van betrouwbare gegevens.
Voor de effecten is voor de 60 middelen waarvoor een Maximum Toelaatbaar Risico (MTR) beschikbaar is, het MTR_{bodem} gebruikt. Daarnaast is voor de soortengroepen bodemorganismen, vliegende insecten, vogels en planten een schatting gemaakt van de gevoeligheid voor atmosferische depositie. Voor deze schatting zijn de laboratoriumgegevens gebruikt van toxiciteitsexperimenten waarin het 50%-effectniveau is bepaald. Van deze gegevens, meestal voor één soort beschikbaar, is vervolgens een No-Observed-Effect-Concentration afgeleid voor de vier onderscheiden soortengroepen. Hierbij is de methode gevolgd die ook wordt gebruikt voor het afleiden van een MTR voor een bepaalde soortengroep. In de meeste gevallen is hierbij een factor duizend gehanteerd. Deze NOEDL (No Observed Effect Deposition Level) kan derhalve worden gezien als een MTR voor de betreffende soortengroepen. De geschatte depositie is vervolgens vergeleken met de NOEDL's en met het MTR_{bodem}. Daarnaast is ook het VR (Verwaarloosbaar Risico) niveau in beschouwing genomen. Dit is gedefinieerd als een factor 100 onder het MTR of het NOEDL. Tot slot zijn de effecten van alle stoffen bij elkaar opgeteld ("Toxic Unit" benadering) teneinde een beeld te krijgen van het effect van gecombineerde blootstelling.
De resultaten laten zien dat het MTR_{bodem} voor drie van de 60 middelen wordt overschreden. Het VR_{bodem} wordt voor 20 van de 60 middelen overschreden.
Wanneer de depositie wordt vergeleken met het VR niveau van de onderscheiden soortengroepen, dan blijkt dat er voor alle soortengroepen voor een aantal middelen overschrijdingen worden gevonden. In totaal wordt voor 68 van de 293 middelen het VR voor minstens één van de onderzochte groepen overschreden. Overschrijdingen van het NOEDL worden vooral gevonden in het geval van hogere planten (voor 11 herbiciden). Voor effecten van fungiciden op niet-doelwit schimmels ontbreken de gegevens.
De Toxic Unit benadering laat zien dat er met name een risico voor planten aanwezig is.

Uit de resultaten blijkt dat ten gevolge van atmosferische depositie van bestrijdingsmiddelen op grotere afstand van de landbouwgebieden het VR niveau voor alle onderscheiden soortengroepen voor een aantal middelen wordt overschreden. Dit betekent dat nader onderzoek en verfijning van de methode noodzakelijk is alvorens uitspraken over mogelijk risico's kunnen worden gedaan. Het MTR niveau wordt met name voor planten overschreden. Hier is nader onderzoek noodzakelijk ten einde uitspraken te kunnen doen over het daadwerkelijk optreden van effecten van atmosferische depositie van bestrijdingsmiddelen op planten.
De conclusies hebben betrekking op gebieden op grotere afstand van de bespoten percelen. In de nabijheid van landbouwgebieden, bijvoorbeeld daar waar de Ecologische Hoofdstructuur grenst aan de landbouwgronden, kunnen hogere deposities optreden. Dit is dan ook een van de punten waarop nader onderzoek zich zou moeten richten. Ook zou in vervolgonderzoek beter naar de bijdrage uit het buitenland moeten worden gekeken.

Summary

In this report, the effects of atmospheric deposition of pesticides on terrestrial organisms are estimated from existing data. Both calculations of RIVM and measurements of pesticide deposition carried out by TNO provided input for the estimation of deposition. For the estimation of the effects it was assumed that all yearly deposition takes place in one day. On the other hand it was assumed that the deposition is spread evenly over the surface of the Netherlands. Only pesticides approved for use in the Netherlands have been taken into account. Transport from abroad has been considered only partly. Import or (illegal) use of pesticides not approved for use in the Netherlands was left out of the analyses, mainly because of a lack of reliable data.

For the effects the MPC (Maximum Permissible Concentration) for soil is used for the 60 compounds for which a MPC is known. Apart from this the sensitivity for atmospheric deposition is estimated for the species groups soil organisms, flying insects, birds and plants.

For this estimation, EC_{50} laboratory data have been used. From these data, which in most cases were only available for one species per group, a NOEC has been derived for the four species groups. The method used is equivalent to the method used to estimate a MPC for a group of species. In most of the cases, a factor 1,000 has been used for this derivation. This NOEDL (No Observed Effect Deposition Level) thus can be seen as MPC for the particular species group. The estimated deposition then was compared with the NOEDL and with the MPC for soil. Further more the NRL (Negligible Risk Level) was taken into account. The NRL is defined as 0.01 times the MPC (or NOEDL). Next, the potential effects of all active ingredients were added (“Toxic Unit” approach) in order to obtain insight in the effects of combined exposure.

The results show that the MPC_{soil} is exceeded for three out of 60 compounds. The NRL_{soil} is exceeded for 20 out of 60 compounds.

For the species groups soil organism, flying insects, birds and plants the NRL is exceeded for a number of compounds. In total the NRL is exceeded for one or more species groups for 68 out of 293 compounds. The NOEDL is exceeded especially in the case of plants (for 11 herbicides). For an estimation of effects of fungicides on fungi, data are lacking.

The Toxic Unit approach results in a potential risk for plants.

The results show that, as a consequence of atmospheric deposition of pesticides at middle distance of agricultural areas, the NRL is exceeded for a number of compounds for all species groups. This means that further research and refinement is needed for assessing the potential risks. The MPC is exceeded especially in the case of plants. Here further research is needed to assess the actual occurrence of effects of atmospheric deposition of pesticides on plants.

The conclusions refer to mid-range effects. Closer to the treated areas, for instance in areas where the Ecological Main Structure borders the agricultural land, higher depositions can be expected. This is one of the aspects for further research. Another important aspect is role of the transport of airborne pesticides from abroad.

1. Introduction

1.1 Background

At RIVM, several research projects relating to pesticides are aimed at the inventory of effects of pesticides on natural ecosystems. The project described in this report was conducted within the framework of project M/716601, Pesticide fate in the Environment, sub-project Regional Risk Assessment. Especially in terrestrial natural areas dispersal by air is of importance. At greater distance of the agricultural area, exposure to airborne pesticides probably is the only exposure route. Quantitatively, atmospheric deposition is the most important emission route (circa 9.2 % of the total use, De Nie, 2002). Based on measurements it is concluded that this emission route eventually leads to deposition of pesticides (Van Dijk and Guicherit, 1999; De Jong and Leendertse, 1999; Duyzer and Vonk, 2002). At the EU level, an EU-FOCUS workgroup has been started, with its scope on atmospheric dispersion of pesticides related to the registration of pesticides (FOCUS-AIR).

Much less attention has been paid to the possible side-effects of deposition of pesticides on organisms, populations or ecosystems. In 1998, the Dutch Health Council organised a symposium on the atmospheric dispersal of pesticides. This symposium mainly aimed at the fate of substances in the atmosphere (Van Dijk *et al.,* 1999). One part was aimed at the estimation of the effects of long-range transport on invertebrates (Van Straalen and Van Gestel, 1999). They conclude that effects are not to be expected, but as uncertainties are large, a safety factor should be applied. Other estimations of effects on plants and fungi show that the exposure to atmospheric deposition is in the order of magnitude of the NOEC (De Jong *et al.,* 1995; Kempenaar *et al.,* 1998). Based on model calculations on atmospheric deposition of herbicides, Klepper *et al.* (1998) predict that the NOEC is exceeded for about 2% of the plant species.

From the above it is concluded that pesticides do disperse via the atmosphere, and that they do deposit, which potentially causes effects. The aim of this report is to make an inventory of the potential effects of atmospheric deposition of pesticides in the Netherlands, based on existing data.

1.2 Problem definition

The main question of this study is:
Is it possible, using existing data, to estimate the occurrence of effects of atmospheric deposition of pesticides on terrestrial ecosystems?

It is expected that atmospheric deposition generally leads to low exposure to the substance, during a longer period. Therefore, in order to estimate the potential effects of this deposition on terrestrial organisms, NOEC data are needed for a variety of terrestrial organisms. These data, however, are hardly available. In addition, individual studies yield only limited amounts of data. To answer the main question, an inventory has been made of the readily available toxicity data for terrestrial organisms, and these data have been used to make an estimation of the potential effects.

