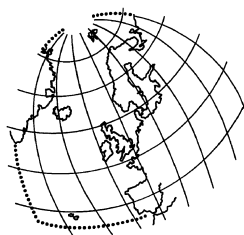


Lindane¹



OSPAR Commission

¹ Secretariat's note: A review statement on lindane (Publication 352c/2008) was adopted in 2008, highlighting new developments since the adoption of the Background Document.

The Convention for the Protection of the Marine Environment of the North-East Atlantic (the “OSPAR Convention”) was opened for signature at the Ministerial Meeting of the former Oslo and Paris Commissions in Paris on 22 September 1992. The Convention entered into force on 25 March 1998. It has been ratified by Belgium, Denmark, Finland, France, Germany, Iceland, Ireland, Luxembourg, Netherlands, Norway, Portugal, Sweden, Switzerland and the United Kingdom and approved by the European Community and Spain.

La Convention pour la protection du milieu marin de l'Atlantique du Nord-Est, dite Convention OSPAR, a été ouverte à la signature à la réunion ministérielle des anciennes Commissions d'Oslo et de Paris, à Paris le 22 septembre 1992. La Convention est entrée en vigueur le 25 mars 1998. La Convention a été ratifiée par l'Allemagne, la Belgique, le Danemark, la Finlande, la France, l'Irlande, l'Islande, le Luxembourg, la Norvège, les Pays-Bas, le Portugal, le Royaume-Uni de Grande Bretagne et d'Irlande du Nord, la Suède et la Suisse et approuvée par la Communauté européenne et l'Espagne.

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Executive Summary

Lindane is the common name for the γ -isomer of hexachlorocyclohexane (HCH) and is used as an insecticide. Lindane contains more than 99% γ -HCH. Priority was given to lindane in the 1992 OSPAR Action Plan, and it was therefore included in 1998 in the OSPAR List of Chemicals for Priority Action. Technical HCH also contains the other isomers but they do not possess a significant insecticidal activity. The use of technical HCH is generally prohibited in Western Europe and North America. Therefore, only lindane is considered in depth in this background document. Lindane is stable in fresh water as well as in seawater. It is removed through secondary mechanisms such as adsorption on sediment or via fish through the gills, the skin or ingestion. Degradation takes place much faster under anaerobic conditions than in the presence of oxygen. A limited degradability has been demonstrated and the occurrence in remote areas is due to long-range transport. Lindane occurs in different compartments and trophic levels of the Arctic and is accumulated by species at low trophic levels, while the biomagnification potential is low at the upper end of the food web. A number of ecotoxicity data for lindane are well within the range of OSPAR Ecotoxicological Assessment Criteria (0,5-5 $\mu\text{g/l}$), which are used for the identification of areas of concern.

Lindane is a contact insecticide with a widespread use in agriculture and forestry, for seed treatment and soil application, in household biocidal products, as a textile preservative and as a wood preservative. Lindane has been intensively used for many years since 1949 but has been replaced in most applications by pyrethroids and other insecticidal chemicals in recent years. From an estimated use of nearly 7900 tonnes in 1970 in Europe, the use decreased to about 2300 tonnes in 1996. France was a major user in the period 1992-1997, with an average consumption of 1600 tonnes per year, compared to an average of 2130 tonnes per year in Europe. In France, the consumption of lindane ceased in 1998.

The emissions of lindane to air in 15 OSPAR states were estimated at 733 tonnes in 1997. The atmospheric input into the North Sea was about 2,5-5 tonnes, and direct discharges and riverine inputs were estimated at 0,95-1,1 tonnes. Seawater in the Atlantic contains between 0,016 and 4,4 ng lindane per litre (on average 0,6 ng/l). Concentrations in the Central North Sea are 0,3-1,3 ng/l (on average 0,8 ng/l), while the concentrations are decreasing from the Southern North Sea (3,4 ng/l), via the German Bight (1,8 ng/l) to the north-western North Sea (0,3 ng/l).

Lindane has been regulated in several international forums. Lindane is one of the chemicals on the list of the UNEP Convention for Prior Informed Consent (PIC) and several countries have prohibited the import of lindane. Products containing less than 99% γ -HCH are banned under Council Directive 79/117/EEC. Eight of the nine countries bordering the North Sea have achieved the 50% reduction target for discharges/emissions of lindane in the period 1985-1999/2000. This included significant reductions in the United Kingdom, while use have ceased in Denmark, Germany, France, Norway and Sweden. The reduction target has not yet been achieved by Belgium and the Netherlands; however, in the Netherlands the authorisation for lindane has expired in 1999. Spain has no longer authorised lindane and Switzerland has severely restricted its use. The use of lindane will cease in the EU in June 2002 due to regulations under Council Directive 91/414/EEC concerning the placing on the market of plant protection products. Hexachlorocyclohexane has been identified as a priority hazardous substance under the Water Framework Directive 2000/60/EC.

The action recommended is: whilst noting the phase out of lindane in the EU, to focus attention on all the other HCH-isomers, to assist the European Commission in monitoring any problems with these isomers in the marine environment and to seek to achieve controls to reduce discharges and emissions; by the end of 2004 to review the likely achievements of these controls and consider the need for further OSPAR action with respect to any remaining uses; to undertake co-ordinated efforts within the framework of the UNECE-LRTAP and UNEP POP Conventions; to continue to monitor lindane in riverine inputs and direct discharges and in atmospheric inputs to the sea, in water, sediment and biota; and to ask other relevant international forums to take account of this background document.

A monitoring strategy for lindane has been added to this background document.

Récapitulatif

Le lindane est le nom commun de l'isomère γ de l'hexachlorocyclohexane (HCH), et est un insecticide. Il contient plus de 99% de γ -HCH. La priorité a été donnée au lindane dans le Plan d'action d'OSPAR en 1992, et il a en conséquence été inscrit en 1998 sur la Liste OSPAR des produits chimiques devant faire l'objet de mesures prioritaires. Le HCH technique contient aussi d'autres isomères sans activité insecticide significative. L'emploi du HCH technique est généralement interdit en Europe de l'ouest et en Amérique du nord. De ce fait, seul le lindane est abordé de manière approfondie dans le présent document de fond. Le lindane est stable dans l'eau douce comme dans l'eau de mer. Il est enlevé par des mécanismes secondaires, tels que l'adsorption sur les sédiments ou par le poisson qui l'absorbent par les ouïes ou la peau ou qui l'ingèrent. La dégradation est beaucoup plus rapide en milieu anaérobie qu'en présence d'oxygène. Une faible dégradabilité a été mise en évidence, et sa présence dans des zones éloignées est due à son transport sur de grandes distances. Le lindane est présent dans divers compartiments et à des niveaux trophiques dans l'Arctique, et s'accumule chez les espèces à de faibles niveaux trophiques, le potentiel de biomagnification étant faible à l'extrémité haute de la chaîne alimentaire. Un certain nombre de données d'écotoxicité du lindane se situent largement dans la fourchette des critères d'évaluation écotoxicologiques d'OSPAR (de 0,5 à 5 $\mu\text{g/l}$), lesquels servent à déterminer les zones préoccupantes.

Le lindane est un insecticide de contact largement utilisé dans l'agriculture et dans la sylviculture, pour traiter les semences et pour l'épandage sur le sol, dans les produits biocides ménagers, comme produit de protection des textiles et comme conservateur du bois. Le lindane est utilisé à grande échelle depuis de nombreuses années (1949), mais a été remplacé ces dernières années dans la plupart des applications par des pyréthroides et autres produits chimiques insecticides. Alors qu'en 1970, la consommation était estimée à près de 7900 tonnes en Europe, elle a été ramenée à 2300 tonnes à peu près en 1996. La France était un gros consommateur pendant la période de 1992 à 1997, sa consommation étant en effet en moyenne de 1600 tonnes par an, alors que la consommation européenne est en moyenne de 2130 tonnes par an. En France, l'utilisation du lindane a cessé en 1998.

Dans les 15 Etats d'OSPAR, en 1997, les émissions atmosphériques de lindane étaient estimées à 733 tonnes. Les apports atmosphériques à la mer du Nord étaient de l'ordre de 2,5 à 5 tonnes, tandis que les rejets directs et les apports fluviaux étaient estimés se situer entre 0,95 et 1,1 tonne. Les eaux de l'Atlantique présentent des teneurs en lindane de 0,016 à 4,4 ng par litre (en moyenne 0,6 ng/l). Les teneurs dans le centre de la mer du Nord vont de 0,3 à 1,3 ng/l (moyenne 0,8 ng/l), tandis que les teneurs sont en baisse du sud de la mer du Nord (3,4 ng/l) au nord-ouest de la mer du Nord (0,3 ng/l), en passant par le German Bight (1,8 ng/l).

Le lindane est réglementé par plusieurs instances internationales. C'est l'un des produits chimiques inscrits sur la liste de la Convention du PNUE sur le consentement préalable informé, et plusieurs pays en ont interdit l'importation. Les produits contenant moins de 99% de γ -HCH sont interdits par la Directive 79/117/CEE du Conseil. Huit des neuf pays riverains de la mer du Nord ont atteint l'objectif de 50% de réduction des rejets/émissions de lindane entre 1985 et 1999/2000. Il s'agit notamment des fortes réductions obtenues au Royaume-Uni, son utilisation ayant en outre cessé au Danemark, en Allemagne, en France, en Norvège et en Suède. La Belgique et les Pays-Bas n'ont pas atteint l'objectif de réduction ; cependant, aux Pays-Bas, l'autorisation du lindane est venue à expiration en 1999. L'Espagne a interdit l'emploi du lindane tandis que la Suisse en a rigoureusement limité l'utilisation. L'emploi du lindane cessera dans l'Union européenne en juin 2002 en conséquence des règlements issus de la Directive 91/414/CEE du Conseil, concernant la mise sur le marché des produits phytosanitaires. L'hexachlorocyclohexane a été classé dans les substances dangereuses prioritaires en vertu de la Directive cadre 2000/60/CE relative à l'eau.

Les mesures recommandées sont les suivantes : tout en prenant acte de l'abandon du lindane dans l'Union européenne, concentrer l'attention sur tous les autres isomères du HCH, afin d'aider la Commission européenne à détecter tous les problèmes que poseraient ces isomères dans le milieu marin, et s'efforcer de mettre des contrôles en place pour réduire les rejets et les émissions ; d'ici la fin 2004, examiner les

résultats de ces règlements et considérer s'il y a lieu d'adopter de nouvelles mesures OSPAR visant toutes les applications qui subsisteraient ; prendre des initiatives coordonnées dans le cadre de la Convention LRTAP de la Commission économique des Nations Unies pour l'Europe et de la Convention du PNUE sur les POP ; continuer à surveiller les apports fluviaux et les rejets directs de lindane ainsi que les apports atmosphériques à la mer, à l'eau, aux sédiments et au milieu vivant ; et demander aux autres instances internationales compétentes de tenir compte du présent document de fond.

Une stratégie de surveillance sur le lindane a été ajoutée à ce document de fond.

1. Introduction

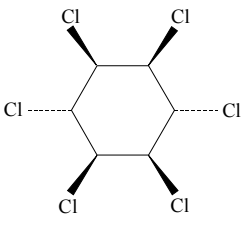
Lindane is a contact insecticide with a widespread use in agriculture and forestry, for seed treatment and soil application, in household biocidal products (e.g. treatment of companion animals, ornamentals and turfs), as textile preservative and as wood preservative. Lindane has been in intensive use for many years since 1949, but has been replaced in most applications by pyrethroids and other insecticidal chemicals in the last years.

Lindane is the common name for the γ -isomer of hexachlorocyclohexane (HCH). The purity of lindane is normally more than 99% γ -HCH. Technical HCH also contains the other isomers: α -HCH (55-80%), β -HCH (5-14%), γ -HCH (8-15%), as well as minor quantities of δ -HCH, ϵ -HCH, and other compounds (BREIVIK ET AL. 1999). All the isomers besides γ -HCH do not possess a significant insecticidal activity (KUTZ ET AL. 1991, quoted by BREIVIK ET AL. 1999) and are therefore not considered in depth in this background document. The use of technical HCH is generally forbidden in Western Europe and North America (WHO, 1991).

The importance of technical HCH as insecticide in general and lindane in specific is declining in Europe. While in a period from 1970 to 1979 HCH made up around 13% of the insecticides used in Europe, it was reduced to less than 5% in the period of 1991 to 1996 (BREIVIK ET AL. 1999). However, there is a large regional variance in the use of insecticides depending on the latitude (less in the north and more in southern parts of Europe) and the crops grown (cereals and soya requiring relatively less insecticides than, for example, maize, rape and further rice and cotton).

The chemical identity of lindane is presented in Table 1, further physico-chemical data are given in Appendix 1.

Table 1 Chemical identity of lindane

| | | |
|-----------------|---|---------------------------------|
| CAS-No. 58-89-9 |  | γ -hexachlorocyclohexane |
|-----------------|---|---------------------------------|

The occurrence of lindane is ubiquitous meanwhile and although its use in agriculture is diminishing rapidly it will still be distributed continuously in the environment within the next years. This is due to the characteristics of lindane as a “Persistent Organic Pollutant” (POP). Therefore, the existing monitoring of lindane should be continued.

The phasing out of lindane in agriculture in the EU started even before Decision 2000/801/EC stated that lindane was not to be included into Annex I of Council Directive 91/414/EEC concerning the placing of plant protection products on the market. The Monograph, on which this Decision is based, however, could not be evaluated within this background document yet, due to the confidentiality of this Monograph.

