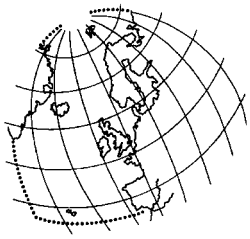
Hazardous Substances Series

Hexachlorocyclopentadiene (HCCP)*



OSPAR Commission

2004

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* OSPAR 2005 agreed to deselect Hexachlorocyclopentadiene from the List of Chemicals for Priority Action (OSPAR 05/21/1, §7.5a).

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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Contents

Executive summary

Hexachlorocyclopentadiene (HCCP) (synonym: 1,3-Cyclopentadiene, 1,2,3,4,5,5-hexachloro-), CAS nr. 77-47-4, was included on the OSPAR List of Chemicals for Priority Action at OSPAR 2000. HCCP is used as an intermediate in the production of many chlorinated pesticides like dieldrin, aldrin, endrin, endosulfan and chlordane. It is also used as an intermediate in the production of substances used in the production of flame-retardants and corrosion-proof polyesters and alkydresins.

The evaluation of environmental risk is based on the draft risk assessment report (RAR) prepared under the EU Council Regulation (EEC) 793/93, the existing substances regulation. The data about degradation in aquatic environment are scarce. A hydrolysis half-life in freshwater of 13,9 days at a temperature of 12°C and pH of 7 has been estimated. On the basis of the available data on aquatic (fresh water) biodegradation, it is concluded in the draft RAR that HCCP is inherently biodegradable. However, since other evidence suggests that biodegradation is a relatively unimportant process in aquatic systems, the RAR gives a worst case rate constant of zero, and concludes that HCCP should be regarded as potentially persistent, thus fulfilling the Technical Guidance Document (TGD) persistency (P) criterion. On the other hand, the estimated half-life in freshwater is well below the OSPAR cut-off value of 50 days. The P criterion is therefore not fulfilled according to the OSPAR criterion.

The octanol/water partition coefficient of HCCP indicates a substantial potential for bioaccumulation, but HCCP has been found to be rapidly metabolised and eliminated in a number of studies. In the draft RAR it is concluded that HCCP does not meet the TGD bioaccumulation (B) criterion. The bioconcentration factor (BCF < 11) is also far below the OSPAR cut-off value, so the OSPAR B-criterion is not fulfilled either. The draft RAR concludes that HCCP meets the TGD toxicity (T) criterion. The OSPAR T-criterion is met as well, as the reported no effect concentrations (NOECs) are below the OSPAR cut-off value.

In conclusion, the available information indicates that HCCP is not a PBT substance, neither according to the EU TGD criteria, nor according to the OSPAR criteria. Therefore, neither identifying the main sources of HCCP and its various pathways into the marine environment, nor reviewing the various controls to limit discharges, emissions and losses of HCCP were deemed necessary. On this basis, no further actions are needed by OSPAR. A decision on whether to delete HCCP from the OSPAR List of Chemicals for Priority Action must await the finalisation of the EC risk assessment. It is not currently appropriate to develop a monitoring strategy for HCCP.

Récapitulatif

L'hexachlorocyclopentadiène (HCCP) (synonyme: 1,3-Cyclopentadiene, 1,2,3,4,5,5-hexachloro-), N° CAS 7-47-4, a été inscrit en 2000 sur la liste OSPAR des produits chimiques devant faire l'objet de mesures prioritaires. Le HCCP sert d'intermédiaire dans la fabrication de nombreux pesticides chlorés, tels que la dieldrine, l'aldrine, l'endrine, l'endosulfan et le chlordane. Il est également employé comme intermédiaire dans la fabrication de substances servant à la production de retardateurs de flammes ainsi que de polyester et de résines alkylées à l'épreuve de la corrosion.