2. Methods

As a starting point, the registered active ingredients of pesticides in the Netherlands at December 2001 were used (source: CTB website: http://www.ctb-wageningen.nl/). As the calculations of pesticide dispersal is based on the period 1998-2000, substances which were approved in 1998 but which were not registered in 2001, were added to the list. This resulted in a list of 293 active ingredients of pesticides (see Appendix 1). Presenting the results it will be indicated which substances are not registered at the moment. At this point it should be mentioned that some substances, although they do not have a registration now, might (temporarily, and under restrictions for use) be used due to a directive aimed at the so-called 'indispensable substances'.

For this selection of compounds, with use of data from the ISBEST-database (a database on pesticide use at the regional level, maintained by Alterra), the aerial distribution and deposition is calculated for the Netherlands as a whole, aggregated for one year. The contribution of pesticides from abroad has not been taken into account, neither is the eventual illegal use. Regional differences were left out of the analyses. Degradation during transport or after deposition was neglected as well. The aim was to obtain a first, general estimation of potential effects. The deposition for the Netherlands then was divided by the area of the Netherlands, in total 3,710,074 ha (excluding IJsselmeer and Wadden Sea, which were left out of the deposition estimations as well), to obtain the deposition per ha.

Furthermore, measured wet and dry deposition data of TNO were used (Duyzer and Vonk, 2002). These measurements included about 70 pesticides. Because these are measured values, they include pesticides used in the Netherlands, pesticides deposited after long-range/mid-range transport via the air, both legal and illegal use, and degradation in air. The data of the different collectors were transformed, using models, by TNO to a deposition for the Netherlands as a whole. Degradation in air was included in these model calculations for as far as possible on the basis of QSAR's (Quantitative Structure-Activity Relationships).

For the compounds whose registration ended in 1998, the use (and therefore the deposition) was first assumed to be zero. Because these compounds actually were used in that specific year (clearance sales and finishing of the stock), a correction has been made assuming the same correlation between use and deposition as in 1995 (the year for which the use and deposition were calculated).

Effects were calculated based on available toxicity data. For the terrestrial environment these data were available for earthworms, honeybees, birds and mammals and in the case of herbicides for plants. However, in general these data concerned EC_{50} values, derived from short-term toxicity experiments. From these data the potential effects of chronic exposure to low dosages was derived. In addition, for a limited number of pesticides an MPC (Maximum Permissible Concentration) was available, in which the available toxicity data were incorporated.

To estimate the effects of atmospheric deposition of the selected pesticides, first a comparison with the MPC was made. Next, for each group of organisms a NOEC for atmospheric deposition was calculated – further referred to as NOEDL (No Observed Effect Deposition Level). Ultimately, the lowest available NOEDL was used to estimate the effects on the terrestrial ecosystem.

As atmospheric deposition will consist of a mixture of pesticides, the combined effect of the different pesticides was studied. Here, the 'Toxic Unit' approach was followed, assuming concentration additivity. In this case, the EC_{50} value (for plants EC_{25}) was used, and for all compounds the PEC/EC_{50} ratio was calculated and added. If this results in a value >1, the combined exposure of all pesticides in atmospheric deposition would result in exceeding of the EC_{50}. Again, it was assumed that in the field, all compounds are deposited at once, thus indicating a worst case situation. On the other hand, it was assumed that the compounds are evenly spread over the Netherlands, giving a good picture of a background exposure. Locally, however, depositions could be much higher.

2.1 MPC

As a first step, the deposition data were compared to the available MPC data for soil organisms. The MPC is set at a level that should protect all species in ecosystems from adverse effects of a substance (VROM, 1990). Pragmatically, a cut-off value is set at the fifth percentile if a species sensitivity distribution of NOECs is used in the refined effect assessment. This is the Hazardous Concentration for 5% of the species, the HC_5^{NOEC} (Van de Meent, 1990). These data were gathered from Crommentuijn *et al.* (1997). Then it was checked if other or more recent MPCs had been derived within the framework of the project 'Setting Integrated Environmental Quality Standards'; when this was the case, the latter MPCs were used. MPC values for lindane and carbofuran were added from Verbruggen *et al.* (2001). With this procedure, an MPC value for soil could be retrieved for 60 of the 293 selected compounds. In addition to the MPC, a level is defined at which effects should be negligible: the Negligible Risk Level (NRL), defined as 0.01 * MPC. This factor is applied to take into account the possible combined effects of the many substances encountered in the environment (VROM, 1989).

For surface water, more MPC values were available than for soil. Where they were lacking, so-called 'indicative MPCs' could be derived. Deriving MPCs for soil from these values is feasible, but in the framework of this project, this was considered too time consuming.

The MPC values found are expressed as μg per kg soil, assuming a standard soil with an organic matter content of 10%, a clay content of 25%, and a dry matter content of 1.4 g per cm^3. It is assumed that all pesticides that deposit are evenly spread through the top 5 cm of the soil, and that they remain available there for exposure to non-target organisms. In order to compare the MPC values with deposition, these values were expressed as deposition in g per ha, by multiplying the MPC values (expressed in mg per kg) by 700.

2.2 Specific groups of organisms

For the terrestrial organism groups – vascular plants, earthworms, honeybees and birds – toxicity data were obtained through the following procedure:

1. If the substance was present in the new RIVM eTOXbase (currently under construction), data from this database were used. This database contains among others data from the pesticide dossiers.
2. If data were lacking in this database, data from the Environmental Indicator of RIVM were used for birds and earthworms. These data mainly originate from the Dutch

registration dossiers, but other sources were used as well, such as the German dossiers for earthworms.

3. When no data were found at all, it was checked whether these data were available in public literature, such as the Pesticide Manual and the Handbook pesticide use and environmental effects.
4. At the end the Ecotox database of EPA (Internet) was searched for additional data. Especially for plants data were obtained from this database.

In general, there is rather a shortage of data than a surplus. In some cases, however, several toxicity data were found for a specific combination of one species and one active ingredient. When these data were related to different endpoints, the value for the most sensitive endpoint was chosen. When the same endpoint was studied, the geometric mean was used. When different life stages were studied, again the value for the most sensitive life stage was used for this study.

The problem arises that generally, EC_{50} values (for plants EC_{25}) are more readily available than NOEC values, while the expected concentrations of the deposition of pesticides further away from the treated areas are in the order of the NOEC rather than the EC_{50}. In order to overcome this drawback, NOEDL values were derived from the EC_{50} values, following procedures for deriving a MPC as described by Traas (2001); see further below at the specific species groups. For plants, Jager and Traas (2001) argue that data from the EPA database are not suited for deriving a NOEDL. Due to the lack of alternatives and other data, for this study the data from the EPA database were used.

2.2.1 Earthworms

In the case of earthworms, toxicity data were available for a relatively large part of the compounds (127 of the 293 selected active ingredients). Because data for earthworms were available in a number of cases where an MPC was lacking, the earthworm data were compared with the estimated exposure. For calculating exposure, a mean density for agricultural soil of 1.4 g per cm^3 was taken (source: EPPO guideline, mean density of agricultural soil). A NOEDL value has to be expressed as exposure in g per ha. This means that the NOEDL has to be multiplied with 700 (1.4 g per cm^3 = 7 g per cm^2 assuming a dispersal over 5 cm soil depth, this is $7 * 10^5$ kg per ha. Thus: a NOEDL value = 1 mg per kg would be found at a deposition of 700 g per ha).

The available data almost exclusively consisted of EC_{50} values. In accordance with Traas (2001), an extrapolation factor of 1,000 has been applied to estimate the NOEDL from the EC_{50} for soil organisms. (The explanation for the factor 1,000 is: a factor 10 for the extrapolation from EC_{50} to NOEC; a factor 10 for the translation from acute to chronic; and a factor 10 for the extrapolation from one species to other species).

Thus, the factor used for the translation of the EC_{50} for earthworms, expressed as mg per kg of soil, into a NOEDL expressed as g per ha is $0.7 * EC_{50}$. In this way the NOEDL for soil organisms was estimated, based on the data for one earthworm species *Eisenia fetida*, for which most data were available.