Results from reviews, compilations and existing assessments, some of which are based on internationally accepted rules, have not been re-evaluated for this background document and were included as such in the present report unless new information and findings differed considerably from the data already assessed.

2. Identification of sources of lindane and its pathways to the marine environment

2.1 Production of lindane and main uses as agricultural insecticide

The overall use of pesticides in Western Europe has been on more or less the same level over the past 15 years, except for a few countries with a known reduction programme such as the Netherlands, Denmark and Sweden (OSPAR, 2000c; with figures from EUROSTAT). However, this constant situation does not reflect the use of lindane which has decreased markedly since the middle of the 1990s and has lost its authorisation as a plant protection agent in the EU in 2000 (phase-out to be completed by June 2002).

There are practically no figures available on the use of lindane for the last two years (2000 and 2001) and most statistics end before 1998. Therefore, production volumes, uses by country, monitoring data etc. compiled in this background document do not represent the actual situation, but rather present a picture of the past, which is outdated due to a virtual cessation of agricultural uses of lindane and an insignificant non-agricultural use in the OSPAR Convention area.

From data gathered within the 'Popcycling-Baltic' project, BREIVIK ET AL. (1999) suggest that a total 382 000 tonnes of technical HCH and 81 000 tonnes of lindane were used in Europe from 1970 to 1996. This is equivalent to an estimated cumulative use of 259 000 tonnes α -HCH, 20 000 tonnes β -HCH, and 135 000 tonnes γ -HCH. The use of technical HCH was the major source of γ -HCH until the late 1970s, and thereafter lindane became the dominating source of this isomer. EU Member States put an end to the use of technical HCH in 1979 through Council Directive 79/117/EEC. In the same year, corresponding measures were taken by the Nordic Countries. By 1996, the use of γ -HCH had been reduced to about one-third of the European use in 1970. Some figures are given in Table 2.

Table 2 Production and trade of lindane before 1995

| | Tonnage (tonnes/year) | Source |
|---|------------------------------|--------------------|
| Import of lindane to Germany (1994) | 15,2 | IUCLID (1994) |
| Export of lindane from Germany (1994) | 14,4 | IUCLID (1995) |
| Production in Germany early 1980s | 4400 | UBA (2001) |
| Production in Germany since 1990 | - | UBA (1998a) |
| Production world-wide 1986 | ca. 38 000 | IUCLID (1994) |
| World Production 1988-1993 | 4400 | UBA (1998a) |
| World Production 1990-1995 | 3222 | UBA (1998a) |
| Production in Western Europe in the 90s | ca. 2055 | UBA (1998a) |
| Production in the EEC (1991) | 1000 – 5000 | RIPPEN (1990/2000) |

BREIVIK ET AL. 1999 present a figure on the estimated use of HCHs in Europe 1970-1996 and specified for α -, γ -HCH and other isomers. The estimation is based on total insecticide consumption. Detailed data on the different HCH isomers were not available from all European countries; therefore the vertical lines in Figure 1 give the 95- and 5-percentile values. For details on the estimation procedure see BREIVIK ET AL. (1999).

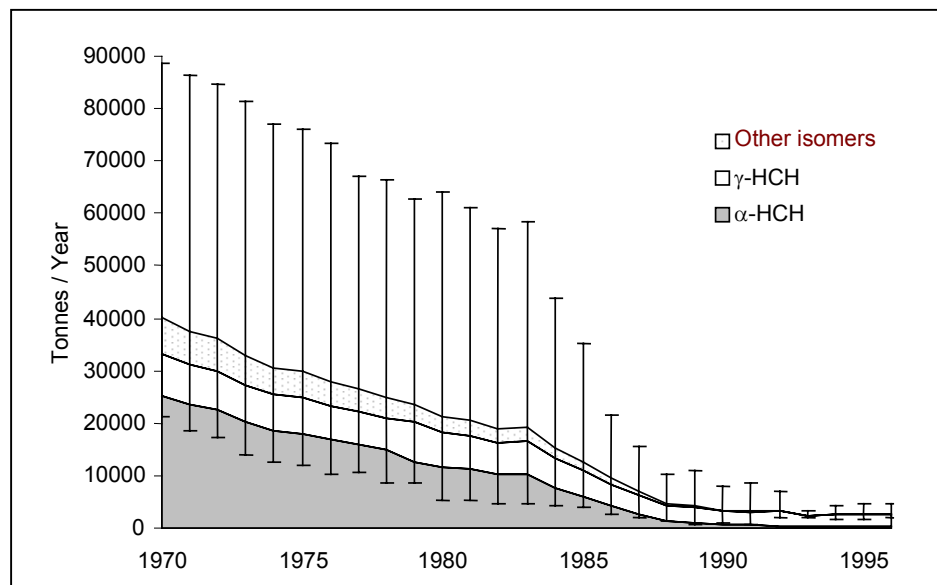


Figure 1: Estimated use of α -, γ -HCH and other isomers in Europe, 1970-1996 (BREIVIK ET AL. 1999; data from the Popcycling-Baltic Project)

Figure 1 shows a substantial decrease in the European use of α -HCH during the study period. In 1970, the use of α -HCH was estimated to be 25 000 tonnes on a European basis, while the use in 1996 was only 366 tonnes, corresponding to 1,4% of the use in 1970. The same relative decrease is observed for β -HCH, as technical HCH is the only source considered for both of these isomers. However uncertain, almost 80% of the remaining uses of α - and β -HCH in Europe in 1996 were assigned to the new states of the former Soviet Union (422 tonnes of technical HCH). The other 20% were attributed to uses in some former Eastern European countries.

For γ -HCH (lindane) the results indicate a decrease in use, but not as steep as for the other two isomers. From an estimated use of nearly 7900 tonnes in 1970, the use decreased to about 2300 tonnes in 1996. The lowest use of γ -HCH in that period was estimated to be 2000 tonnes in 1993. According to Centre International d'Études du Lindane (CIEL 1998; quoted by BREIVIK ET AL. 1999) the average annual lindane consumption in Europe was 2130 tonnes during the period from 1992 to 1997. France was a major user of lindane in Europe in this period, with an annual average consumption of 1600 tonnes/year. In France, the consumption of lindane terminated in July 1998.

2.2 Non-agricultural uses of lindane

In 1999 the OSPAR Commission published a report on non-agricultural uses of certain pesticides by OSPAR Contracting Parties (OSPAR, 1999). This report summarises the information received through a questionnaire up to the end of 1997. However, the report reflects the time period during the late 1980s/early 1990s and points to the fact, that an actual complementary inventory on non-agricultural uses of pesticides will probably emerge with the implementation of the EU Biocide Directive 98/8/EC.

In this report, HCH (lindane, γ -HCH) is referred to as a “category A pesticide” (“agricultural pesticides used in large quantities and in most countries”) which is not true anymore today.

Lindane was used in all responding states except for the Nordic countries (Denmark, Finland, and Sweden) as well as the Netherlands. Uses were for wood preservation, insect control in public and private areas, and medicinal uses for the control of ectoparasites on humans and animals. The use in Switzerland was only for human and veterinary medicines. In the UK, lindane was also approved for the control of

certain insects on amenity turf and grass and for use as a rodenticide; the amount of lindane used by local authorities as a rodenticide being very low, about 2,5 kg/year.

The only responding states who were able to give figures for tonnage used were:

- Belgium (0,375 tonnes/year for the use as an insecticide in private/public areas and as wood preservative – no figures being available for medicinal use), and
- Ireland (4,3 tonnes/year for the use as preservative, in pest control and as miscellaneous biocide – no information being given on the types of use).

In terms of the total non-agricultural uses of lindane:

- the use in wood preservation was “important” (> 25% of the total non-agricultural use) for Belgium and the UK, “of minor importance” (< 4%) for Germany;
- the use as an insecticide in public/private areas was “important” (> 25%) for the UK, “of medium importance” (5%-24%) for Ireland and “of minor importance” (< 4%) for Germany;
- the medicinal use was “of minor importance” (< 4%) for Belgium (another 3 countries also reporting this kind of use);
- in the UK, the extent of the use was “of medium importance” (5%-25%) on amenity turf/grass and “of minor importance” (< 4%) as rodenticide.

Certain risks arise from these non-agricultural uses. They are discussed in Chapter 3.4.

3. Monitoring data, quantification of sources and assessment of the extent of problems

3.1 Quantification of sources

3.1.1 Emissions to the atmosphere and atmospheric inputs to the European mainland and the North Sea

The AMAP Assessment Report (1998) gives the following indication of long-range atmospheric transport of HCHs to the Arctic Ocean:

HCH compounds volatilise soon after application, especially in the tropics (TAKEOKA ET AL. 1991; quoted in AMAP 1998), and are atmospherically transported to the Arctic and other remote regions. The low Henry's Law Constants of HCHs favour partitioning from air, especially at low temperatures. At equilibrium, the ratio of α -HCH concentration in seawater to that in air is 3000 at 25°C, but 22 000 at 0°C. The world's oceans are the major reservoirs of HCHs. Global models estimate that approximately 20% of the HCH present in the environment is held in the ocean surface layer, which is taken to be the upper 75m (STRAND & HOV 1996; quoted in AMAP 1998) or 200 m (MACKAY ET AL. 1995; quoted in AMAP 1998). Even though the heaviest use of HCHs has been in tropical and subtropical regions, levels in surface seawater are an order of magnitude higher in the Arctic (IWATA ET AL. 1993; SCHREITMÜLLER & BALLSCHMITTER 1995; quoted in AMAP 1998). Concentrations of HCHs in tree bark increase with latitude, being relatively high in Canada, Nordic Countries, Alaska, and Russia, and lower in tropical countries (SIMONICH & HITES 1995; quoted in AMAP 1998). Thus the transport and distribution of HCHs exemplifies the ‘cold condensation’ effect (WANIA & MACKAY 1993, 1995; quoted in AMAP 1998).

The emission of lindane to the atmosphere was estimated for Europe for 1990 and 1997 (Table 3; UBA, 1997). This does not necessarily give the atmospheric load to the North Sea and the North East Atlantic, because some of the load is transported outside the OSPAR Convention area and some is deposited over land; in fact, the atmospheric load to the North Sea is much lower than the emissions (Table 4).

Within EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe) an investigation and assessment of POP transboundary transport and

accumulation in different media (EMEP 2000) was conducted. Using the MSCE-POP model (Meteorological Synthesizing Centre – East, Moscow) the simulation of lindane (γ -HCH) transport within the European region for the period of 1970-1997 was made. Emission estimates obtained under the POPCYCLING-Baltic project for the indicated time period were used in modelling. On the basis of calculations, mean deposition and concentration values in environmental compartments (air, soil, sea water, vegetation) were determined in European countries for 1997. Preliminary estimates of lindane emitted in each country and deposited in other European countries were estimated and a primary analysis of the relation between emission and concentration levels in individual countries was made. To verify the model, measured and calculated data for 1990-1997 are compared.

Table 3 Emission of lindane to the atmosphere in Europe

| Atmospheric emission | Emission volume (tonnes/year) | Source |
|--|-------------------------------|-------------|
| Emission from Europe (38 countries) (1990) | 1310 | UBA (1997) |
| Emission from Europe (38 countries) (1997) | 765 | EMEP (2000) |
| Emission from 15 OSPAR countries (1990) | 417 | UBA (1997) |
| Emission from 15 OSPAR countries (1997) | 733 | EMEP (2000) |

The comparison of calculated and measured data indicates that the model results for all the compartments are underestimated (see also HOLOUBEK ET AL. 2000). To some extent, this is explained by high volatility of this compound (according to the calculations about 60% of its emission is transported outside the EMEP grid). For this reason, in order to obtain a correct assessment of the pollution level in the European region, lindane modelling should be made on the hemispherical scale (compare LI ET AL. 1998).

Figure 2 indicates that major emission sources (according to emission data used for 1997) are located in south-western Europe (France, Belgium and Spain). However, widespread agricultural uses of lindane have come to an end in the EU in the meantime.

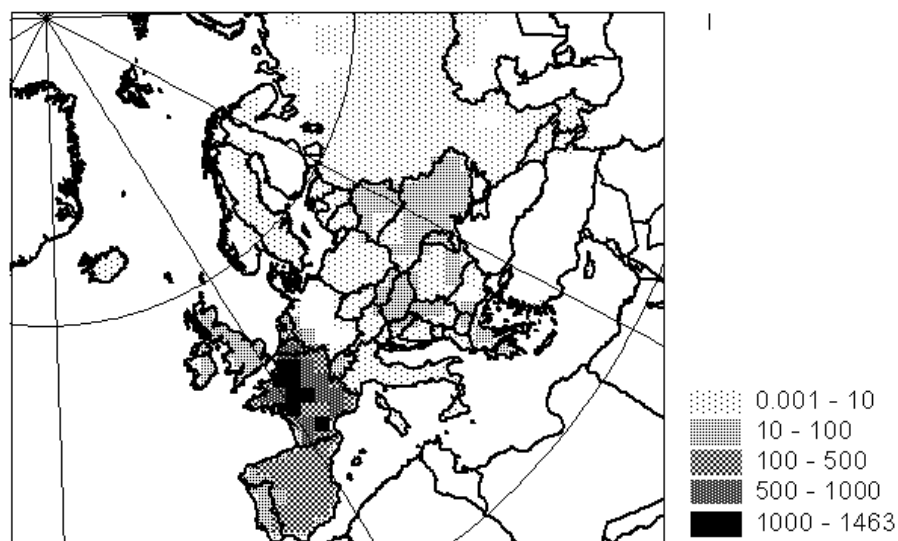


Figure 2 Lindane emission to air in 1997 (MSCE-model) in g/km²/a (from EMEP 2000)

Further to the emissions, the air concentration and atmospheric deposition over Europe in 1997 is calculated by MSC-East. Further calculated data on concentrations in air, sea water, soil and vegetation are given in Chapter 3.2.1.