L'évaluation du risque environnemental est basée sur le projet de rapport d'évaluation des risques (RAR) dressé en vertu du règlement du Conseil (CEE) 793/93, de l'Union européenne, à savoir le règlement visant les substances existantes. Les données relatives à la dégradation dans le milieu aquatiques sont rares. L'on a estimé que la demi-vie de l'hydrolyse dans l'eau douce était de 13,9 jours à une température de 12° C et un pH de 7. Sur la base des données en possession sur la biodégradation en milieu aquatique (eaux douces), il est conclu dans le projet de RAR que le HCCP est intrinsèquement biodégradable. Toutefois, puisque d'autres indices donnent à penser que la biodégradation est un processus présentant relativement peu d'importance dans les systèmes aquatiques, le RAR donne un taux constant de 0 dans le pire des cas et conclut que le HCCP devrait être considéré comme potentiellement persistant, et qu'il remplit ainsi le critère de persistance (P) fixé dans le document d'orientation technique (DOT). D'un autre coté, l'estimation de la demi-vie dans l'eau douce est nettement inférieure à la valeur de coupure fixée par OSPAR, soit 50 jours. Le critère de P fixé par OSPAR n'est par conséquent pas rempli.

Le coefficient de partage octanol/eau du HCCP indique un important potentiel de bio-accumulation, quoique dans plusieurs études, l'on ait constaté que le HCCP était métabolisé et éliminé rapidement. Dans le projet de RAR, il est conclu que le HCCP ne répond pas au critère de bio-accumulation (B) du DOT. Le coefficient de bio-concentration (BCF <11) est également nettement inférieur à la valeur de coupure fixée par OSPAR, de telle sorte que le critère B d'OSPAR n'est pas rempli non plus. Dans le projet de RAR, il est conclu que le HCCP répond au critère de toxicité (T) du DOT. Le critère T d'OSPAR est également rempli, car les teneurs sans effet qui ont été signalées (NOEC) sont inférieures à la valeur de coupure OSPAR.

En conclusion, les renseignements disponibles indiquent que le HCCP n'est pas une substance PBT, et ceci que ce soit en fonction des critères du DOT de l'Union européenne ou du critère OSPAR. Par conséquent, il est considéré qu'il est inutile de déterminer les principales sources de HCCP ainsi que ses diverses voies de pénétration dans le milieu marin, ni de revenir sur les divers règlements qui visent à limiter les rejets, les émissions et les pertes de HCCP. Sur cette base, il n'y a pas lieu qu'OSPAR prenne de quelconques nouvelles mesures. Il est impératif que la décision de retrait ou non du HCCP de la liste OSPAR des produits chimiques devant faire l'objet de mesures prioritaires attende que l'évaluation communautaire des risques soit terminée. Il n'y a pour l'instant pas lieu d'élaborer de stratégie de surveillance du HCCP.

Introduction

Hexachlorocyclopentadiene (HCCP), CAS nr. 77-47-4, is on the OSPAR List of Chemicals for priority action. HCCP was included in the list at OSPAR 2000, after the first application of the DYNAMEC mechanism. At the Commission meeting the Netherlands offered to act as the lead country for drawing up a background document.

Sources of Information

The presented information has been taken exclusively from the first draft risk assessment report (RAR) (see Appendix 1) prepared by the Netherlands as the rapporteur for HCCP under the EU Council Regulation (EEC) 793/93 (Existing substances regulation).

PBT assessment

According to the draft RAR, the degradative processes for removal of HCCP from water include photolysis, hydrolysis and biodegradation. A reported experimentally determined hydrolysis half-life of 3,3 days at pH 7 and 30°C has been recalculated in the draft RAR to a hydrolysis half-life of 13,9 days at 12°C. In shallow or flowing waters, photolysis is the predominant fate process, proceeding much faster than hydrolysis. On the basis of the available data on aquatic biodegradation, it is considered in the draft RAR that HCCP is inherently biodegradable, however not under all circumstances. Therefore, as a worst case assumption, a rate constant of 0 h^{-1} is given, and the conclusion is that the substance should be regarded as potentially persistent, thus fulfilling the Technical Guidance Documents (TGD, 2002) P-criterion. However, the half-life of 13,9 days for hydrolysis at 12°C is well below the DYNAMEC cut-off value of 50 days (see Dynamic Selection and Prioritisation Mechanism for Hazardous Substances (DYNAMEC), OSPAR Commission 2000).