2.2.2 Honeybees

In the case of honeybees, data are available for two exposure routes: contact and oral. An estimation of the exposure of honeybees via food due to atmospheric deposition is deemed less useful, because of the many uncertainties. For direct exposure, it is deemed possible to make an estimation of potential exposure and effects. In toxicity tests, honeybees are directly exposed to a compound and a LD_{50} value is estimated in µg per bee. Analogous to the method used for earthworms, a factor 1,000 is used for the extrapolation of the LD_{50} for honeybees to a NOEDL for flying insects (NOEDL = LD_{50}/1000). Toxicity data of *Apis mellifera* were used. Analogous to Canton *et al.* (1990), it is assumed that a female bee worker has a body surface of 0.5 cm^2. This means that exposure of one bee, at a dosage of 1 g per ha is equal to 5 * 10^{-9} g. To translate the LD_{50}, expressed in µg per bee, to a NOEDL in g per ha, the LC_{50} is multiplied by 5 (5 * 10^{-9} (surface), divided by 10^6 (µg to gram) and 1,000 (LD_{50} to NOEDL)). Following this method, a NOEDL could be calculated for 168 compounds. For about 50 active ingredients, apart from the deposition also the concentration in air was measured by TNO (Duyzer and Vonk, 2002). However, extrapolation of these data to contact exposure of honeybees seemed not feasible, given the limited amount of data currently available.

2.2.3 Plants

For plants, only toxicity data were used from experiments in which plants were exposed by direct spraying. Experiments, in which plants or seeds were directly exposed to a compound, for instance by dipping, were left out of account. The extrapolation of this kind of data to atmospheric deposition would bring about too much uncertainty, and it would be difficult to get a realistic extrapolation to atmospheric exposure. For plants, the EC_{25} value was divided by 3 to get an indication of the NOEDL. This factor is underpinned by the relation of EC_{25} and the NOEC based on a database of the pesticide industry (2.38 (n=48) for juvenile plants and 1.67 (n=40) for seedlings). Next, a factor 10 is used for the extrapolation from acute to chronic exposure. If data were available for 1, 2 or 3 species, a factor 10 was used for the translation to all species. If data were available for more than 3 species, the HC_5 (Hazardous Concentration for 5% of the species) is calculated. Where different endpoints were studied, the most sensitive endpoint was chosen. When more test results were available for the same species and parameter, the geometric mean was used.

2.2.4 Birds

In order to obtain an indication of the possible effects of atmospheric deposition of pesticides on birds and mammals, a realistic worst case scenario was followed. For birds in risk assessment a number of different scenario's exist (EU-SANCO, 2002). A long-term exposure of wheat shoot eating birds served as a starting point. Wheat shoots result in the risk-assessment in the highest levels of a compound per quantity of food. Furthermore, it was assumed that the birds ate no other food. For calculation of the exposure, a so-called 'shortcut factor' of 22 was used (EU-SANCO, 2002). With this factor, the Daily Dietary Dose (DDD) was calculated in the following way: DDD = shortcut value * application rate in kg per ha [mg per kg Body Weight (BW) per day]. The application rate in kg per ha is calculated by dividing the deposition in g per ha by 1,000. The resulting DDD was directly compared to the toxicity data for birds, expressed in mg per kg BW. These toxicity data were derived from LD_{50} values by applying a factor 1,000 for a NOEC for all bird species, in accordance with Traas (2001). When data were available for more species, the geometric mean was used.

2.2.5 ECO-ter

Finally, the data were used to indicate any effect on the terrestrial ecosystem (ECO-ter). For this aim, the lowest of all values for the above mentioned species groups were taken.

3. Results

3.1 Exceeding MPC

The exceeding of the MPC_{soil} is shown in figure 1 for those compounds for which the quotient of deposition and MPC >0.01. The results show that for three active ingredients (dichlorvos, metam-sodium[1] and propoxur), the estimated deposition exceeds the MPC. For 20 compounds the quotient exceeds 0.01. This is the Negligible Risk Level. This result is found for 20 of the 60 compounds for which a MPC was available.

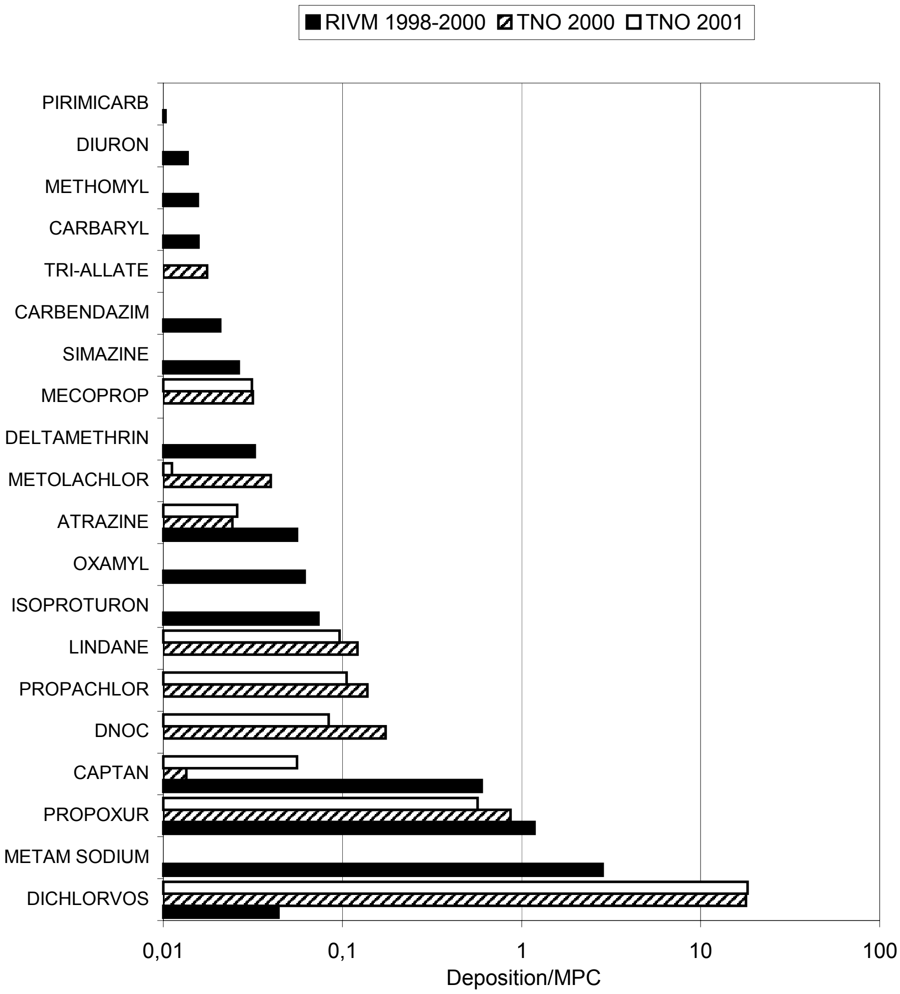

Figure 1 Atmospheric deposition of active ingredients versus the MPC_{soil}

[1] In practice, metam-sodium will immediately be transformed to its active component methylisothiocyanate (MITC). Because it is applied as metam-sodium, the term metam-sodium is used.

3.2 Soil invertebrates

The active ingredients showing a ratio > 0.01 for deposition versus the estimated NOEDL for soil invertebrates are shown in figure 2. The NOEDL has been calculated for 127 active ingredients. From figure 2, it appears that the NOEDL for soil invertebrates is not exceeded for any active ingredient. For 7 active ingredients the 0.01 level is exceeded; for the 0.01 level an analogy with the Negligible Risk level is present.