Other figures in Table 4 show the estimated atmospheric input into the North Sea from the beginning of the 1990s.

Table 4 Atmospheric input of lindane into the North Sea

| Approaches to estimating atmospheric inputs | Atmospheric Input (kg/year) | Source |
|--|-----------------------------|------------------|
| Wet deposition (mean from 5 values 1990-1994; calculated for 525 000 km ²) | 5000 | OSPAR (1996a) |
| dto. 1990-1995 | 4500 | OSPAR (1997) |
| Extrapolated to the North Sea area from deposition data from Westerland/Sylt | 2600 | UBA (1997) |
| Dry and wet deposition (1990s) | 1580 | SELKE (1994) |
| Extrapolated from 0,88 g/km ² for 1991; wet deposition only | 500 | HÜHNERFUß (1995) |

3.1.2 Riverine inputs to the North Sea

Next to relatively high volatility of lindane, which results in a considerable transport via the air, lindane is also more water soluble (see Annex 1) than most of the other chlorinated hydrocarbons, and a major input to the maritime environment is via rivers from the application areas.

Since 1990, OSPAR is collecting data on lindane, among others, according to the “Principles of the Comprehensive Study of Riverine Inputs” (see e.g. OSPAR, 2000h). The figures in Table 5 show data on the discharge from Germany into the North Sea from 1990 to 1997 (UBA, 1998b). Since the authorisation of lindane came to an end in Germany only in the middle of the 1990s, a diminishing effect is not visible in the load figures up to 1997. Lindane is mainly transported in the water phase and only to a lesser extent adsorbed to particulate suspended matter (see Table 5 and Table 6). Additional riverine inputs to the North Sea were registered from the UK (Table 6) and other countries (Table 7). In comparison to the riverine inputs, those from direct municipal or industrial discharges are negligible (Table 8 and Table 9).

Table 5 Riverine input of suspended matter and lindane from Germany to the North Sea from 1990 to 1997 (UBA, 1998b)

| Year | Outflow (Million m ³ /year) | Suspended matter (tonnes/year) | lindane load (kg/year) |
|------|---|-----------------------------------|---------------------------|
| 1990 | 26 631 | 2 537 546 | 340 |
| 1991 | 24 303 | 3 290 630 | 180 |
| 1992 | 33 053 | 3 483 009 | 200 |
| 1993 | 35 212 | 4 911 721 | 200 |
| 1994 | 56 066 | 1 460 194 | 250 |
| 1995 | 53 502 | 1 360 800 | 290 |
| 1996 | 33 769 | 1 170 734 | 180 |
| 1997 | 34 861 | 1 419 422 | 350 |

Table 6 Riverine Input of suspended matter and lindane from UK to the North Sea from 1985 to 1996 (assembled from GOVERNMENT STATISTICAL SERVICE, 1998)

| Year | Suspended matter (tonnes/year) | lindane load (kg/year) |
|------|--------------------------------|------------------------|
| 1985 | no data | 1 560 |
| 1990 | 2 900 000 | 780 |
| 1994 | 3 260 000 | 636 |
| 1995 | 2 480 000 | 608 |
| 1996 | 1 790 000 | 370 |

Table 7 Riverine inputs to the North Sea

| Year | Source | lindane load (kg/year) |
|-------------|---|---------------------------|
| 1990s | COMMPS (median from EU-wide monitoring data; extrapolated to 176 km ³ /year *) | 2130 |
| 1990s | IKSR (median from Rhine monitoring data at Lobith; extrapolated with 70 km ³ /a (Rhine outflow) to 176 km ³ /year *) | 792 |
| 1990-1995 | Riverine Inputs and Direct Discharges (RID) OSPAR (1998b) | 970 |
| before 1995 | Discharges of rivers to the North Sea (CLEEMANN ET AL. 1995 with references therein) | 1100 |
| 1996 | Elbe (median from Elbe monitoring data at Schnackenburg; extrapolated with 24 km ³ /a (Elbe outflow) to 176 km ³ /year *); IKSE | 2980 |

*) 176 km³ per year is the long-term average total river discharge into the North Sea (OSPAR, 1998a).

Table 8 Direct discharges (kg) of lindane to the North Sea by country (Data Reports on the Comprehensive Study of RID, quoted in OSPAR, 2000c; OSPAR, 2000g,h)

| Country | 1990 | 1991 | 1992 | 1993 | 1994 | 1995 | 1996 | 1997 | 1998 |
|-------------------|--------------------|--------------------|--------------------|--------------------|-------------------|-------------------|------------------|--------------------|-------------------|
| Denmark | 0-30,3 | 0-30,3 | 0-30,3 | 0-30,3 | 0-30,3 | 0-30,3 | n.i. | n.i. | n.i. |
| Germany | 4,8 | 4,8 | 0,0-0,3 | 0,0-0,3 | 0,0-0,3 | 0,4 | 0,04-0,05 | 0,02-0,3 | 0,02-0,3 |
| Netherlands | 5,7 | 5,0 | 3,9 | 3,9 | 3,9 | 3,9 | n.i. | 0,0 | 0,0 |
| UK * | 150-152 | 128-154 | 103-104 | 110-111 | 85-86 | 90-92 | 61-66 | 118-134 | 60-108 |
| Total (kg) | 160,5-192,8 | 137,8-194,1 | 106,9-138,5 | 113,9-145,5 | 88,9-121,5 | 93,9-126,2 | 61,0-66,1 | 118,0-134,3 | 60,0-108,3 |

Table 9 Riverine inputs (kg) of lindane to the North Sea by country (Data Reports on the Comprehensive Study of RID, quoted in OSPAR, 2000c; OSPAR, 2000g,h)

| Country | 1990 | 1991 | 1992 | 1993 | 1994 | 1995 | 1996 | 1997 | 1998 |
|-------------------|------------------|-----------------|----------------|------------------|-----------------|------------------|----------------|----------------|----------------|
| Belgium | 102 | 100-102 | 77-88 | 62-98 | 52-97 | 54-57 | n.i. | 56-76 | 101-120 |
| Denmark | 0-30,3 | 0-30,3 | 0-30,3 | 0-30,3 | 0-30,3 | 0-30,3 | n.i. | n.i. | n.i. |
| France | 175 | 175 | 175 | 175 | 175 | 175 | 120 | n.i. | n.i. |
| Germany | 342 | 183 | 207-209 | 193-194 | 250-272 | 291 | 234-243 | 370 | 261 |
| Netherlands | 11 | 2,4 | 6,6 | 360 | 150-310 | 352-385 | 300 | 300 | 258-261 |
| Norway | 420 | 216 | 70 | 61 | 61 | 70 | 48 | 46 | 57 |
| UK * | 147-234 | 224-351 | 142-239 | 245-298 | 196-265 | 154-251 | 96-140 | 70-108 | 61-130 |
| Total (kg) | 1197-1314 | 900-1060 | 678-818 | 1096-1216 | 884-1210 | 1096-1259 | 798-731 | 842-900 | 738-829 |

Notes to Table 8 and Table 9:

n.i.: No information.

* There is a further significant discharge from the UK to the Irish Sea and the Atlantic.

3.2 Monitoring data

3.2.1 Global and European monitoring data

Atmosphere

From various sources (MOLTMANN ET AL. 1999a) a global atmospheric background range for lindane may be compiled to about 0,015–0,3 ng/m³.

Some atmospheric monitoring data on lindane for Western Europe (Sweden and Germany) were collected from handbooks (HOWARD 1991; RIPPEN 1990/2000 – data from the time before 1990) as well as for Eastern Europe (HEINISCH ET AL. 1994 – data from the 1980s). Quite high concentrations of lindane were measured in Gdansk in the early 1990s (FALANDYSZ 1998). One report from eastern Canada (Resolute Bay, 1992) gives an extremely high value of 9,8 ng/m³ (BIDLEMAN ET AL. 1995) for lindane, while another from Sable Island (1988/89) lies around 0,02 ng/m³ (BIDLEMAN ET AL. 1992). This value is of the same order of magnitude as measurements in the tropospheric border layer over the Atlantic with a range 0,0015-0,331 ng/m³ (SCHREITMÜLLER & BALLSCHMITTER 1995).

Measurements from the Arctic are reported by SELKE (1994) and in a review report (HARGRAVE & VASS 1987). LI ET AL. (1998) reported a relationship between global uses of technical HCH and air concentrations of α -HCH in Arctic air between 1979 and 1994. Elevated levels of persistent organic pollutants (including HCHs) are positively correlated with long-range transport episodes from use areas in the mid-latitudes of North America, Europe and Asia. During transport episodes with air masses originating mainly in Europe, elevated levels of PCBs and HCHs were observed at an air monitoring station on Svalbard (Spitzbergen) (OEHME ET AL. 1996, quoted in OSPAR 2000b).

For the Arctic, NORSTROM & MUIR (1994) have shown that HCHs occur frequently in samples from air, snow, and sea water (at 10 and 225 m) as well as in zooplankton, while samples from higher trophic levels (amphipods, arctic cod, beluga, ringed seal, polar bear) contain HCHs only in 10 % or less of the samples.

The MSC-East model (see Chapter 3.1.1) assumes that lindane in air is present in the gas phase only. Regions of highest emissions in 1997 (see Figure 2 above) also show the highest concentrations in air (1-5,5 ng/m³) (Figure 3 from EMEP, 2000).

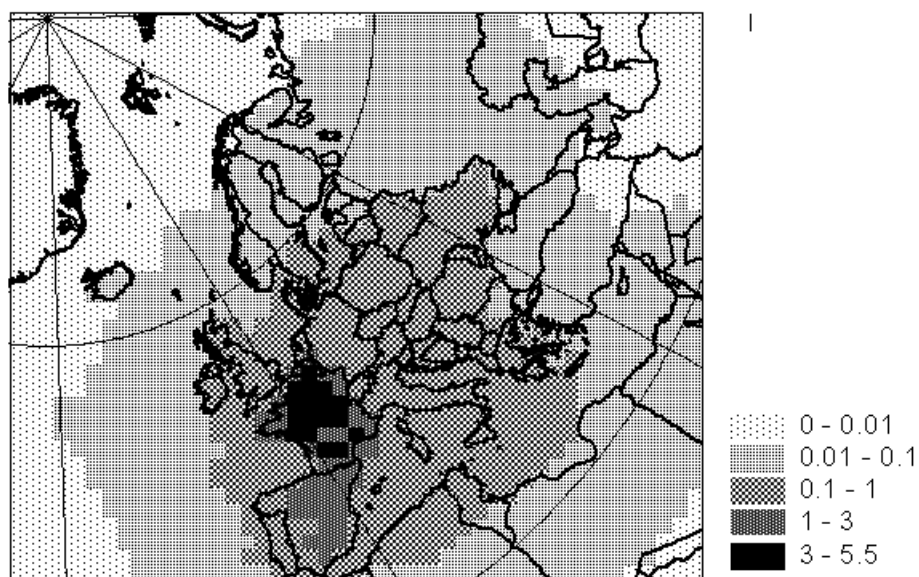


Figure 3: Lindane air concentration in 1997 in ng/m³ (from EMEP 2000).

For comparison, EMEP (2000) provides mean annual air concentrations of some EMEP observation stations in Finland, Norway, Sweden, Iceland and the Czech Republic (Table 10). Unlike the overall view, calculations overestimate the air concentration for these remote areas (ratio of measurement to calculation is < 1). The only exemption is the Norwegian station on Svalbard (Spitzbergen) near the EMEP grid boundary which receives considerable input from sources located outside the EMEP region. Again, this supports the need for hemispherical modelling.

Table 10 Mean annual air concentrations for lindane from EMEP stations in 1996/97 in ng/m³ (EMEP 2000, modified)

| Station | Country | Year | Measurement [ng/m ³] | Calculation [ng/m ³] | Meas./Calc. |
|-------------|-----------|------------------|----------------------------------|----------------------------------|-------------|
| FI96 | Finland | 1996 | 0,011 | 0,022 | 0,49 |
| Svalbard | Norway | 1997 | 0,015 | 0,002 | 8,75 |
| Lista | Norway | 1997 | 0,062 | 0,085 | 0,73 |
| SE2 | Sweden | 1996 | 0,025 | 0,172 | 0,14 |
| Stórhöfði | Iceland | 1997 | 0,006 | 0,016 | 0,42 |
| Kosetice | Czech. R. | 1997 | 0,034 | 0,326 | 0,11 |
| Mean | | 1992-1997 | 0,039 | 0,074 | |

Finland reported high deposition of HCHs in Lapland in 1993 and 1994 in early summer (0,5 to 1 µg/m²/month). The large magnitude of precipitation in southern Finland in June 1993 and 1996 was one reason for the very high level of HCHs per square metre (2,4 to 2,7 µg/m²/month). The main isomer found in both Lapland and southern Finland was γ-HCH. Also α-HCH was found but β-HCH much less than both other isomers (KORHONEN ET AL. 1998).

The annual deposition of HCHs per square metre in Finland is about the same level as in Sweden. In Denmark the HCH deposition is estimated to be twice as much as in Finland; in Norway three times and in Germany five times the deposition of Finland (*ibid.*; see also references for the different countries).