With respect to bioaccumulation, the draft RAR mentions a number of experimental studies in which HCCP was found to be rapidly metabolised and eliminated. Although the log K_{ow}-value of 5,04 and the Quantitative Structure Activity Relationship (QSAR) estimates for the bioconcentration factor (BCF) point to a significant bioaccumulation potential, several reliable experimentally determined BCFs are relatively low. US-EPA concluded to use a BCF of < 11 (with adjustment for lipid content). In the draft RAR it is concluded that HCCP does not meet the TGD B-criterion (TGD, 2002). The BCF of < 11 is also far below the OSPAR cut-off value (500) (see DYNAMEC).

Based on the lowest no observed effect concentrations (NOECs) found for freshwater and marine organisms, 0,0037 and 0,0003 mg/l, respectively, the draft RAR concludes that HCCP meets the TGD T-criterion (TGD 2002). The OSPAR T-criterion is met as well, as the reported NOECs are below the OSPAR cut-off value (0,1 mg/l) (see DYNAMEC).

Conclusion

The available information indicates that HCCP is not a PBT substance, neither according to the TGD criteria, nor according to the OSPAR criteria. On this basis, the Netherlands considered that no further actions are needed by OSPAR or other relevant international organisations. Therefore, neither identifying the main sources of HCCP and its various pathways into the marine environment, nor reviewing the various controls to limit discharges, emissions and losses of HCCP were deemed necessary.

Appendix 1: Extracts from the draft EU Risk Assessment Report prepared under Council Regulation (EEC) 793/93 (EU RAR 2003)

(Hexachlorocyclopentadiene (HCCP), Risk Assessment, Draft Report 26 February 2003, The Netherlands)

"(…)" indicates that text was left out as it was considered irrelevant for the purpose of this appendix.

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3.1.1.2.4 Discussion of degradation data

The half-life for HCCP in the atmosphere has been estimated to be 29 days. However, using the ionisation potential as physical parameter, the half-life of HCCP after reaction with hydroxyl radicals would be estimated to be less than 1 day. Based on the highly chlorinated structure of HCCP, it is expected that reaction of this compound with ozone molecules in the atmosphere would be too slow to be environmentally significant.

Degradative processes for removal of HCCP from water include photolysis, hydrolysis and biodegradation. Hydrolysis of HCCP in water occurs much more slowly than photolysis. In shallow or flowing waters, photolysis is the predominant fate process; in deeper waters hydrolysis and biodegradation may be more important environmental fate processes (US-EPA, 1984).

In the study from Wolfe et al. (1982) a hydrolysis half-life of 3,3 days was found at pH 7 and 30°C. For risk assessment purposes for fresh water a pH of 7 and a temperature of 12°C will be established which is conform to the standard environmental parameters. The hydrolysis half-life reflecting an average EU outdoor temperature can be recalculated by the equation:

DT50(X°) = DT50(t) . e(0,08.(T-X))

DT50(12°C) = 3,3 (days) . e(0,08.(30-12))

DT50(12°C) = 3,3 (days) . 4,2 = 13,9 days

(….)

HCCP can be biodegraded in aquatic media under laboratory conditions as was seen in the study from Atallah et al. (1981), although evidence is considered to be weak. However, another study on the fate of HCCP found biodegradation to be a relatively unimportant process in aquatic systems, based on the observation that there was no detectable difference in hydrolysis rates between sterile and non-sterile studies and measured numbers of micro-organisms (Wolfe et al., 1982). It is difficult to differentiate removal or degradation via abiotic processes (adsorption, volatilisation, and hydrolysis) from that via biodegradation.

HCCP is a volatile, hydrophobic substance, which will be metabolised, strongly adsorbs to organic carbon and will not be mineralised aerobically. Under anaerobic conditions dehalogenisation will occur and one or more chlorinated metabolites will be formed. HCCP will hydrolyse to some extent.

On the basis of the available data on aquatic biodegradation, HCCP is considered to be inherently biodegradable, not fulfilling specific criteria (rate constant 0 h^{-1}). This is a rather worst case assumption, but adequate data are lacking to make a more balanced decision on this issue. The rate constant k will be greater than 0 h^{-1} under some conditions (expert judgement).