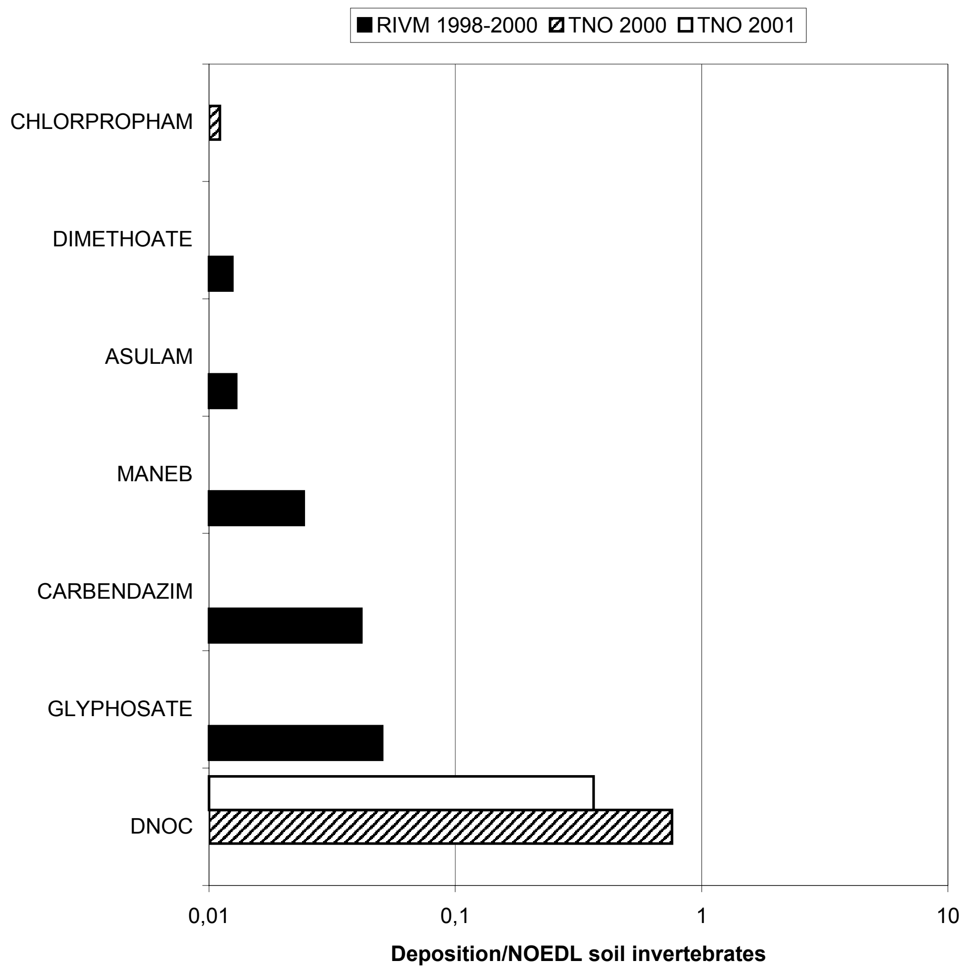

Figure 2 Atmospheric deposition of active ingredients versus the NOEDL for soil invertebrates caused by atmospheric deposition

3.3 Flying insects

The active ingredients with a ratio >0.01 between deposition and NOEDL for flying insects are shown in figure 3. The NOEDL has been calculated for 168 active ingredients. Figure 3 shows that the NOEDL is not exceeded. For five compounds the quotient exceeds 0.1. For 19 active ingredients the 0.01 level is exceeded.

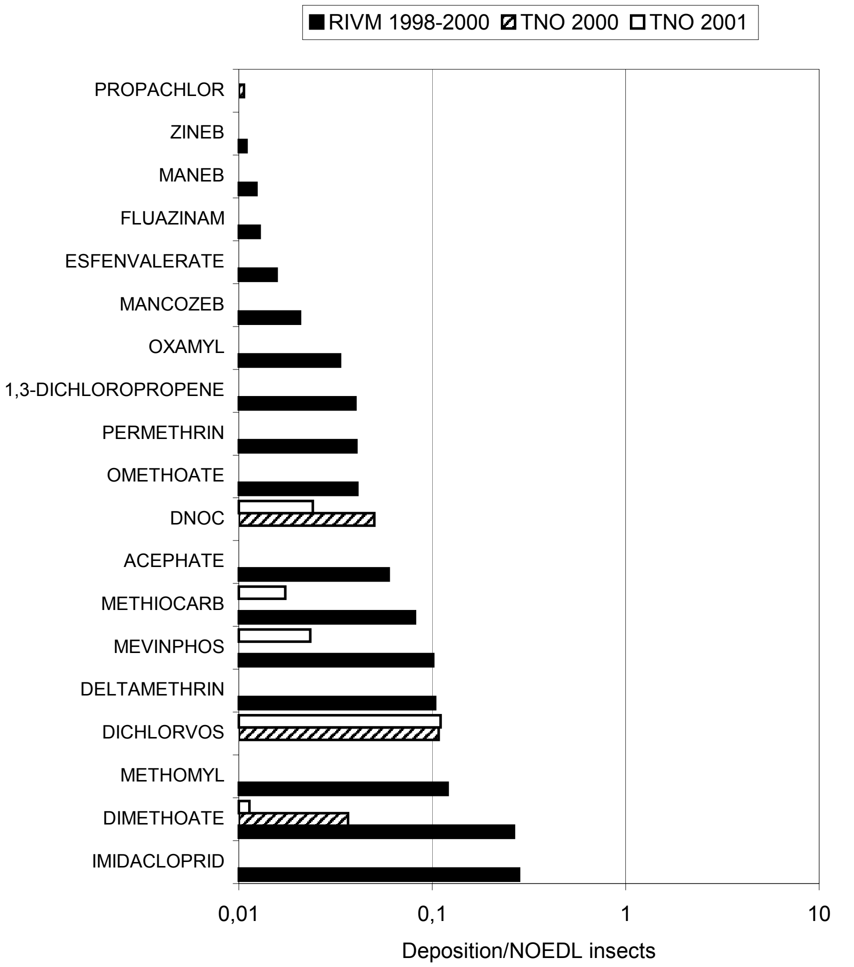

Figure 3 Atmospheric deposition of active ingredients versus the NOEDL for flying insects

3.4 Plants

The active ingredients with a ratio >0.01 between deposition and NOEDL for plants are shown in figure 4. The NOEDL was estimated for 36 herbicides. Figure 4[2] shows that for 10 herbicides the NOEDL is exceeded, and for 30 herbicides the 0.01 level is exceeded.

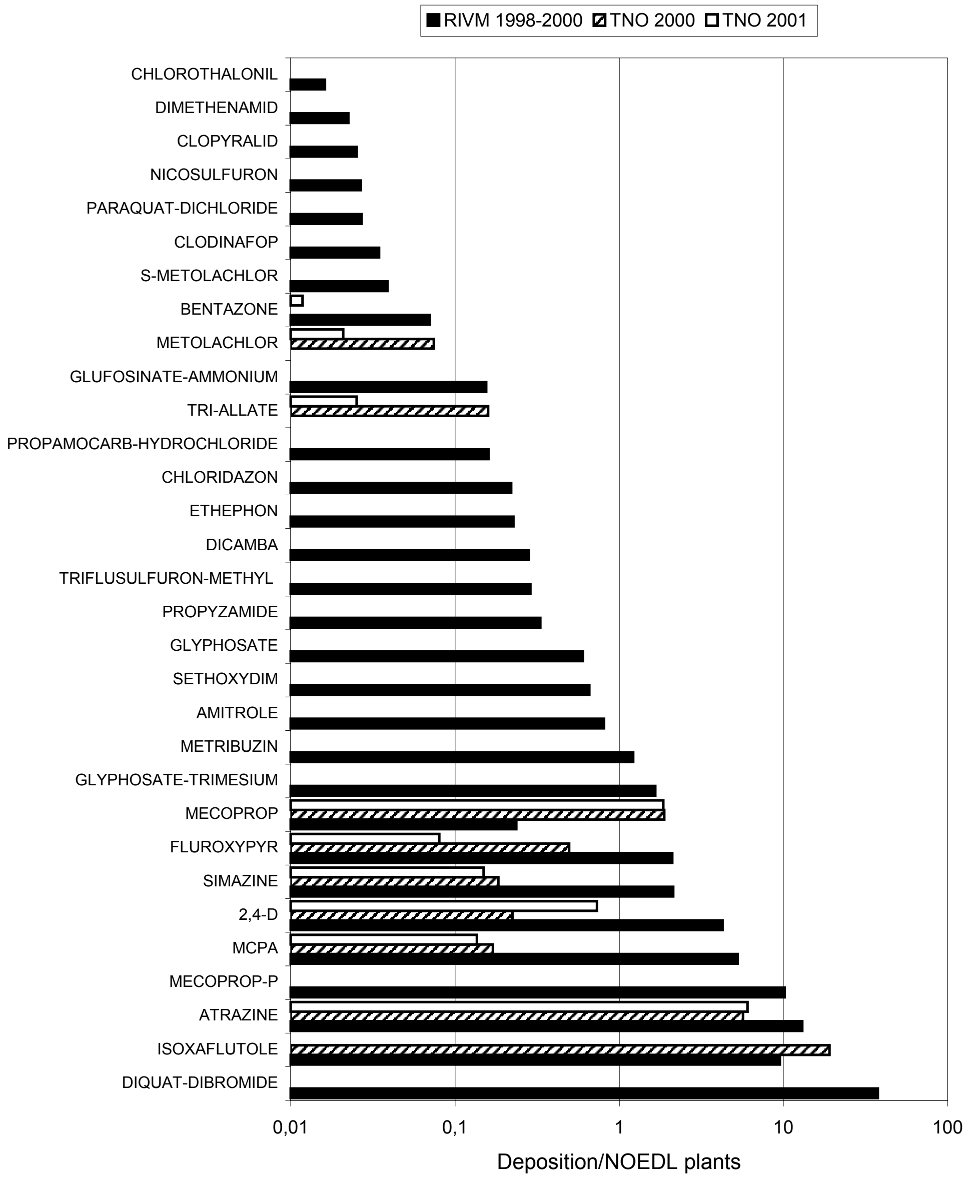

Figure 4 Atmospheric deposition of active ingredients versus the NOEDL for plants

3.5 Birds

The active ingredients with a ratio >0.01 between deposition and NOEDL for birds are shown in figure 5. The NOEDL was calculated for 226 active ingredients. From figure 4 it is clear that for DNOC the NOEDL for birds is exceeded. For 12 active ingredients the quotient

[2] Mecoprop has been replaced by Mecoprop-P; since for both compounds different toxicity data were available, compounds are presented separately here.

between deposition and NOEDL exceeds 0.1. For 37 active ingredients the Negligible Risk level is exceeded.