Seawater

A global marine water phase background value for lindane was estimated at about 0,6 ng/l, ranging from 0,016 ng/l in lower reaches of the Atlantic (1985; KRÄMER & BALLSCHMITTER 1988) to a maximum of 4,4 ng/l off the coast of Iceland in the Arctic (1985; HARGRAVE & VASS 1987). These values are derived from global baseline studies as mentioned before (and e.g. WITTLINGER & BALLSCHMITTER 1990). Some newer values (1990-1992) of 0,021-0,075 ng/l were presented by SCHREITMÜLLER & BALLSCHMITTER (1995).

The occurrence of lindane in sea water as calculated by the MSC-East model, due to atmospheric input (and subsequent dispersion with currents), is shown in Figure 4. The analysis of the calculation results points out that the marine environment exerts the largest influence on lindane dispersion over a long time scale. Lindane, which is accumulated in the sea, can support air concentration levels during 10-15 years. Re-emission of lindane takes place in some regions of the Mediterranean Sea. It is conditioned by the temperature regime and the relationship of air and sea concentrations in this region; air concentrations are relatively low whereas sea concentrations show maximum concentrations (EMEP, 2000).

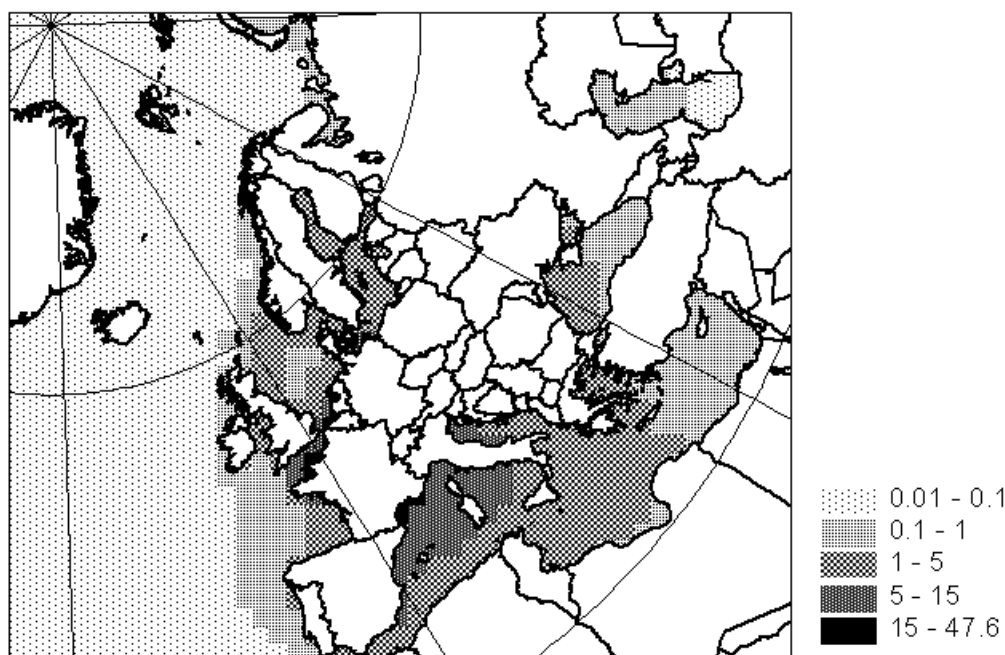


Figure 4 Lindane seawater concentrations in 1997 in µg/m³ (from EMEP 2000)

Marine sediment

Some local data on concentrations of HCHs and lindane in sediments from the North Sea are given in chapter 3.2.2.

Rivers

Within the EU Water Framework Directive (2000/60/EC) the European Commission has set up a legal framework and a clear methodological basis for the prioritisation of substances potentially hazardous to the aquatic environment. A combined monitoring-based and modelling-based priority setting procedure (COMMPS) was applied in the selection process of the proposed priority substances. In this context

monitoring data from many European rivers were collected, weighted and aggregated (EC 1999). For lindane and other HCH-isomers the following overall concentrations were calculated (Table 11).

Table 11 Results of the aggregation of monitoring data for HCHs and lindane in the aquatic phase and sediment from the COMMPS procedure (EC 1999; from Tables A9 and A10)

| CAS No. | Compound | 90-perctle. | Median [µg/l] | Arimeth. Mean | Stand. Deviat. | Sample Stat. | Entries used | Entries > DL |
|--------------------|-----------------|----------------|----------------|----------------|----------------|--------------|--------------|--------------|
| Water phase | | (µg/l) | (µg/l) | (µg/l) | (µg/l) | (-) | (-) | (-) |
| 319-84-6 | α-HCH | 0,0248 | 0,0036 | 0,0094 | 0,0082 | 77 | 1974 | 1190 |
| 319-84-7 | β-HCH | 0,0378 | 0,0064 | 0,0129 | 0,0080 | 44 | 1226 | 751 |
| 319-86-8 | δ-HCH | 0,0218 | 0,0033 | 0,0092 | 0,0163 | 18 | 208 | 106 |
| 58-59-9 | γ-HCH (lindane) | 0,0370 | 0,0083 | 0,0168 | 0,0279 | 546 | 11666 | 8260 |
| Sediment | | (µg/kg) | (µg/kg) | (µg/kg) | (µg/kg) | (-) | (-) | (-) |
| 319-84-6 | α-HCH | 76,96 | 2,67 | 19,42 | 31,23 | 27 | 594 | 398 |
| 319-84-7 | β-HCH | 62,8 | 3,08 | 42,26 | 143,64 | 27 | 822 | 528 |
| 58-59-9 | γ-HCH (lindane) | 10,96 | 3,19 | 9015 | 15,57 | 53 | 953 | 689 |

Medians for lindane are far lower than the German quality standard for lindane and other HCHs in surface water with respect to aquatic biocoenoses (0,3 µg/l; see Chapter 5.1, and even the 90-percentile value is lower than the drinking water threshold for plant protection products (0,1 µg/l).

Soil and vegetation

Lindane concentrations in soil and in vegetation resulting from atmospheric deposition show a similar distribution as in Figure 3 above (calculated with the MSC-East model). Maximum concentrations in France (1997) reached 15-25 µg/kg in soil and 100-170 µg/kg dw in vegetation (spruce needles) (EMEP, 2000).

Measurements of the soil concentration of 1,01 µg/kg at a station in the Czech Republic (Kosetice) 1997 in comparison with the calculated 3,58 µg/kg show that the model overestimated the lindane content in soil (ibid.). On the other hand, the lindane concentration in vegetation measured in 1997 is 4,84 µg/kg and somewhat higher than the calculated value of 2,92 µg/kg (ibid.).

Groundwater

A survey of pesticides in ground water for 1990-1995 was carried out in Germany (UBA 2001), i.e. for the time when lindane was phased out for agricultural use in Germany. Lindane was ranked number 20 of the most common active substances found in groundwater. In the following years (1996-1998) lindane was still found regularly at 5-7% of the observation points, and in few cases with more than 0,1 µg/l.

3.2.2 Monitoring data for the North Sea

The atmospheric value for the North Sea area is derived from CAMP-data from a single station at the Norwegian coast for 1993 and 1994 (OSPAR 1995, 1996a). The range for all monthly values of both years is 0,02–0,46 ng/m³, and the average is 0,09 ng/m³. This value is well within the range of the above stated atmospheric background level.

For the central North Sea, 27 single measurements in seawater for 1990-1994 ranged from 0,3–1,3 ng/l (MUDAB, 1998; UBA, 2001). This gives an arithmetic mean of 0,8 ng/l, which is more than 10 times higher than the above stated background value for water. In water, lindane concentrations are higher in the southern North Sea (up to 3,4 ng/l) and the German Bight (up to 1,8 ng/l) than in north-western North Sea (around 0,3 ng/l; data from THEOBALD ET AL. (1996), quoted in OSPAR, 2000c). In the Dutch part of the Wadden Sea concentrations up to 8 ng/l were found in the period 1992-2000 (see www.waterstat.nl).

OSPAR has agreed on an Ecotoxicological Assessment Criterion (EAC) for lindane in water in the range of 0,0005-0,005 µg/l (0,5-5 ng/l) (OSPAR, 1996 and OSPAR, 1997, quoted in OSPAR, 2000c; see also OSPAR 1997, Summary Record, Annex 6). The lindane concentrations in water reported above are within the range of this EAC. For sediment and mussels, EACs were not considered relevant for the current monitoring programme, although values in mussels from the East Frisian Wadden Sea and at a site in the Scheldt estuary exceeded EAC levels (OSPAR, 2000c).

The concentration values for the North Sea sediments are collected from various sources (1988-1992; MOLTSMANN ET AL. 1999a) and range from 5-3000 ng/kg (mean value around 500 ng/kg). UBA (2001) states a value of up to 700 ng/kg for the Wadden Sea. OSPAR (1996b) gives a range of <100-5000 ng/kg (dry weight) for sediments from Belgium and the Scheldt estuary in 1992.

3.2.3 Lindane in biota

Concentrations of lindane in fish liver and mussel tissue generally decreased during the period 1990 to 1995, especially in relatively polluted regions of estuaries, fjords and near coastal zones (OSPAR, 2000a). In contrast, a significant upward trend was observed in dab muscle from Lista (southern Norway) for the same period. Concentrations of lindane in mussels around the Irish Sea, Bristol Channel, Celtic Sea and Atlantic coasts are in the µg/kg wet weight (ww) range and, in livers of flatfish from the Irish Sea, are approximately ten times higher. In a study, the highest levels of lindane in fish occurred in Liverpool Bay. Concentrations of α - and γ -HCH in the blubber of stranded male harbour porpoises from the Irish Sea and west of Scotland during the past ten years have been approximately 2-4 mg/kg lipid (ibid.).

There is evidence that lindane concentrations (α - and γ -HCH) in biota have generally decreased. A number of significant downward trends were observed in southern and south-western Norwegian fjords, in the Kattegat, and in the southern North Sea (along the west coast of Belgium, the Oyster Ground, the German Bight) (ibid.).

SIMMONDS ET AL. (1994; quoted in OSPAR, 2000f) report concentrations of 0,30 and 0,38 mg/kg ww (range: 'not detectable' to 0,92 mg/kg ww) in blubber samples from adult pilot whales killed at two sites in the Faroe Islands in 1986.

OSPAR (2000b) presents a compilation of data on the range of concentrations for selected organochlorine compounds (including HCHs) in biota in the Arctic Ocean from the first half of the 1990s (ibid. Table 4.2, p. 52). All values are in the range of µg/kg ww (Table 12).

Table 12 Range of concentrations for HCHs in biota in the Arctic Ocean from the first half of the 90s (OSPAR 2000b)

| Animal Group | Tissue | Concentration Range (µg/kg ww) |
|----------------|----------------|--------------------------------|
| Mussels | soft tissue | < 0,5 – 0,82 |
| Fish | liver | < 0,6 – 153 |
| Sea birds | liver | 0,2 – 25 |
| Marine Mammals | blubber fat | 17 – 473 nd – 1150 |

nd: Not detected

BREIVIK ET AL. (1999) have undertaken the work to compare data on the use of HCH (all isomers) with measured concentrations in biological material. Several studies (quoted in BREIVIK ET AL. 1999) have investigated the spatial distribution of HCH in plant biomass in Europe (ERIKSSON ET AL. 1989; JENSEN ET AL. 1992; CALAMARI ET AL. 1994; SINKKONEN ET AL. 1995) or at a global scale (CALAMARI ET AL. 1991; SMONICH & HITES 1995). Unfortunately, both differences in analytical methodology, species sampled and limited years covered reduce the feasibility for a comparison with the use. Furthermore, as stated by WANIA (1997) and quoted by VALLACK ET AL. (1998), the multi-hop nature of the environmental transport of POPs complicates the estimation of source-receptor links.

Recently, BINGERT ET AL. (1998; quoted in BREIVIK ET AL. 1999) reported data covering time-series of organochlorines in biota from the Baltic area. The longest time trend for HCH was presented for α -HCH in muscle of pike (*Esox lucius*) from lake Storvindeln, close to the Arctic Circle in Sweden. As there is little agricultural activity in this remote area, atmospheric transport and deposition are expected to be the only significant source of α -HCH to the lake. BREIVIK ET AL. (1999) showed that the decreasing trends in use of HCH in countries adjacent to the Baltic are comparable to the decreasing concentrations measured in pike of the last number of years. This is also true for herring.

3.2.4 Lindane in animal food items

In the period from 1995 to 1998, lindane was found in 10-50% of meat samples and in more than 50% of fish, crab and mollusc samples in Germany (UBA 2001). Other HCH isomers were found less often.

3.3 Assessment of the extent of the problem

3.3.1 Physico-chemical properties

Representative physico-chemical data of lindane are presented in Appendix 1.

3.3.2 Biotic and abiotic degradation

Aerobic degradation

ULMANN (1972) indicates that lindane is very stable in fresh water as well as in salt water environments. It is removed from the water column through secondary mechanisms, such as adsorption to the sediment or to fish through the gills, the skin or ingestion. To a certain extent, biodegradation also takes place (EXTOXNET, Cornell University).

The IUCLID database shows the degradation in a screening test as follows (Council Directive 79/831/EEC; C.6; ThOD; no other sources):

- 99% degradation after 28 days;
- 5 days (84,2%);
- 14 days (> 100%);
- 28 days (99%).

However, the validity of the results of the screening test is questioned by the German authorities.

Lindane is moderately persistent in water, with a half-life ranging between 20 and 200 days (according to the Support Document of the AQUIRE Database). The higher value, however, clearly characterises lindane as a persistent substance.

Static screening flask tests with 5 and 10 mg/l do not show any transformation of lindane even after adaptation (TABAK ET AL. 1981; GHISALBA 1983; RICHARDS & SHIEH 1986; quoted by RIPPEN 1990/2000). In a Closed-Bottle-Test (according to OECD 301 D) the biodegradability measured by oxygen demand reached 18% in 28 days (GSF 1982; quoted by RIPPEN 1990/2000), which classifies lindane as “not ready biodegradable”. Mineralisation in activated sludge is low: 0,1% CO₂ development only (RICHARDS & SHIEH 1986).

Aerobic degradation takes place in sewage treatment plants to a certain extent: in one case elimination was 25% (less than 1% by stripping; PETRASEK ET AL. 1985; quoted by RIPPEN 1990/2000), in others 0-98% (mean 41%; n=42; VAN LUIN & VAN STARKENBURG 1985; quoted by RIPPEN 1990/2000). However, the elimination with activated sludge is practically 0% (10 mg/l lindane; RICHARDS & SHIEH 1986), also with older sludge, but about 23% with fresh activated sludge (KIRK & LESTER 1988; quoted by RIPPEN 1990/2000).

In a pilot plant for sewage water treatment (SCHRÖDER 1987; quoted by RIPPEN 1990/2000), the bulk of lindane was found in the outflow (81% and 91% in the first and second stage, respectively), while the rest remained in the sludge (19% and 9% respectively). A degradation was only observable with a low loading (thus resulting in 6% degradation). In another pilot plant, degradation was assessed at 28% excluding elimination by sorption and stripping (BHATTACHARYA ET AL. 1990; quoted by RIPPEN 1990/2000).

In one system consisting of river water and sediment, lindane was degraded or eliminated at a rate of 80% within a period of three weeks, in another only at a rate of 15% in 87 days (CALLAHAN ET AL. 1979; quoted by RIPPEN 1990/2000). When using only river water without sediment, transformation in 84 days achieves 0%, 0% and 20% (ibid.).

Some half-lives are given for an eutrophic pond (92 h / 3,8 d at 25 °C, pH=9,3), a dystrophic reservoir (770 h / 32 d at 25 °C, pH=7,3), and an oligotrophic water (650 h / 27 d at 25 °C, pH=7,8). The persistence in river water, seawater and groundwater is given to be 3-30 days, 30-300 d, and more than 300 days, respectively (US-EPA, 1988).

KALSCH ET AL. (1998) report half-lives of about 90 to 697 days for the temperature range of 5-15 °C (this is considered to be worst case). KALSCH ET AL. report a mineralisation of 47% to ¹⁴CO₂ only after 91 days in one of their experiments. Between 1 and 9% of the radioactivity volatilised during the tests. At the end (day 91), 23 to 66% were bound to sediments and 15 to 45% of the total activity was neither extractable by water nor by cyclohexane (ibid.).

MACKAY ET AL. (2000; quoted by BEYER & MATTHIES 2001) report the following half-lives for lindane: water – 708 days, sediment – 2 292 days.

The metabolisation pathway for lindane is described in REINECKE (2001).

Anaerobic degradation

Under anaerobic conditions, degradation is found to take place much faster than under aerobic conditions (FIEDLER ET AL. 1990).

The comprehensive US-EPA report (CALLAHAN ET AL. 1979) states several examples for anaerobic degradation (water/sediment, sludge, soil) with transformation of lindane up to more than 95 % in several weeks. FISCHER & PECHER (1993; quoted by RIPPEN 1990/2000) found half-life values of 0,3 to 16,5 for different leachates originating from anaerobic municipal waste decomposition.

Degradation in soil

Degradation half-lives in soil are given by JURY ET AL. (1983, 1984; 260 and 266 days) and DIETER (1990; 1 year). OTTOW (1985) reports a transformation of 75-100% in 3-30 years (all quoted by RIPPEN 1990/2000). BRUBAKER & HITES (1998; quoted in BEYER & MATTHIES 2001) come to half-life in soil of 708 days.

Other references point to the fact that no results from standardised soil degradation tests are available (US-EPA, 1988).

Also half-lives from field experiments vary considerably. The maximum values are over 300 days. The calculation of a PEC_{soil} from a dt_{50}^1 of 180 days does not seem to represent the realistic worst case which would have to be set at 292 days from the data presented in the EU Draft Monograph for lindane.

3.3.3 Overall persistence and atmospheric long-range transport

The fact that lindane only has a limited degradability has been demonstrated in Chapter 3.3.2 and the occurrence in remote areas due to long-range transport has been discussed in Chapter 3.2.1.

To further discuss the property of overall persistence, which is considered to be a key parameter in chemicals evaluation, as well the long-range transport potential, reference is made to the work of BEYER & MATTHIES (2001).

The overall persistence is the average residence time of a molecule in the environment, but accounting only for irreversible loss processes.

According to BEYER & MATTHIES (2001), the overall persistence is calculated using a level III multimedia model and assuming an emission scenario based on actual production, use, and disposal of the chemical. The scenario describes the persistence of the chemical in the source region. Alternatively, the overall persistence is calculated using the same model as above but assuming 100% emission into air. This scenario describes the persistence in a remote region. Here, a remote region is defined as a region having no direct emissions. The longer persistence of the two scenarios is selected in order to get a conservative estimate of the real persistence of the substance.

BEYER & MATTHIES (2001) calculate the overall persistence τ_{ov} for different emission scenarios (as explained above):

- | | |
|---|------------------------|
| • 100% emission into air (standard scenario) | $\tau_{ov} = 613$ days |
| • 20% emission to air, 80% emission to soil | $\tau_{ov} = 873$ days |
| • 10% emission to air, 10% emission to water, 80% emission to soil | $\tau_{ov} = 859$ days |
| • 100% emission to water | $\tau_{ov} = 796$ days |

¹ dt_{50} : disappearance time of 50% of the substance.

When comparing these values with half-lives given by other authors in Chapter 3.3.2 (half-life in air: 43,3 days), the overall persistence for lindane is generally far higher than for α -HCH.

The characteristic travel distance (CTD) describes the long-range transport potential as the distance at which the initial concentration drops to 37 % (1/e). For the standard scenario (100% emission to air), the CTD is calculated to be 7401 km.

3.3.4 Bioaccumulation and biomagnification

Information on bioaccumulation (in terms of bioconcentration, BCF) for lindane can be derived from the log P_{OW} using the formula:

$$\log BCF = 0,77 \log P_{OW} - 0,70 \quad (\text{MEYLAN ET AL. 1999});$$

$$\text{thus } \log BCF = 0,77 \cdot 3,66 - 0,70 = \underline{2,12}; \quad BCF = \underline{131,3} .$$

The log P_{OW} value of > 3 itself indicates a “potential for bioaccumulation”.

Measured BCF values were obtained from the AQUIRE database (<http://www.epa.gov/ecotox/>; Table 13). The LC_{50} (as indicator for toxicity) and the water solubility (~ 7 g/l) are well above the test concentrations (2,0-5,6 $\mu\text{g/l}$), so as not to have a negative influence on the test performance.

Table 13: Selected data on the bioaccumulation of lindane (γ -HCH) – CAS No. 58-89-9

| Test organism | Trophic level / Habitat | LC_{50} / Test concentration | Test conditions | BCF | Source / Year |
|----------------------------|-------------------------|--|-----------------------|--------|---------------|
| <i>Brachydanio rerio</i> | sCons FW | $> 100 \mu\text{g/l}$ $5,6 \mu\text{g/l}$ | 0,1-5 d; FLO; FORM | 13 400 | [1] / 1991 |
| <i>Lepomis macrochirus</i> | sCons FW | $> 40 \mu\text{g/l}$ $2,0 \mu\text{g/l}$ | 0,1-5 d; FLO; FORM | 15 400 | [1] / 1991 |
| <i>Oncorhynchus mykiss</i> | sCons FW | $> 20 \mu\text{g/l}$ $2,0 \mu\text{g/l}$ | 0,1-5 d; FLO; FORM | 14 600 | [1] / 1991 |
| <i>Poecilia reticulata</i> | sCons FW | $> 16 \mu\text{g/l}$ $2,0 \mu\text{g/l}$ | 0,1-5 d; FLO; FORM | 12 800 | [1] / 1991 |

[1] AQUIRE / 6919; LA ROCCA ET AL. 1991.

sCons: secondary consumers; FW: freshwater species; FLO: flow-through test; FORM: Formulation instead of active substance was used in the test.

The data selected here for the bioaccumulation of lindane (Table 13) are all taken from the same study; other BCF values in the AQUIRE database are usually 10 times lower.

The BCF-values in Table 13 indicate that also a “not so lipophilic” substance like lindane with a log P_{OW} of 3,66 may be a candidate for biomagnification, although other laboratory results do not indicate a biomagnification potential, proven by complete depuration (BEEK ET AL. 2000; BUTTE ET AL. 1991: BCF for γ -HCH: 920 ± 131).

BEEK ET AL. (2000) discussed the fact that not only the degree of lipophilicity but also the degree of chlorination and the position of the chlorine atoms, and particularly the elimination pathways determine the potential of biomagnification:

During the 1987/88 a mass mortality of bottlenose dolphins occurred along the U.S. Atlantic coast. γ -HCH was among the nine of the most frequently detected pesticides (KUEHL ET AL. 1991).

However, comparing biomagnification efficiencies and residues of top predators with the same diet, e.g. fish eating tuna fish and dolphins, there are data suggesting that not the predator status *per se*, but the lack of branchial elimination pathways of mammals as compared to the elimination potential of gill-breathing fish may explain the higher residues and bioaccumulation/biomagnification potential in marine mammals (MARSILI ET AL. 1995).

This is in conformity with investigations on the bioaccumulation and transfer of γ -HCH, PCBs and DDTs in pike (*Esox lucius*) (LARSSON ET AL. 1993). Lipids and concentrations of contaminants in hard foe were 10 times higher as compared to muscle suggesting that the transfer via roe is an important elimination pathway for the individual and a prerequisite for persisting residues in the offspring.

NORSTROM & MUIR (1994) have demonstrated in different compartments and trophic levels of the Arctic that HCH occurs in air, snow and sea water and is efficiently accumulated by species at low trophic levels, while the biomagnification potential is low at the upper end of the food web (arctic cod, beluga, ringed seal, polar bear).

γ -HCH residues have also been found in water, sediments, eggs of pelicans and eels, the main pelican prey. Data suggest a biomagnification with a factor of 1,8 between eel and pelican eggs. The log BCFs/BAFs for eel and pelican eggs were 3,33 and 3,58, respectively, related to water, i.e. nearly as high as the log P_{OW} for γ -HCH, making evident the risk of underestimating the bioaccumulation from laboratory investigations (ALBANIS ET AL. 1995; quoted in BEEK ET AL. 2000).

Further examples for the terrestrial compartment are presented by BEEK ET AL. (2000).

3.3.5 Acute and chronic ecotoxicity effects

Lindane is less toxic to *Daphnia* than to other organisms (Table 14). *Eurypanopeus depressus* shows a very low LC₅₀ value. When compiling data on ecotoxicity of lindane, accepted data from an OSPAR-Workshop (OSPAR, 1996) are included, even if originating from the time before 1980.

Table 14: Data on the ecotoxicity of lindane (γ -HCH) - CAS No. 58-89-9

| Test organism | Trophic lev. / Habitat | Endpoint / Effect | Test conditions | Endpoint concentration | Source Year |
|--------------------------------|------------------------|-------------------------|-------------------|------------------------|-------------|
| <i>Scenedesmus subspicatus</i> | pProd FW | NOEC GRO | 3 d; STA; | 1 400 μ g/l | [1] 1994 |
| <i>Daphnia magna</i> | pCons FW | LC ₅₀ MOR | 2 d; STA; FORM | 1 600 μ g/l | [2] 1986 |
| <i>Gammarus pulex</i> | pCons FW | NOEC GRO | 10 d; REN | 2,67 μ g/l | [3] 1996 |
| <i>Eurypanopeus depressus</i> | sCons SW | LC ₅₀ MOR | 4 d; REN; FORM | 0,66 μ g/l | [4] 1987 |
| <i>Salmo trutta</i> | sCons FW | LC ₅₀ MOR | 4 d; STA; ACT | 1,7 μ g/l | [5] 1980 |

[1] UBA-Database 1994.

[2] In IUCLID Data Set (Rev. 2/1994); Data from 1986.

[3] UBA-Database 1996.

[4] AQUIRE /4358; SHIRLEY & MCKENNEY 1987.

[5] AQUIRE /666; JOHNSON & FINLEY 1980.

Complete references may be obtained from the AQUIRE-Database.

pProd: primary producers; pCons: primary consumer; sCons: secondary consumer; FW: freshwater species; SW: saltwater species; GRO: Growth; MOR: Mortality, Immobilisation; STA: static test; REN: semi-static (renewal) test; ACT: active substance; FORM: Formulation instead of active substance was used in the test.

OSPAR has agreed on an Ecotoxicological Assessment Criterion (EAC) for lindane in the range of 0,0005-0,005 µg/l (0,5-5 ng/l) (OSPAR, 1996; OSPAR, 1997, quoted in OSPAR, 2000c; see also OSPAR 1997, Summary Record Annex 6). The above stated ecotoxicity data are well above this range.