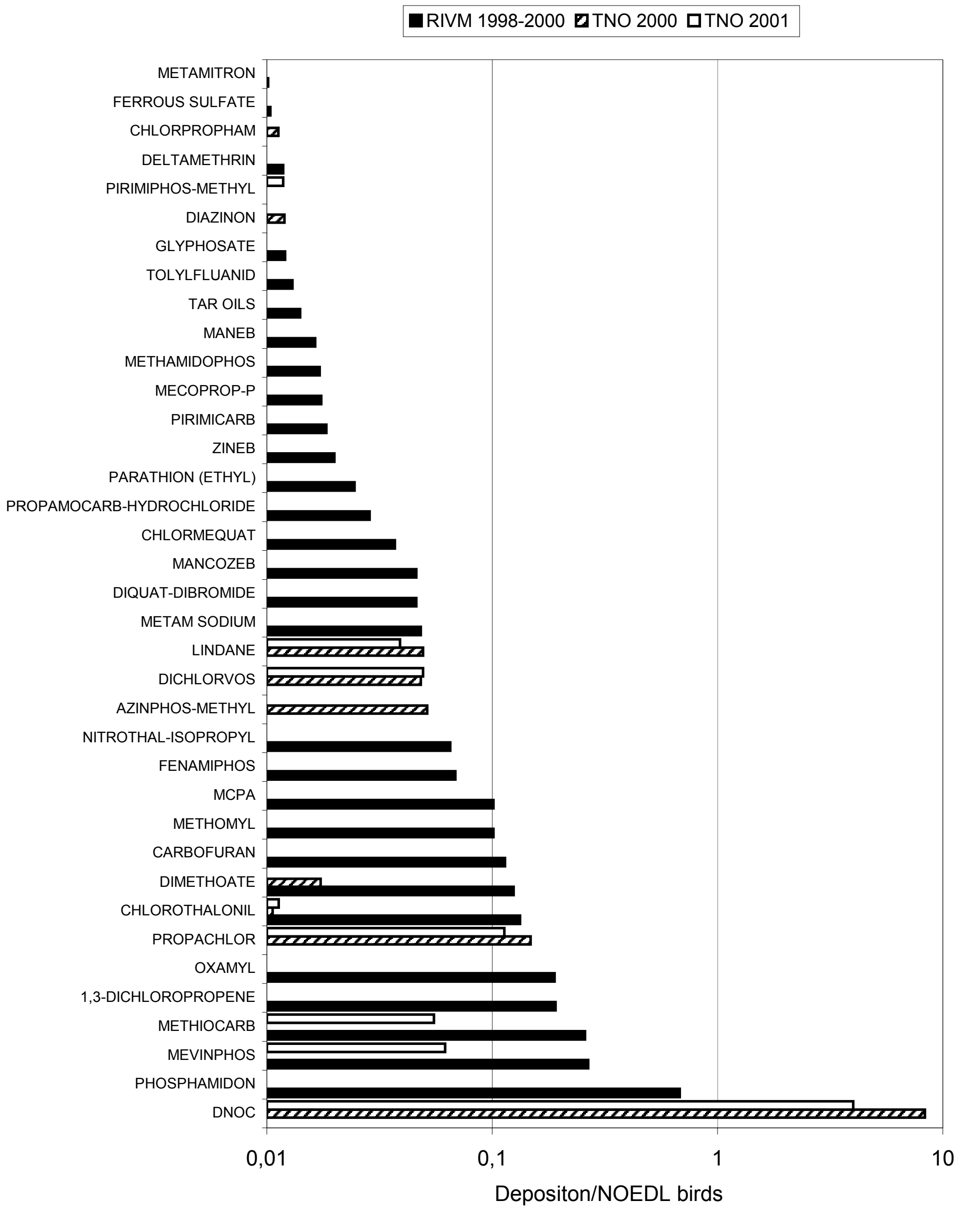

Figure 5 Atmospheric deposition of active ingredients versus the NOEDL for birds

3.6 ECO-ter

Figure 6 shows all active ingredients exceeding a ratio of 0.01 between deposition and NOEDL for at least one of the organism groups studied. For every active ingredient the organism group with the highest ratio is chosen. In this way, a NOEDL could be estimated for 236 of 293 active ingredients studied. From figure 6, it is clear that the NOEDL is exceeded for 11 of the 293 active ingredients under investigation. For 24 active ingredients the ratio between deposition and NOEDL is between 0.1 and 1, and for 33 active ingredients between 0.01 and 0.1.

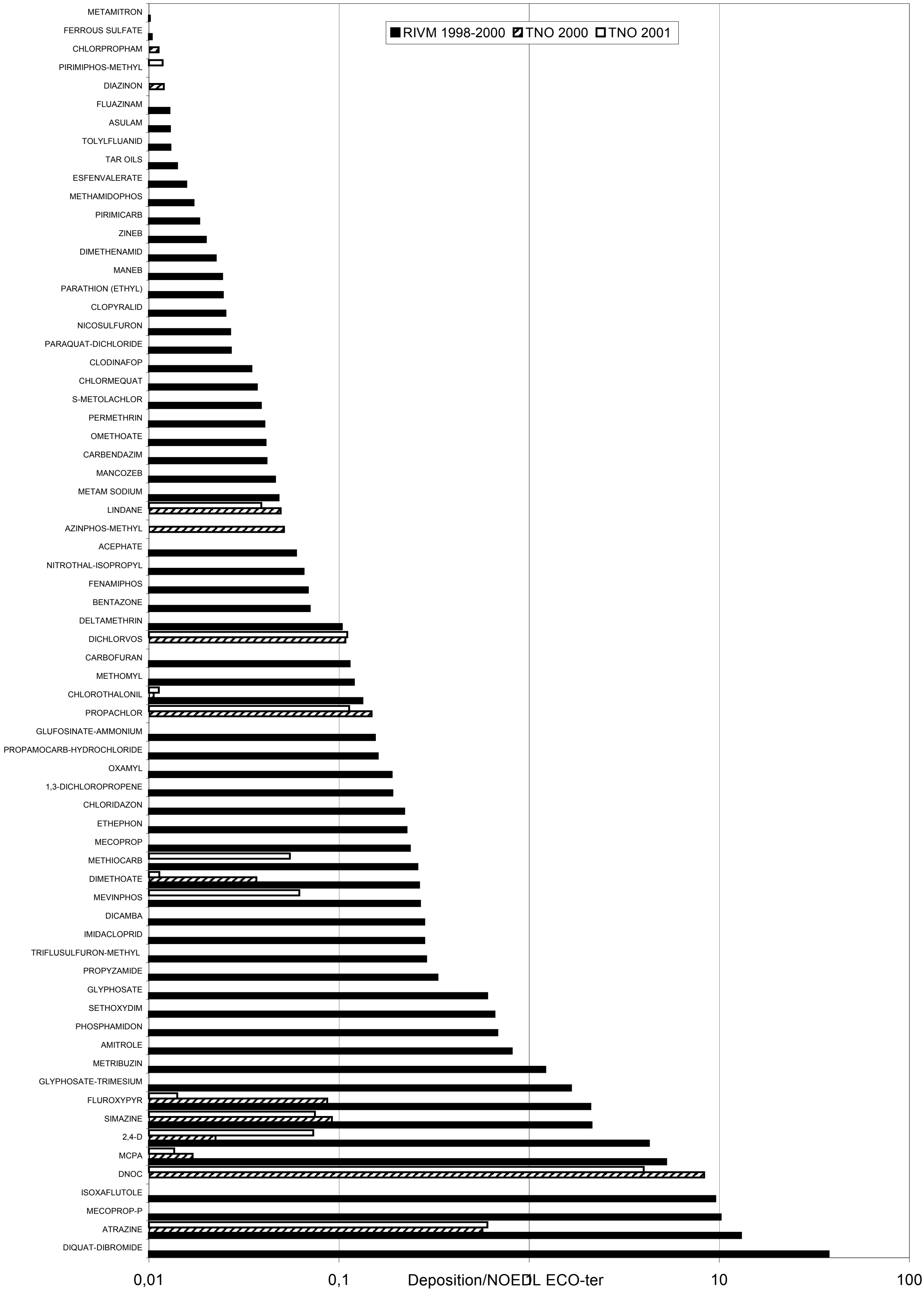

Figure 6 Atmospheric deposition of active ingredients versus the NOEDL for terrestrial organisms

3.7 Toxic Units

For the Toxic Unit approach, the average deposition data for 2000 and 2001 (from TNO) were used. These data were available for about 50 active ingredients. For the other compounds, the data resulting from the RIVM model calculations were used. Toxic Units are based upon an exceeding of the 50% effect levels (for plants 25%). The results are shown in table 1.