EACs were established for assessing chemical monitoring data from the OSPAR Convention area (see Chapter 3.2.2). EACs are the concentrations of specific substances in the environment below which no harm to the environment or biota is expected. Criteria for lindane were derived using all available ecotoxicological data that passed predefined quality criteria (OSPAR, 1996). Although based on laboratory toxicity tests, usually employing freshwater organisms, these reference values were established for use as the best available assessment criteria. Levels below these values suggest that no harm to the marine environment should be expected. However, caution should be exercised in using (...) assessment criteria in specific situations since their use does not preclude the use of common sense and expert judgement with regard to natural concentrations. Furthermore, the EACs do not take into account specific, long-term biological effects such as carcinogenicity, genotoxicity and endocrine disruption, or the combined effects of substances (OSPAR, 2000c).

3.3.6 **Carcinogenicity, mutagenicity or harmful reproductive effects**

The following toxicological information is taken from the Pesticide Information Profile of the EXTOTOXNET database (<http://ace.ace.orst.edu/info/extotoxnet/pips/lindae.htm>; revision date is June 1996). Reference is frequently made to IPCS Environmental Health Criteria (WHO, 1991).

Carcinogenic effects: No tumours were found in groups of 20 mice fed with beta-, gamma-, and delta-isomers of HCH at about 64 mg/kg/day, but tumours occurred in 100% of the mice fed with alpha-isomer only. In rats similar findings were noted at doses of about 49 mg/kg/day. Other work suggests that rodents may suffer from liver tumours from high doses of the gamma-isomer (lindane). HCH was not found to promote tumours initiated by benz[a]anthracene. The available evidence is contradictory, and does not allow assessment of the potential for carcinogenic effects in humans from lindane exposure.

The International Agency for Research on Cancer (IARC) has concluded that lindane is a “possible human carcinogen” (Class 2B).

Mutagenic effects: Most tests on mice and on micro-organisms have shown no mutagenicity due to lindane exposure. However, lindane has been shown to induce some changes in the chromosomes of cultured human lymphocytes at 5 and 10 ppm in the culture medium. Some chromosomal damage was also noted at a concentration of 1 ppm in this study. An *in vivo* (in live animals) study of the effects of lindane on rat leukocytes (white blood cells) did not find chromosomal abnormalities after a single administration of 75 mg/kg. It is unlikely that lindane would pose a mutagenic risk in humans at normal exposure levels.

Reproductive effects: In rats, doses of 10 mg/kg/day for 138 days resulted in marked reductions in fecundity and litter size, and half that dose (5 mg/kg/day) reportedly had no effect. In another study in rats, doses as low as 0,5 mg/kg/day over 4 months caused observable disturbances in the rat oestrus cycle, lengthened gestation time, decreased fecundity, and increased foetal mortality. Lindane was found to be slightly estrogenic to female rats and mice, and also caused the testes of male rats to become atrophied. Semeniferous tubules and Leydig cells (important for production of sperm) were completely degenerated at doses of 8 mg/kg/day over a 10-day period. Reversible decreases in sperm cell production were noted in male mice fed approximately 60 mg/kg/day for 8 months. It is unlikely that lindane will cause effects at the low levels of exposure expected in human populations.

Teratogenic effects: In rats, doses as low as 0,5 mg/kg/day over 4 months caused decreased growth in offspring. Beagles given 7,5 or 15 mg/kg/day from day 5 throughout gestation did not produce pups with any noticeable birth defects. Pregnant rats given small amounts of lindane in their food had offspring unaffected by the pesticide. It appears that lindane is unlikely to cause developmental effects at levels of exposure expected in human populations.

3.3.7 Toxicological effects

The following toxicological information is taken from Pesticide Information Profile of the EXTTOXNET database (ace.ace.orst.edu/info/exttoxnet/pips/lindae.htm; revision date is June 1996).

Acute toxicity: Lindane is a moderately toxic compound via oral exposure, with a reported oral LD50² of 88 to 190 mg/kg in rats. Other reported oral LD50 values are 59 to 562 mg/kg in mice, 100 to 127 mg/kg in guinea pigs, and 200 mg/kg in rabbits.

Gamma-HCH is generally considered to be the most acutely toxic of the isomers following single administration. It is moderately toxic via the dermal route as well, with reported dermal LD50 values of 500 to 1000 mg/kg in rats, 300 mg/kg in mice, 400 mg/kg in guinea pigs, and 300 mg/kg in rabbits. Notably, a 1% solution of lindane in vanishing cream resulted in a six-fold increase in acute toxicity via the dermal route in rabbits, with a reported dermal LD50 of 50 mg/kg. It is reported to be a skin and eye irritant. Younger animals may be more susceptible to lindane's toxic effects. Calves are especially susceptible to dermal application.

Effects of high acute exposure to lindane may include central nervous system stimulation (usually developing within 1 hour), mental/motor impairment, excitation, clonic (intermittent) and tonic (continuous) convulsions, increased respiratory rate and/or failure, pulmonary oedema, and dermatitis.

Other symptoms in humans are more behavioural in nature such as loss of balance, grinding of the teeth, and hyper-irritability. Most acute effects in humans have been due to accidental or intentional ingestion, although inhalation toxicity occurred (especially among children) when it was used in vaporisers. Workers may be exposed to the product through skin absorption and through inhalation if handled incorrectly. Lotions (10%) applied for scabies have resulted in severe intoxication in some children and infants. It is reported that single administrations of 120 mg/kg inhibited the ability of white blood cells to attack and kill foreign bacteria in the blood of rats, and 60 mg/kg inhibited antibody formation to human serum albumin. It is not clear whether these effects were temporary, or for how long they may have lasted.

Chronic toxicity: Doses of 1,25 mg/kg/day in mice, rats, and dogs produced no observable effects over periods of up to 2 years. Doses of 40 to 80 mg/kg/day were rapidly fatal to dogs in a study over 2 years, and doses of 2,6 to 5,0 mg/kg/day resulted in convulsions in some test animals. This same dose level caused liver lesions in rats. In one study, 6 to 10 mg/kg/day was reported to have no observable effects on mice, but in another study, that dose caused apparent metabolic changes in the liver. Other studies in mice have demonstrated liver damage at higher doses. In a 2-year rat study, significant liver changes were attributed to the dietary intake of approximately 5 mg/kg/day.

Sufficiently high repeated administration of lindane has caused kidney, pancreas, testes, and nasal mucous membrane damage in test animals. There have been reported links of lindane to immune system effects; however, these have not been amply demonstrated in test animals or in humans in a long-term study. Long-term toxicity of the gamma-isomer may be less than that of the alpha- and beta-isomers due to its more rapid transformation and elimination and lesser storage in the body. Sixty male workers in a lindane producing factory had no signs of neurological impairment or perturbation after 1 to 30 years exposure. Another study of chronically exposed workers showed subtle differences between their electrocardiographs (graphs of the heart beat impulses) and those of unexposed workers.

Organ toxicity: Data from animal tests indicate that lindane may affect the central nervous system, liver, kidney, pancreas, testes, and nasal mucous membrane.

² LD50: lethal dosis.

3.4 Risk to the environment

The relatively high volatility of lindane, together with the obvious long-range transport into the Arctic characterise lindane as a persistent organic pollutant (POP) in the framework of the UN ECE POP-Protocol. OSPAR (2000f; p. 91) gives the following evaluation of persistent organic contaminants in general:

Traces of persistent organic contaminants are ubiquitous in all compartments of the deep ocean ecosystems. However, as many of these compounds are uniquely anthropogenic their source must be human activities. There is no evidence for persistent organic contaminants having a deleterious effect on species within Region V, but it remains an open question as to whether this reflects a lack of studies or a true absence of impact. For deep ocean biota, dilution effects (in terms of the volume of water and the diffuseness of the biota) probably keep the severity of any impact well below those experienced by shallow-water biota. There remains the persistent concern that the risks associated with the constant stream of new synthetic substances being introduced into world markets are currently underplayed by the chemical industries. Despite widespread awareness of the serious and deleterious consequences that have arisen from the introduction of some of these substances (e.g. DDT, CFCs and dioxins), the public perception is that genetic engineering and ecosystem manipulation associated with mariculture are far more threatening. Hence there is relatively little pressure on the chemical industry to develop more effective testing procedures for new synthetic products. It is suspected that high concentrations of some persistent organic contaminants could be leading to pathological responses, for example through the depression of immune response. Specific actions to reduce the inputs of persistent organic contaminants to Region V (Wider Atlantic) are not required since successful measures to reduce inputs to, and impacts on, shallow water ecosystems will be equally effective at reducing impacts on deep ocean ecosystems.

As far as the non-agricultural uses are concerned (see Chapter 2.2), for the use as wood preservative (in Belgium, Germany and the UK), surface waters, sediment and biota were considered to be the risk areas. Germany felt that contamination of water from a point source was “important”, whereas Belgium and the UK classified this route of contamination to be of “minor importance”. Germany also felt that contamination via point sources from direct discharge was “likely” but, lindane was hardly used any more for wood preservation in Germany (OSPAR, 1999).

As an insecticide in private/public areas, it was felt that surface waters, biota and possibly sediment could be at risk. However, user countries considered contamination of water “unlikely”. No areas of risk were identified for the use of lindane as an insecticide on amenity turf/grass nor the limited use as a rodenticide in the UK (*ibid.*).

The medicinal use of lindane (for control of ectoparasites on humans and animals) was considered to be either of “minor importance” or “unlikely” to pose a risk to the aquatic environment (*ibid.*).

These appraisals reflect the situation of the early 90s, as pointed out in the OSPAR Report. Meanwhile, the situation has changed to a diminished use of lindane in the non-agricultural sector too (compare Chapter 5.1).

4. Desired reduction

Hexachlorocyclohexane isomers (HCH) are on the OSPAR List of Chemicals for Priority Action since 1998 (see the OSPAR Strategy with regard to Hazardous Substances). The OSPAR objective with regard to hazardous substances on this list is to prevent pollution of the maritime area by continuing to reduce discharges, emissions and losses of hazardous substances, with the ultimate aim of achieving concentrations in the marine environment near background values for naturally occurring substances and close to zero for man-made synthetic substances. Every endeavour will be made to move towards the target of cessation of discharges, emissions and losses of hazardous substances by the year 2020.

Taking into account this objective, it can be concluded that in the light of the information and data compiled on lindane (γ -HCH) in the previous chapters in terms of:

- the identification and quantification of sources and pathways to the marine environment;
- the available environmental monitoring data;
- the assessment of the extent of the problem, in particular the intrinsic properties;

there is sufficient clear and scientifically proven evidence to justify that from the marine environmental point of view all open uses of lindane are to be banned.

The need to avoid all uses of lindane has been widely acknowledged at national and international level (see Table 15 below). Several OSPAR Contracting Parties have already banned HCH completely. In addition, its further use will be banned or severely restricted by international agreements and legislation, such as the European Commission Decision 2000/801/EC by which all uses of lindane as an agricultural pesticide in the EU will have to cease in June 2002 at the latest.

However, there are some other remaining open uses of lindane which affect the OSPAR maritime area and which should be banned as well. Within OSPAR Contracting Parties, there might be minor uses for which appropriate alternatives are difficult to find. While every effort should be made to substitute lindane also in minor uses, it will be more important, in order to achieve the OSPAR objective and target, to achieve a reduction of the quantities of lindane which are being transported (e.g. via atmospheric deposition) into the maritime area from sources outside the Convention area.

5. Identification of possible measures

5.1 Review of agreed national and international measures for the regulation of lindane

5.1.1 OSPAR Strategy with regard to Hazardous Substances

HCH isomers have been on the OSPAR List of Chemicals for Priority Action right from 1998 (see Annex 2 to the OSPAR Strategy with regard to Hazardous Substances; reference number: 1998-16). Annex 3 of the OSPAR Strategy with regard to Hazardous Substances lists substances which are candidates for selection, assessment and prioritisation according to paragraph 3.1.c or 3.1.e of the OSPAR Strategy. Lindane has been on the list of substances identified as of concern by OSPAR in the period 1991-1997, which was part of the OSPAR Action Plan. At the same time lindane was on the OSPAR List of Potential Endocrine Disruptors – Part A, which have been reported in the scientific literature to induce changes to the endocrine system of varying severity in the course of in vivo tests.

5.1.2 Ban of HCH and lindane and PIC-Convention

Lindane is one of the chemicals on the list for Prior Informed Consent (PIC) and several countries have prohibited its import.

Products containing less than 99% γ -HCH are banned under Council Directive 79/117/EEC. Discharges of HCH in waste waters leaving an industrial or treatment plant are regulated under Council Directive 84/491/EEC.

Table 15 gives an overview on measures against the use of HCH and lindane in some European countries and in the EC.

Table 15: Measures against the use of HCH and lindane in European countries before the EU withdrawal of approval in 2000 (Table 15 and references modified from BREIVIK ET AL. 1999)

| Country | Action | Year | Restrictions / remaining uses | Reference |
|----------------|---|---------------------------|--|------------------------------------|
| Austria | γ-HCH banned | 1992 | NA | UNEP 1995 |
| Belgium | γ-HCH severely restricted | 1989 | Veterinary use and wood protection | UNEP 1995 |
| Bulgaria | γ-HCH banned | 1993 | | UNEP 1995 |
| Cyprus | γ-HCH severely restricted | 1987 | Paints and wood protection; agricultural use banned | UNEP 1995 |
| Czechoslovakia | γ-HCH severely restricted | NA | NA | UNEP 1995 |
| Denmark | γ-HCH banned | 1995 | | Miljø and Energi Ministeriet 1997* |
| EC | γ-HCH severely restricted banned for agricultural use | 1978/ 1988 2000 | Council Directive 79/117 on HCH containing less than 99% of the γ-isomer | UNEP 1995 2000/801/EC |
| Finland | HCH cancelled, γ-HCH banned | 1977 1988 | Mixed isomers cancelled 1977 | AMAP 1998 |
| France | γ-HCH banned | 1998 | | BREIVIK (1999) |
| Germany | γ-HCH banned | 1988 | | UNEP 1995 |
| Greece | γ-HCH still approved | 2001 | | EC 2001 ** |
| Hungary | γ-HCH severely restricted | 1968 | Registered as grain treatment for winter wheat and nurseries | UNEP 1995 |
| Iceland | HCH never registered | | γ-HCH authorisation for use as pesticide withdrawn in 1993, for veterinary use withdrawn 1999, the use stopped a few years earlier | AMAP 1998 |
| Ireland | γ-HCH still approved | 2001 | | EC 2001 ** |
| Italy | γ-HCH severely restricted | NA | NA | UNEP 1995 |
| Netherlands | γ-HCH severely restricted, subsequently banned | 1991 1999 | Use for agricultural applications prohibited | UNEP 1995 |
| Norway | γ-HCH banned | 1991 | | Landbr. 1998* |
| Poland | Phase-out | NA | | UNEP 1995 |
| Portugal | HCH banned γ-HCH still approved | 1974 2001 | Ban on products using mixed isomers | UNEP 1995 EC 2001 ** |
| Sweden | γ-HCH severely restricted and subsequently banned from use in agriculture | 1970 1988 | Remaining use as wood preservative suspended 1988 | AMAP 1998 UNEP Legal File |
| Spain | HCH banned γ-HCH no longer authorised | 1994 2000 | No exemptions | Spanish OSPAR Delegation |
| Switzerland | γ-HCH severely restricted | 1986 | Prohibition of sale of mixed isomers. Sale of γ-HCH for some applications | UNEP 1995 |
| UK | HCH banned; γ-HCH authorisation withdrawn | 1979 2001 | Sale and supply of γ-HCH banned 2002, use up period ends 2003 | UK OSPAR Delegation |
| USSR | γ-HCH severely restricted for use as pesticide | 1988 | Techn. HCH and γ-HCH probably still in use. | AMAP 1998 |

NA = no information available

*) Personal communication (see BREIVIK ET AL. 1999).

***) Status in January 2001 of current authorisations of lindane (European Commission / Internet).

5.1.3 North Sea Conference

The Progress Report to the Fifth North Sea Ministerial Conference indicates that for HCH (incl. lindane) the reduction target of 50% for discharges/releases to water and emissions to air from 1985 to 1999/2000 has been achieved in 8 out of 9 countries discharging into the North Sea. This includes significant reductions in Great Britain, while use was discontinued in Germany, Denmark, France, Norway, and Sweden. The reduction target of 50% has not been achieved in Belgium (41,5 tonnes/year) and the Netherlands (39,4 tonnes/year). The Netherlands reported that authorisation for lindane expired in October 1999. The use of lindane will end in the EU in June 2002 at the latest (see below).

5.1.4 UN ECE POPs Protocol / UNEP POPs Convention

A Protocol on Persistent Organic Pollutants (POPs) under the framework of the United Nations Economic Commission for Europe (UN ECE) 1979 Convention on Long Range Transboundary Air Pollution (LRTAP) was signed in Aarhus, Denmark, on 28 June 1998. The Protocol lists HCH under its Annex II "Substances Scheduled for Restrictions on Use". However, the Stockholm Convention on Persistent Organic Pollutants (POPs) of 23 May 2001, which was concluded in the framework of UNEP, does not contain any regulations for HCH, although both the goals of the POPs Protocol and the POPs Convention are very similar.

The UN ECE POPs Protocol restricts the use of technical HCH to the use as an intermediate in chemical manufacturing. It restricts products containing lindane to the following uses, which are due to reassessment two years after the entry into force of the Protocol:

- seed treatment;
- soil applications directly followed by incorporation into topsoil surface layer;
- professional remedial and industrial treatment of lumber, timber and logs;
- public health and veterinary topical insecticidal application;
- non-aerial application to tree seedlings, small-scale lawn use and use for nursery stocks and ornamentals;
- indoor industrial and residential applications.

Besides the fact that the UN ECE POPs Protocol and the UNEP POPs Convention have not been ratified by all European countries and the EU, some of the above stated permitted uses have become irrelevant in many countries anyway.

A background document on lindane is also in preparation for UN ECE under the lead of Germany and Austria. The format of this background document for the UN-ECE differs, however, from the format of the OSPAR background document and will include additional information on uses in countries outside the OSPAR Convention area.

5.1.5 Council Directive 91/414/EEC

In the year 2000 it has been decided, and published under Decision 2000/801/EC, that lindane is not anymore permitted as active substance in crop protection products under Council Directive 91/414/EEC concerning the placing of plant protection products on the market. Therefore, all Member States of the EU have to withdraw all authorisations within 6 months. Any existing stocks have to be used in no more than 18 months or in no more than one growing season. In the case of Germany, as in other European countries too, the authorisation of lindane as agricultural pesticide has already ended before that date, namely on 31 December 1997. The above stated decision has therefore practically been put into action in Germany already without making it necessary to explicitly ban lindane in the Ordinance on the Use for Plant Protection.

Based on the information available in the framework of the re-evaluation of lindane under Council Directive 91/414/EEC and the proposed conditions for use, the Standing Committee on Plant Health of the European Commission concluded that no plant protection product containing lindane is expected to fulfil the safety requirements laid down in Article 5(1) (a) and (b) of Council Directive 91/414/EEC. This conclusion has been reached primarily because the evaluation has identified concerns with regard to the safety of this active substance, in particular with regard to operators exposure, its fate and behaviour in the environment and its effects on non-target organisms (EC, 2000).

5.1.6 EU Water Framework Directive 2000/60/EC

Following the adoption of the Water Framework Directive (2000/60/EC), the European Commission was required to identify, from a list of (originally) 32 priority substances, the priority hazardous substances that are of particular concern for the aquatic environment (COM/2001/0017 final – COD 2000/0035). These substances shall be subject to cessation or phase-out of discharges, emissions and losses into surface, transitional and coastal waters within 20 years of the adoption of the pertinent proposals. The "List of Priority Substances in the Field of Water Policy" was adopted on 20 November 2001 and contains 33 substances or groups of substances, 11 of which have been designated as priority hazardous substances.

Among others, hexachlorocyclohexane has been identified as "priority hazardous substance". As a typical representative of this group of chemicals, lindane will be subject to controls and measures, without prejudice to the inclusion of other individual representatives of HCH, where appropriate. As a consequence of the adoption of priority substances, the European Commission shall produce proposals for emission controls and quality standards to be submitted within 2 years of the inclusion of the substance concerned on the List of Priority Substances. During 2001, the Commission established an Expert Advisory Forum. This forum will address all technical issues in relation to Article 16 for the priority hazardous substances, including lindane, among others, the elaboration of quality standards and emission controls, as well as other issues relating to strategies against pollution of waters in the Community.

5.1.7 Quality standards

In Germany, quality standards for pesticides in drinking water sources were established by a sub-committee on EU Quality Criteria (Länderarbeitsgemeinschaft Wasser – Unterausschuss EU-Qualitätskriterien; LAWA-UA "EUQ") and its predecessors. Quality standards are derived from ecotoxicological data for the most important trophic levels. Quality standards should not be exceeded, if possible, i.e. they give guidance for protection. In case that actual concentrations exceed the quality standard value, priorities for future measures of water protection may be justified on this basis. The quality standard for lindane with respect to aquatic biocoenoses has been set to 0,3 µg/l, while for drinking water resources a general quality standard of 0,1 µg/l is applied. Concentrations in excess of the quality standard have been registered only twice in Eastern Germany in the last years and were attributed to polluted sites on the former chemical industry area of Bitterfeld.

For the Directorate for Environmental Protection and the Directorate for Chemicals of the Netherlands, RIVM has calculated maximum permissible concentrations for surface water and soil based on direct effects as well as for birds and mammals (VAN DE PLASSCHE 1994).

Table 16: Maximum permissible concentrations (MPC) for HCHs for surface water and soil based on direct effects for birds and mammals (VAN DE PLASSCHE 1994)

| Compound | MPC freshwater (µg/l) | MPC saltwater (µg/l) | MPC aquatic combined (µg/l) | MPC soil (mg/kg) |
|----------|-----------------------|-------------------------|----------------------------------|------------------|
| α-HCH | 2,2 | 5 | 3,5 | - |
| β-HCH | 17 | 1 | 6,1 | - |
| γ-HCH | 0,86 | 0,00017 | 1,0 | 0,005 |
| Compound | MPC bird (mg/kg food) | MPC mammal (mg/kg food) | MPC bird/mammal combined (mg/kg) | |
| α-HCH | - | 5 | 5 | - |
| β-HCH | 63 | 0,2 | 0,2 | - |
| γ-HCH | 0,16 | 2,5 | 1,2 | 0,005 |

These MPCs for direct effects are also extended to secondary poisoning in this report.

5.1.8 Administrative rules concerning substances hazardous to water

Administrative rules may be found in the UNEP Legal File (<http://irptc.unep.ch/irptc/legint.html>). Quality objectives (the concentrations indicated are the minimum requirements necessary to protect aquatic life from pollution), which will be measured at a point sufficiently close to the point of discharge, are laid down in Council Directive 84/491/EEC with the object of eliminating pollution (all concentrations relate to the arithmetic mean of the results obtained over one year):

- the total HCH (means the isomers of 1,2,3,4,5,6-hexachlorocyclohexane) concentration in inland surface waters affected by discharges must not exceed 100 ng/l;
- the total concentration of HCH in estuary waters and territorial sea waters must not exceed 20 ng/l;
- in the case of water used for the abstraction of drinking water, the HCH content must conform to the requirements of Council Directive 75/440/EEC. This applies to standards at the tap only.

In addition to the above requirements, HCH concentrations in inland surface waters must be determined by the national monitoring network and the results compared with a total HCH concentration of 50 ng/l. The total concentration of HCH in sediments and/or molluscs and/or shellfish and/or fish must not increase significantly over time. Where several quality objectives are applied to waters in an area, the quality of the waters must be sufficient to comply with each of those objectives.

The WHO Guidelines for Drinking Water Quality, Volume 1 (1993 and 1998) indicate that lindane is of health significance in drinking-water. The threshold in drinking water is 2 µg/l.

According to German rules, lindane is classified as “severely hazardous to water” (Water Hazard Class 3, 1996). The classification forms the basis for water protection requirements for industrial plants in which hazardous substances are handled.

In Great Britain, an inland surface water is assigned the DS1 classification provided the prescribed annual mean concentration of 0,1 µg/l HCH (total of all isomers) and the prescribed annual mean of all other Schedule 1 substances are not exceeded. This is an implementation of an approximation of Council Directive 84/491/EEC. A coastal water and relevant territorial water is assigned the DS2 classification provided the prescribed annual mean concentration of 0,02 µg/l for all isomers of HCH and the prescribed annual mean of all other schedule 2 substances are not exceeded.

In classifying waters for suitability for abstraction for supply (after treatment) as drinking water, an inland surface water is assigned the classification DW1 where the concentration does not exceed 1 µg/l (for total of the pesticides HCH, dieldrin and parathion), DW2 where the concentration does not exceed 2,5 µg/l (for the pesticides as above), or DW3 where the concentration does not exceed 5 µg/l (for the pesticides as above), provided the concentrations of other scheduled substances do not exceed their prescribed maximum concentrations. This system of classification, which is additional to that prescribed by the Surface Water (Dangerous Substances) (Classification) Regulations 1989 (SI 2286), relates to the level of treatment required. This is an implementation of an approximation of Council Directive 75/440/EEC.

The International Maritime Organization classifies lindane with the Hazard Class 6.1 (Poisonous Substance) and PP (Severe Marine Pollutant) according to the IMDG Code (effective 1991).

5.1.9 Other administrative rules

Administrative rules may be found in the UNEP Legal File (<http://irptc.unep.ch/irptc/legint.html>).

In Germany, for example, lindane is prohibited for the use in antifouling paints according to the Hazardous Substances Ordinance (effective from 1 January 2000), as well as for the use in mills, flour silos, in stocks of grain and grain products according to the Ordinance on the Use for Plant Protection (effective from 1992, 1999). Lindane may not be used against bark beetles in removed bark, and as pouring agent and spreading agent. However, the use of lindane as wood preservative and seed treatment has become irrelevant in Germany. Products containing lindane are sometimes still used against cockroaches on retting dumps and against head lice and scabies mites according to the Act on Prevention of Infectious Diseases.

5.2 Choice of substitutes

The OSPAR Commissions' report on non-agricultural uses of pesticides (OSPAR 1999) lists several possible substitutes for lindane. According to the intended uses during the early 1990s (which was the time period covered in the report), these substitutes were:

- for wood preservative use: azaconazole, boric acid, carbendazim and pyrethrins (for domestic use); azaconazole, boric acid, chromic acid, copper/chrome/arsenic and sodium fluoride (for industrial use);
- for insecticidal use: organophosphates, permethrin, pyrethrins;
- for rodenticidal use: anticoagulants (e.g. warfarin, brodifacoum) and acute poisons (e.g. calciferol, zinc phosphide);
- for medicinal use (human and veterinary): organophosphates, pyrethrins.