Table 1 Cumulative Toxic Units for the effects of atmospheric deposition

Species group	Σ Toxic Units EC_{50} all active ingredients	Σ Toxic Units EC_{50} active ingredients 2002
Soil invertebrates	0.00078	0.00020
Flying insects	0.0011	0.00095
Birds	0.0084	0.0011
Plants	0.254*	0.245*

* Based on EC_{25} values

From table 1 it can be concluded that in none of the cases, the figure for the cumulative Toxic Units exceeds the value of 1. In the case of plants, however, the value is of such a magnitude that some effects could be expected. When interpreting the results, however, two uncertainty factors should be kept in mind: at the one hand, a worst-case situation was simulated, by assuming that all deposition occurs at one day. At the other hand, as a consequence of the way of measuring, the calculated deposition figures could be seen as background concentrations. Locally, for instance downwind of agricultural areas, considerably higher concentrations may be expected.

In the right column of table 1 it is assumed that active ingredients that are no longer approved in the Netherlands are no longer present in the deposition. These data should be considered with care, for several reasons. To start with, the use of some of the not-approved compounds is still temporarily authorised, for reasons of 'indispensability for agriculture'. Moreover, as the route through the air does not stop at country borders, import is to be expected. In addition, there is the possibility of illegal use. Furthermore, it is probable that the use of compounds that are not approved anymore, will be replaced by the use of other compounds (existing or new), leading to a higher use of those compounds. When the use of a compound is stopped because of its toxicity for a certain group of organisms, it may be assumed that the replacing compounds will be less toxic, so it is realistic to assume that toxic pressure will diminish. All these aspects are left out of consideration when calculating column 2 of table 1. And finally, it was assumed that the Dutch (and European) policy is 100% effective.

With the assumptions mentioned, toxic pressure diminishes for soil invertebrates and birds. Dutch policy thus appears to be principally effective, but actual effectiveness will depend on the above-mentioned factors: the use in neighbouring countries, implementation of policy and the nature of replacing compounds. For plants, toxic pressure has hardly diminished. This seems to be a logical consequence of the lack of attention for non-target plants in the registration process.

4. Discussion

The effects of atmospheric deposition on terrestrial organisms had to be estimated from existing data. Most of these data were not generated for a risk assessment as described in this report. The available data therefore had to be extrapolated. The toxicity data used were based on single, acute exposure. These data were extrapolated to chronic effects of low dosages. The use of a factor 1,000 for the extrapolation of 50% effect levels to a NOEDL is based on safety factors as used for standard setting. For most compounds the use of these safety factors can be seen as a worst case estimation.

For deposition, the total estimated deposition of one year was used, and it was assumed that this amount was available at one moment. The deposition data as measured by TNO are the result of use in the Netherlands, as well as import and export from abroad and degradation in air. The RIVM calculations only include use in the Netherlands, and decomposition has not been taken into account. In neither of the datasets used, decomposition after deposition is accounted for. In practice, deposition will take place over a certain time span, and degradation will occur. Ignoring degradation and the time factor thus leads to an overestimation of the actual concentrations. On the other hand, these deposition data may be regarded as background concentrations, to be expected further away of the areas of use. TNO selected the locations in such a way as to exclude direct influence of local pesticide use. Closer to agricultural fields, higher depositions will occur. Given the large areas with pesticide use in the Netherlands (2,300,000 ha agricultural area, about half of the Netherlands, of which 800,000 ha arable land with intensive pesticide use, about one fifth of the Netherlands (IJsselmeer and Wadden sea excluded; CBS statline, data 2000), a considerable area may be exposed to higher depositions.

Furthermore, the use of compounds in the surrounding countries is only partly included in the study. In the model, only the Dutch use is included. The measured values include import from abroad, but these measurements were only available for a limited number of compounds (40 of the compounds considered in this study). Compounds that are not approved in the Netherlands were not included; neither is illegal use.

The analysis comprised all 293 active ingredients approved for use in the Netherlands. TNO measurements inherently include import from abroad. In this analysis, no special attention is paid to active ingredients that are banned in the Netherlands but approved in surrounding countries. The share of active ingredients from abroad in the total toxic pressure, therefore, is unknown.

4.1 MPC-soil

The MPC value is aimed at protecting organisms against the adverse effects of substances. The MPC_{soil} calculated, therefore, is aimed at protecting soil organisms against (active ingredients of) pesticides. Initially, the starting point was the protection of the soil *within* the treated parcel. In the case of atmospheric transport and deposition in remote areas, much lower depositions are found. The standard soil is selected to be representative for agricultural soils. The soil in nature areas can differ substantially from agricultural soils. Not only may pH and organic matter content deviate, but in nature areas also no ploughing and fertilising takes place. These factors may result in considerable differences in decomposition of a compound compared to a standard soil. Differences in dry matter content of soils have direct consequences for exposure expressed in mg per ha. Organic matter content is assumed to be inversely proportional to toxicity (Traas, 2001). This means that on a vulnerable soil with for instance an organic matter content of 2% (instead of 10% as in standard soil), toxicity is five times as high (10: 2), and the MPC will be exceeded with a five times lower dosage compared to standard soil. For other factors, such as clay content, such relations are not known. Furthermore, it is well possible that in a nature area the distribution of the deposition into the soil differs from a standard soil. It was assumed that the compound spreads evenly over the top 5 cm of the soil. When, for instance, a compound spreads into a top layer of less than 5 cm, this leads to a higher concentration. On the other hand, in that case, a smaller part of the soil is contaminated.

For 20 out of 60 active ingredients for which a MPC_{soil} is known, the Negligible Risk Level is exceeded. Since a priori, there is no reason to assume that the other compounds should have lower or higher MPC values, in practice the NRL could be exceeded for a higher number of active ingredients.

4.2 Earthworms

The soil type is one of the determining factors determining soil density and the relationship between deposition and the concentration in the soil. The presence of earthworms, however, is also determined by the composition of the soil; the organic matter content, then again, influences the availability of the compounds. Given the other uncertainties, these factors will be of less importance for the risk assessment of atmospheric deposition. Given the fact that earthworms are not very sensitive, even if they are exposed to the actual field dosage, it can be questioned whether they are the most suitable parameter for assessing effects of deposition. An alternative would be to include epigeic (on the soil living) organisms.

4.3 Honeybees

The estimation of the exposure of honeybees to atmospheric deposition has been based on many assumptions, leaving many uncertainties. However, since a worst-case scenario was followed, it is unlikely that effects of atmospheric deposition will occur in practice. The inclusion of herbivorous insects, which would be exposed by food, would nevertheless give a better picture of the potential risks on flying insects.

4.4 Plants

The NOEDL for growth and development is exceeded for a number of individual plant species. The results are in accordance with field studies with glufosinate-ammonium (De Snoo *et al.*, 2001) in which very low dosages (2% of field dosage) induced short-term phytotoxic effects. Furthermore, the Toxic Units value for 25% effect indicates that effects may be expected. The estimation of the effects of deposition is derived from direct spray exposure data. This exposure route differs from practice, in which dry deposition will be an important route as well as exposure to concentrations in air. Research aimed at this route is currently being carried out, among others by Plant Research International (WUR). This research is restricted due to a lack of research data.