Substitutes from other insecticide compound groups (e.g. pyrethroids, carbamates, organic phosphoric esters and neonicotinoid compounds) are not necessarily more environmentally acceptable. Nevertheless, it has been demonstrated that major crops which previously used to be treated with lindane may also be grown without the use of synthetic insecticides (i.e. by organic farming).

6. Choice for action

The data and information given in this background document provide clear evidence and proof that lindane poses a serious threat to the marine environment. Therefore any further use of lindane, which could lead to inputs to and pollution of the OSPAR maritime area, should be avoided, banned or at least severely restricted.

6.1 The use of lindane as an agricultural pesticide

With the Commission Decision 2000/801/EC of 20 December 2000 excluding lindane from Annex I of Council Directive 91/414/EEC concerning the placing on the market of plant protection products and the

consequent withdrawal of authorisations for plant-protection products containing this active substance, all uses of lindane as an agricultural pesticide in the EU will cease in June 2002 (i.e. 18 months after the date of the Decision). Provided that there are no illegal uses of lindane after this deadline, no further action is needed by OSPAR to regulate or control the use of lindane as an agricultural pesticide. Therefore,

- OSPAR should simply note that all uses of lindane as an agricultural pesticide will cease in June 2002, which means that one of the main sources of inputs of lindane to the marine environment in previous decades will be phased out.

6.2 The need to severely restrict or ban other remaining uses of lindane

The data and information given in this background document demonstrate that other remaining uses of lindane, minor though they may be, should be severely restricted or banned, in order to achieve the goals of the OSPAR Strategy with regard to Hazardous Substances and its 2020 timeframe with respect to chemicals on the OSPAR List of Chemicals for Priority Action.

Under the EU Water Framework Directive 2000/60/EC (WFD) a list of priority substances, including the identification of priority hazardous substances, has been established. The hexachlorocyclohexane isomers are included on this list as a priority hazardous substance, with lindane being the "indicative parameter" for all HCH isomers. This also means that the remaining uses of lindane will be subject to EU controls and measures which will contribute to reaching the OSPAR goals by 2020. Under Article 16(4) of the Water Framework Directive, the list of Priority Substances is due to be reviewed by December 2004. If, however, lindane will lose its role as representative for the HCH isomers due to the initiated phase-out in Europe, the focus of attention should be directed towards the other HCH isomers. This would enlarge the scope of safeguarding the marine environment from sources of technical HCH (i.e. a mixture of several HCH isomers) from outside the EU. Therefore,

- OSPAR / EU Member States should assist the European Commission in the above review with a view to supporting monitoring and measures for environmental control of all HCH isomers by providing information on the risk for the marine environment of all other HCH isomers.

Since it is not yet possible to judge to what extent measures resulting from the work in progress in the EU will enable the OSPAR 2020 target to be fully achieved for lindane in general, OSPAR should by the end of 2004:

- review what is likely to be achieved by the EC measures that have by then been adopted;
- consider the need for further OSPAR actions with respect to any remaining other uses of lindane in order to achieve the year 2020 target.

6.3 The need to reduce the atmospheric inputs of lindane into the maritime area

There is proof that lindane undergoes atmospheric long-range transport and deposits of these chemicals can be found even in remote areas, including Arctic regions of the OSPAR maritime area.

Lindane is included on the list of priority persistent organic pollutants (POPs) established under the POP Protocol of the UN-ECE Convention on the Long-Range Transfer of Airborne Pollution and a background document on lindane is in preparation for UN-ECE under the lead of Austria and Germany. However, lindane is not included in the list of POP substances established under the UNEP POP Convention, 2001. Therefore,

- OSPAR Contracting Parties should undertake co-ordinated efforts within the framework of UN-ECE LRTAP towards a severe restriction or ban of all uses of lindane in countries where products containing lindane are still in use, in order to reduce the atmospheric long-range transport and the inputs of these chemicals into the OSPAR maritime area via airborne deposition;

- OSPAR Contracting Parties which are or will become Contracting Parties to the UNEP POP Convention, 2001 should undertake co-ordinated efforts that lindane is included in the next revision of the list of POP substances established under this Convention.

6.4 The need for a continuation of monitoring and assessment activities with respect to lindane

Even if there will be in the near future no remaining uses of lindane in OSPAR Contracting Parties, there is a continued need to monitor these chemicals under OSPAR in order to establish the effectiveness of national and internationally agreed measures and to ensure that their implementation fulfils the obligations under OSPAR's Strategy with regard to Hazardous Substances. Furthermore, such monitoring could reveal the sources and inputs to the marine environment from continued uses of HCHs outside of the OSPAR Convention area. Therefore,

- OSPAR should continue monitoring lindane in terms of:
 - inputs via rivers and direct discharges (RID programme) and via the atmosphere (CAMP programme);
 - marine environmental concentrations in water, sediment and biota;
- OSPAR should invite Germany to review the existing monitoring requirements and assessment tools (including ecotoxicological assessment criteria) with respect to lindane under OSPAR and other relevant international organisations with a view to developing proposals for examination by OSPAR after the implementation of the WFD monitoring requirements, as appropriate, for:
 - the level and the type of future monitoring and/or assessment activities required;
 - an improved co-ordination of the monitoring and assessment activities carried out by Contracting Parties individually or collectively in the framework of OSPAR and other international organisations, and especially with the EU and WFD activities;
 - modifications or additional monitoring and assessment activities to be initiated under OSPAR.

6.5 The need for consistency with actions by other bodies

To ensure that the information in this background document and the conclusions reached by OSPAR are brought to the attention of the European Commission,

- OSPAR should formally communicate this background document to the European Commission.

To ensure that the information in this background document can be considered in the context of other international agreements, which deal with hazardous substances and to which Contracting Parties are associated,

- OSPAR should send copies of this background document to the appropriate bodies dealing with those agreements and invite Contracting Parties who are common parties to OSPAR and those other agreements to promote action to take account of this background document by those other international bodies in a consistent manner.

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Appendix 1: Physico-chemical Properties of lindane

| Property | Value | Source |
|--|--|---|
| Chemical name | 1,2,3,4,5,6-Hexachlorocyclohexane, (1,2,4,5/3,6)- γ -Isomer | IVA (2000) |
| CAS-No. | 58-89-9 | Rippen (1990/2000) |
| EEC No. (EINECS) | 200-401-2 | Rippen (1990/2000) |
| Molecular formula | C ₆ H ₆ Cl ₆ | Rippen (1990/2000) |
| Molecular Mass M [g/mol] | 290,83 | Rippen (1990/2000) |
| Physical state of matter at 20 °C | solid (crystals) | Rippen (1990/2000) |
| Colour / odour | colourless / essentially odourless | Rippen (1990/2000) |
| Detection limit [µg/l] (min. - max.; medium) | 0,00005 – 0,1 0,02 | Bach et al. (2000) |
| Solubility in water S [g/l] | 6,2 (20 °C) 7,3 (25 °C) 7,4 (25 °C) 8,35 (25 °C, ph 5) | Mean from Rippen (1990/2000) EC-JRC (1998) Mean from Rippen (1990/2000) IUCLID (1994) |
| Solubility in ethanol [g/l] | > 30 - 40 | IVA (2000) |
| log P _{ow} | 3,2 – 3,7 3,5 3,5 (22 °C) 3,61 3,61; 3,72 3,66 3,69 | WHO (1991) IVA (2000) IUCLID (1994) Howard (1991) EC-JRC (1998) Mean from Rippen (1990/2000) Hühnerfuß (1995) |
| Adsorption coefficient log K _{oc} [l/kg] | 2,97 3,28 (n=18) 3,51 (n=5) | Hühnerfuß (1995) Mean from Rippen (1990/2000) Mean from Rippen (1990/2000): Susp. matter in estuary |
| Melting point T _m [°C] | 112,6 | Rippen (1990/2000) |
| Boiling point T _b [°C] | 288 (760 hPa) 323 (decomp.) 323,4 (571 hPa) | IUCLID (1994), WHO (1991) Rippen (1990/2000) EC-JRC (1998), Howard (1991) |
| Vapour pressure P [Pa] | 1,25·10 ⁻³ 1,33·10 ⁻³ (20 °C) 2,48·10 ⁻³ (24 °C) 2,9·10 ⁻³ (20 °C) 4,34·10 ⁻³ (20 °C) 4,4·10 ⁻³ (24 °C) 5,6·10 ⁻³ (20 °C) 7,30·10 ⁻³ (25 °C) 7,43·10 ⁻³ (25 °C) | EC-JRC (1998) “ IUCLID (1994) Mean from Rippen (1990/2000) WHO (1991) IVA (2000) The Pesticide Manual (1983) Mean from Rippen (1990/2000) Howard (1991) |
| Henry's Law constant [-] | 5,6·10 ⁻⁵ (20°C) ^a 8,3·10 ⁻⁵ (23°C) ^c 1,2·10 ⁻⁴ (25 °C) ^a | Rippen (1990/2000) Rippen (1990/2000) Howard (1991) |
| Volatilisation from soil | in 24 h 28% in 75 h 32% | Rippen (1990/2000) (volat. from plants is higher) |

| Property | Value | Source |
|---|--|---|
| Rate of dry deposition [cm/s] | 0,04 | Rippen (1990/2000) |
| Reactivity with OH-radicals atmospheric half-life [d] | Air: 1,6 ^b Aerosol: 67 $\tau = 96 \text{ d } (9,7 \cdot 10^5 \text{ rad/cm}^3 \text{ at } 277 \text{ }^\circ\text{K})^a$ $\tau = 84 \text{ d } (5 \cdot 10^5 \text{ rad/cm}^3 \text{ at } 298 \text{ }^\circ\text{K})^a$ | Rippen (1990/2000) “ BRUBAKER & HITES 1998 pers. comm. B. WAGNER (UBA) |
| Hydrolysis half-life [d] | under normal environmental conditions: months to years | Rippen (1990/2000) |
| Toxicological classif. (EU) | T – toxic Xi – irritant N – dangerous to the environment | IVA (2000) UNEP Legal File |
| R-Phrases | R 23/24/25, R 36/38 R 50, R 53 | IVA (2000) UNEP Legal File |
| S-Phrases | S 1/2, S 13, S 45, S 60, S 61 | UNEP Legal File |

a) Calculated values

b) Estimated values

c) Measured values

Appendix 2: Monitoring strategy for lindane

As part of the Joint Assessment and Monitoring Programme (*reference number 2003-22*), OSPAR 2004 adopted an Agreement on monitoring strategies for OSPAR Chemicals for Priority Chemicals (*reference number 2004-15*) to implement the following monitoring for tracking progress towards the objectives of the OSPAR Hazardous Substances Strategy (*reference number 2003-21*) with regard to lindane. The Monitoring Strategy for lindane will be updated as and when necessary, and redirected in the light of subsequent experience.

The mainly diffuse sources of lindane are well known and have been detailed in the Background Document and are also identified in the EC WFD fact sheet on lindane.

Lindane is heavily regulated by existing legislation. OSPAR will therefore evaluate progress with respect to the implementation of such legislation in the EC as a tool for assessing the progress towards the objectives of the Hazardous Substances Strategy.

Inputs of lindane to the NE Atlantic are already monitored through RID and CAMP and monitoring will be continued, to demonstrate, if inputs of lindane still occur.

Lindane is not currently measured as part of the CEMP, but will be part of the monitoring under the EC Water Framework Directive in coastal and transitional waters in which context the target matrix will be water (whole water). OSPAR will seek to make use of the results of monitoring carried out under the WFD. The review of the CEMP in 2005-06 will consider whether to add an additional appendix on lindane and other HCH isomers. This will review will take into account:

- a. whether the data that will be available from WFD monitoring are sufficient for OSPAR to assess whether Lindane can still be detected in the wider marine environment;
- b. whether monitoring in biota mussels or fish liver is the most appropriate target, given that lindane has potential for bioaccumulation at the lower end of the food chain.

In the meantime Contracting Parties are urged to extend their monitoring programmes to cover Lindane and other HCH isomers.

Since the WFD is using Ecological Quality Standards (EQS) as legally binding limit values for lindane in water and the OSPAR ultimate aim for lindane should be close to zero concentrations, it is not considered necessary to develop OSPAR EACs as assessment tools.

| LINDANE MONITORING STRATEGY | |
|---|---|
| Implementation of actions and measures | <ul style="list-style-type: none"> Examination of progress in the implementation of regulations on marketing and/or use or emission and/or discharge which have been agreed, or are endorsed, by the Background Document |
| Atmospheric inputs | <ul style="list-style-type: none"> Monitoring will continue under the CAMP |
| Riverine inputs | <ul style="list-style-type: none"> Monitoring will continue under the RID study |
| Maritime area: | |
| Concentrations in water | <ul style="list-style-type: none"> Where available, data will be periodically compiled from EC WFD monitoring |
| Concentrations in biota | <ul style="list-style-type: none"> The review of the CEMP in 2005-06 will consider whether to add an additional appendix on lindane and other HCH isomers. Any proposal for such an appendix should consider whether for biota monitoring in mussels or fish liver is most appropriate <p><i>Additional voluntary activity:</i></p> <ul style="list-style-type: none"> <i>In the meantime, Contracting Parties will be encouraged to extend their monitoring programmes to cover HCH isomers and to report results on a voluntary basis through the data-handling mechanism operated by ICES for the CEMP</i> |