Mathiassen and Kudsk (2001) studied the possible effects of mecoprop-P deposition in rain on a number of plant species. In this study the ED_{10} was used to estimate the NOEL (as dose required reducing fresh or dry weight by 10%). They concluded that effects are not to be expected, since the yearly deposition rates are one third of the ED_{10} of the most susceptible plant species. When comparing their results with the results of this study, two things catch the eye. The deposition rate found in the Netherlands is twice as high. And second, in the present study, an extra factor of 10 is applied to simulate chronic exposure, and a HC_5 value is calculated for the resulting data. The consequence is that compared to Mathiassen and Kudsk, in the present study, a higher deposition is weighed against a lower NOEDL, leading to a considerable exceeding of the NOEDL. The higher deposition in the Netherlands can have different causes, for instance higher use, more import from other countries etc. Since data for chronic exposure are lacking the factor 10 was applied as an extra safety factor, taking the precautionary principle as a starting point. Using the HC_5 value is a statistical method that aims to protect 95% of the species, thus following Dutch environmental policy. Applying the HC_5 method to the data of Mathiassen results in a value of 0.39 g a.i./ha, compared to the lowest EC_{10} value of 0.5 g a.i./ha, found for *Capsella bursa pastoris.* Applying of the factor 10, however, results in a NOEDL of 0.039 g a.i./ha. For the present study the extrapolation factor 10 was applied because of the lack of information about effects of chronic exposure.

4.5 Birds

A worst-case scenario was followed to assess the risk of atmospheric deposition for birds. Differentiation in food choice has not been taken into account, and neither has avoidance. Considering the low dosages involved, the latter is not very likely to occur. Given the worst-case assumptions, effects of atmospheric deposition are not to be expected.

5. Conclusions and recommendations

5.1 Conclusions

Before drawing conclusions, it has to be stated that the estimations are based on many assumptions. This could lead to a considerable uncertainty in the results. The assumptions have been made in relation to both estimation of the deposition and the effects. To make it possible to calculate the effects, many simplifications were necessary when calculating the effects, at some points leading to possible overestimation; in other cases to possible underestimation of effects. These uncertainty factors and their possible influence on the effects have been discussed extensively in chapter 4.

The aim of the study was to estimate the possible effects of the atmospheric deposition of agricultural pesticides on terrestrial ecosystems, using existing data. The main conclusion is that the Negligible Risk Level is exceeded for a number of compounds for all species groups studied (soil organisms, flying insects, birds and plants) and for the NRL_{soil}. In the case of vascular plants the NOEDL, comparable to the Maximum Permissible Concentration is exceeded as well (for 11 herbicides). The calculations show that the MPC is not exceeded in the case of flying insects and soil organisms. In the case of birds one compound (DNOC) shows a clear exceeding of the NOEDL. In the Netherlands, the use of this compound is not approved at the moment. The MPC_{soil} is exceeded for 3 compounds. For conclusions about possible effects of fungicides on non-target fungi, data are lacking.

Exceeding of MPC_{soil}
The results show that the Negligible Risk Level (NRL) is exceeded for 20 of 60 compounds for which a NRL was available (of which 9 are not approved in the Netherlands at the moment). For three compounds (dichlorvos, metam-sodium (MITC) and propoxur), a risk can be expected for soil organisms, based on the estimated exceeding of the MPC. Locally higher depositions were not included, and neither has the combined use of different compounds been taken into account. Locally, and under certain soil conditions, an exceeding of the MPC could occur. Further away of the agricultural areas, the risk of large-scale exceeding of the MPC seems limited.

Soil invertebrates
Based on the available data and the assumptions and starting points mentioned before for 6 compounds, the 0.01 value is exceeded. For one compound only (DNOC), the PEC/NOEDL quotient exceeds 0.1. This compound is not approved at the moment. Based on these results, large-scale acute effects of atmospheric deposition of pesticides on soil invertebrates are not to be expected.

Flying insects
For a total of 19 active ingredients (of which 15 are approved in the Netherlands) the 0.01 level (NRL) is exceeded. For none of the active ingredients in the study, the NOEDL is exceeded. For an interpretation of the effects of the exceeding of the NRL further research and refinement is needed.

Plants
For terrestrial vascular plants, the NOEDL is exceeded for eleven herbicides (metribuzin, glyphosate-trimesium, mecoprop, fluroxypyr, simazine, 2,4-D, MCPA, mecoprop-P, atrazine, isoxafutole and diquat-dibromide). Eight of these are at this moment approved in the Netherlands. In total, the 0.01 level (NRL) is exceeded for 37 herbicides (of which 24 still approved). The results indicate that effects of atmospheric deposition of herbicides on vascular plants cannot be excluded. The effect of excluding the compounds that are no longer approved is limited. This was expected, since side-effects on terrestrial vascular plants are not included in the registration procedure.

Birds
For birds, the NOEDL is only exceeded for DNOC. This compound is not approved in the Netherlands at the moment. For a total of 37 active ingredients (25 still approved), the 0.01 level is exceeded. Based on these results further refinement of the method is needed to exclude effects on birds on larger distances of the treated areas. Closer to the parcels, the atmospheric deposition might cause such concentrations that birds could be affected.

ECO-ter
When all results are combined, the NOEDL is exceeded for 9 of the 293 active ingredients in the study, of which 7 are presently approved in the Netherlands. For a total of 66 active ingredients, the 0.01-level (NRL) is exceeded for at least one of the groups studied.

TOXIC UNITS
The toxic unit approach is used to indicate combined effects. This approach is based on EC_{50} values. The results show that mainly in the case of plants a risk appears to be present.

5.2 Recommendations

The results of this study show that a-priori, only effects on plants are likely. It is assumed that the compounds are evenly spread over the country. Since in practice, this is not the case, it is recommended that further studies be focused on actual depositions, accounting for more realistic time intervals and regional distributions. It then will become clear whether exceeding of the NOEDL is to be expected. A way to present this kind of results for instance could be to map the exceeding or to present the percentage of nature areas in which the Negligible Risk Level or the MPC is exceeded.

In further studies on the deposition of active ingredients in the Netherlands, import from surrounding countries should be included. For these estimations compounds which are approved in the Netherlands as well as compounds which are only approved in the surrounding countries should be included. An inventory of the import could be rather difficult, since for most countries, data on the use of active ingredients is not available in detail.

Furthermore, decomposition in air and in soil should be included to obtain more realistic exposure data.

In the case of plants, effects are to be expected. Further research aimed at the nature and extent of these effects is desirable. For the effect of fungicides on non-target fungi, a literature study should be conducted to get an overview of the available information and the potential risks.

In this study major assumptions and simplifications were necessary in order to be able to estimate potential effects of atmospheric deposition of pesticides. The results indicate that refinement of the procedure is necessary. We therefore recommend to improve and refine deposition estimations (both spatial and temporal resolution should be enlarged). Furthermore we recommend to fill datagaps, especially for fungi, so that MPC and NRL can be estimated without extrapolation factors.

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Appendix 1 Selected active ingredients

GR=Growth Regulator, H=Herbicide, I=Insecicide, F=Fungicide, A=Acaricide, R=Rodenticide, L=leaf killing; NA = not approved at the moment.

NAME	MODE OF ACTION
2-(1-NAPHTHYL)ACETAMIDE	GR
1-NAPHTHYLACETIC ACID	GR
2,4-D	H
INDOL-3-YLACETIC ACID	GR
4-INDOL-3-YLBUTYRIC ACID	GR
ABAMECTIN	I
ACEPHATE	I
ACLONIFEN	H,L
ALDICARB	I
ALKYLDIMETHYLBENZYL-AMMONIUMCHLORIDE	
ALKYLDIMETHYLETHYLBENZYL-AMMONIUMCHLORIDE	ALG
PHOSPHINE	R
ALUMINIUM-SULFAAT	
AMIDOSULFURON	H
AMITRAZ	I
AMITROLE	H,L
AMMONIUMSULFAAT NA	H,L
ASULAM	H
ATRAZINE NA	H,L
AZACONAZOLE	F
AZAMETHIPHOS	I,A
AZINPHOS-METHYL NA	I,A
AZOCYCLOTIN NA	I,A
AZOXYSTROBIN	F
BACT.PREP.BAC.THUR.	I
BENAZOLIN NA	H,L
BENFURACARB NA	I,A
BENOMYL	F
BENTAZONE	H
6-BENZYLAMINOPURINE NA	GR
BENZYLBENZOAAT	
BIFENOX	H,L
BIFENTHRIN	I,A
BITERTANOL	F
BORAX	H,F,I
BRODIFACOUM	R
BROMACIL NA	H,L
BROMADIOLONE	R
BROMOXYNIL	H
BROMUCONAZOLE NA	F
BROMOPROPYLATE NA	I,A
BUMINAFOS NA	H,L
BUPIRIMATE	F
BUPROFEZIN	I
BUTOCARBOXIM NA	I
BUTOXYCARBOXIM	I
CAPTAN	F
CARBARYL NA	I,A
CARBETAMIDE	H
CARBENDAZIM	F
CARBOFURAN	I
CLOFENTEZINE	A
CHLORBROMURON NA	H,L
CHLOROPHACINONE	R
CHLORFENVINPHOS	I
CHLORMEQUAT	GR
CHLORPROPHAM	H
CHLORPYRIFOS	I
CHLOROTHALONIL NA	F
CHLOROTOLURON NA	H,L
CHLORIDAZON	H
CINIDON-ETHYL-1999	H
CIPC NA	GR
1,3-DICHLOROPROPENE	H
CLODINAFOP	H
CLOMAZONE (2000)	H
CLOPYRALID	H
CLOQUINTOCET	H
CODLEMONE	FEROMOON
CREOSOOT/CRESOL	H,L
CYCLOXYDIM	H
CYDIA POM.GRANU.VIRUS - 1997	BIOL
CYFLUTHRIN	I
CYHEXATIN	A
CYMOXANIL	F
CYPROCONAZOLE	F
CYPRODINIL	F
CYROMAZINE	I
DAMINOZIDE	GR
DAZOMET	N
DELTAMETHRIN	I
DESMEDIPHAM	H
DESMETRYN NA	H,L
DIAZINON NA	I,A
DICAMBA	H
DICHLOBENIL	H
DICHLOFENTHION NA	I,A
DICHLORPROP-P NA	H,L
DICHLORVOS	I,A
DICHLORAN NA	F
DIDECYLDIMETHYLAMMONIUMCHLORID	ALG,BACT
DIENOCHLOR NA	A
DIETHOFENCARB	F
DIFENACOUM	R
DIFENOCONAZOLE	F

DIFETHIALONE	R
DIFLUBENZURON	I
DIFLUFENICAN	H
DIMETHENAMID	H
DIMETHOATE	I
DINOTERB NA	H,L
DIQUAT-DIBROMIDE	H
DITHIANON	F
DIURON	H,L
D-KARVON NA	GR
DNOC NA	H,L
DODEMORPH	F
DODINE	F
EPOXYCONAZOLE	F
EPTC NA	H,L
ESFENVALERATE	I
ETHEPHON	GR
ETHOFUMESATE	H
ETHOPROPHOS NA	G
ETRIDIAZOLE	F
ETRIMFOS NA	I,A
FENAMIPHOS	N
FENARIMOL	F
FENBUTATIN OXIDE	A
FENHEXAMID	F
FENITROTHION	I,A
PHENMEDIPHAM	H
FENOL NA	H,L
PHENOTHRIN	I
FENOXAPROP NA	H,L
FENOXYCARB	I
FENPICONIL NA	F
FENPROPATHRIN NA	I,A
FENPROPIMORPH	F
FENTIN ACETATE NA	F
FENTIN HYDROXIDE NA	F
FERRIC PHOSPHATE	M
FERROUS SULFATE	H
FIPRONIL	I,A
FLORASULAM	H
FLUAZIFOP-P	H
FLUAZINAM	F
FLUCYCLOXURON	A,I
FLUDIOXONIL	F
FLUROXYPYR	H
FLUTOLANIL	H
TAU-FLUVALINATE	I,A
FOLPET	F
FORMALDEHYDE	F
PHOSALONE NA	I,A
FOSETYL-ALUMINIUM	F
PHOSPHAMIDON NA	I,A
PHOXIM	I,A
FURALAXYL NA	F
GIBBERELINE	GR
GIBBERELLIC ACID	GR
GIBBERELLIN_A4_+_A7	GR
GLUFOSINATE-AMMONIUM	H
GLYPHOSATE	G
GLYPHOSATE-TRIMESIUM	H
GUAZATINE NA	F
HALOXYFOP-METHYL	H
HEPTENOPHOS NA	I,A
HEXYTHIAZOX	A
HYDRAMETHYLNON	I
HYMEXAZOL	F
IMAZALIL	F
IMAZAMETHABENZ-METHYL NA	H,L
IMIDACLOPRID	I
IOXYNYL	H,L
IPC NA	GR
IPRODIONE	F
ISOPROPANOL	H,L
ISOPROTURON	H
ISOXAFLUTOLE	H
KASUGAMYCIN	F,B
COPPER HYDROXIDE NA	F
COPPER OXYCHLORIDE	F
LAMBDA-CYHALOTHRIN	I
LENACIL NA	H,L
LINDANE NA	I,A
LINURON	H
MAGNESIUM PHOSPHIDE	I,R
MALATHION	I,A
MALEIC HYDRAZIDE	GR
MANCOZEB	F
MANEB	F
MCPA	H
MECOPROP NA	H,L
MECOPROP-P	H
MEPANIPYRIM	F
MESOTRIONE	H
METALAXYL	F
METALAXYL-M	F
METALDEHYDE	M
METAMITRON	H
METAM SODIUM	G
METAZACHLOR	H
METHAMIDOPHOS NA	I,A
METHIDATION NA	I,A
METHIOCARB	M
METHOMYL	I,A
METIRAM	F
METOBROMURON NA	H,L
METOLACHLOR NA	H,L
METOXURON	H
METRIBUZIN	H,L
METSULFURON-METHYL NA	H
MEVINPHOS NA	I,A
FORMIC ACID	F
MINERAL OIL	I
MONOLINURON NA	H,L

SODIUM_P-TOLUEENSULFONCHLORAMIDE	
SODIUMDICHLORISOCYANURATE	
NICOSULFURON	H
NITROTHAL-ISOPROPYL NA	F
OMETHOATE NA	I,A
OXAMYL	I,A
PACLOBUTRAZOL	GR
PETROLEUM OIL	A,I,H
PARAQUAT-DICHLORIDE	H
PARATHION (ETHYL) NA	I,A
PENCONAZOLE	F
PENCYCURON	F
PENDIMETHALIN	H
PER-ACETIC ACID	
PERMETHRIN	I,A
PIPERONYL BUTOXIDE	I
PIRIMICARB	I
PIRIMIPHOS-METHYL	I
PROCHLORAZ	F
PROCYMIDONE	F
PROMETRYN NA	H,L
PROPACHLOR NA	H,L
PROPAMOCARB-HYDROCHLORIDE	F
PROPAQUIZAFOP NA	H,L
PROPETAMPHOS NA	I,A
PROPICONAZOLE	F
PROPOXUR	I
PROPYZAMIDE	H
PROSULFOCARB	H
PYMETROZINE	I
PYRAZOPHOS NA	F
PYRETHRINS	I,A
PYRIDATE	H
PYRIDABEN	I,C
PYRIFENOX NA	F
YRIMETHANIL	F
QUATERN.AMMONIUMVERB	H,L
QUINMERAC NA	H,L
QUIZALOFOP-P-ETHYL	H
RIMSULFURON	H
SETHOXYDIM NA	H,L
SIMAZIN NA	H,L
S-METOLACHLOR	H
SPODOPTERA EXIGUA NPV	BIOL
TAR OILS NA	I,A
STREPTOMYCIN- SESQUISULFATE	BAC
STREPTOMYCES GRISEOVIRIDIS	BIOL
SULCOTRIONE	H
TEBUCONAZOLE	F
TEBUFENPYRAD	A
TEFLUBENZURON	I
TERBUTRYN NA	H,L
TERBUTHYLAZINE	H
TETRACHLORVINPHOS NA	I,A
TETRADIFON NA	A
TETRAMETHRIN	I
THIABENDAZOLE	F
THIODICARB	I,M
THIOFANATE-METHYL	F
THIOMETON NA	I,A
THIRAM	F
TOLCLOFOS-METHYL	F
TOLYLFLUANID	F
TRIADIMEFON NA	F
TRIADIMENOL	F
TRI-ALLATE	H
TRIAZAMATE	I
TRIAZOPHOS NA	I,A
TRICHLORFON	I,A
TRICLOPYR	H
TRIFLUMIZOLE	F
TRIFLUSULFURON-METHYL	H
TRIFORINE NA	F
TRINEXAPAC-ETHYL	GR
VALIDAMYCIN NA	F
VAMIDOTHION NA	I,A
VERTICILLIUM DAHLIAE KLEB.	BIOL
VERTICILLIUM LECANII	BIOL
FATTY ACIDS	H
VINCLOZOLIN	F
WARFARIN NA	R
WATERSTOFPEROXIDE	F
ZILVERTHIOSULFAAT	GR
ZINEB	F
ZIRAM	F, REP
SULFUR	F,A