The persistence of HCCP in soil is low, with degradation of >90% of applied HCCP to non-polar products within approximately 7 days. Factors contributing to this loss include abiotic and biotic degradation processes and volatilisation, although the relative importance of each is difficult to quantify given the limited information available. As no half-life in soil can be derived from the experimental data presented, the use of screening data may be considered. Degradation half-life classes for soil, partly based on K_p can be used. As HCCP has a K_{psoil} of lower than 100 l/kg and the substance is considered to be inherently biodegradable, a half-life of 300 days is chosen (TGD, 2002).

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3.1.1.3.2 Accumulation and metabolism

The log Kow of HCCP has been experimentally determined to be 5,04, which would indicate a substantial potential for bioconcentration, bioaccumulation and biomagnification. With EPIWIN a BCF of 1516 was estimated (log Kow 5,04) and with EUSES the BCF would amount to 3800. Actual determinations indicate that HCCP does not seem to accumulate to a great extent (Podowski, 1979), mainly because it is metabolised rapidly (studies described in more detail below).

In the following section bioconcentration factors (BCF) and bioaccumulation factors (BAF) are reported. Bioconcentration implies that tissue residues result only from exposure to the ambient environment (e.g. air for terrestrial or water for aquatic species). Bioaccumulation considers all exposures (air, water and food) of an individual organism as the source of tissue residues.

Podowski and Khan (1979, 1984) conducted several experiments concerning the uptake, bioaccumulation and elimination of ^{14}C-HCCP in goldfish and concluded that the species eliminated absorbed HCCP rapidly. In one experiment, fish were transferred daily into fresh solutions of ^{14}C-HCCP for 16 days. This transfer of three fish/jar resulted in accumulative exposure of 240 μg of HCCP. Nominal HCCP concentrations of 10 μg/l resulted in measured water concentrations in the range of 3,4-4,8 μg/l, because of rapid volatilisation of the compound. Radioactivity accumulated rapidly in fish tissue, reaching a maximum on day 8 corresponding to 6 mg HCCP/kg. Since an undetermined amount of the radioactivity was present as metabolites, no reliable bioconcentration factor can be calculated. From day 8 to day 16, tissue levels declined in spite of daily renewal of exposure solutions, indicating that excretion of HCCP and/or its metabolites was occurring more rapidly than uptake. In a static exposure to an initial measured HCCP concentration of 5 μg/l, radioactivity was taken up by the fish to a level corresponding to 1,6 mg HCCP/kg on day 2, accompanied by a slight decrease of HCCP in the water. By day 4, approximately 50% of the absorbed activity had been excreted, and the water level increased. Over the following 12 days, radioactivity in both water and fish declined slowly.

Podowski and Khan (1979, 1984) also studied elimination, metabolism and tissue distribution of HCCP injected intraperitoneally into goldfish and concluded that goldfish eliminate injected HCCP both rapidly and linearly (biological half-life approx. 9 days). Fish (27-45 g) were injected with 39,6 μg of ^{14}C-HCCP and analysed 3 days later. Of the 97% of the radiolabelled dose accounted for, the fish eliminated approx. 18,9%, leaving a residual of 78,1%. Of the residue found in the fish, 47,2% was extractable in organic solvent; 10,6% was water-soluble metabolites, and 20,3% was unextractable. None of the metabolites were identified. Biphasic elimination was observed, rapid at first, followed by a slower phase. Based on a study of goldfish injected with ^{14}C-HCCP, the elimination of HHCP occurs in multiple stages, with a reported half-life in the organism of 7 days and predicted clearance of 90 to 95% of the chemical after 162 and 211 days, respectively (Podowski et al., 1991).

Veith (1979, cited in US-EPA, 1984) determined the BCF for HCCP to be 29 in the fathead minnow. In a 32-day flow-through study, 30 fish were exposed to HCCP at a mean concentration of 20,9 μg/l and were sacrificed five at a time for residue analysis at 2, 4, 8, 16, 24 and 32 days. The study was conducted using Lake Superior water at 25ºC (pH 7,5, dissolved oxygen >5,0 mg/l and hardness 41,5 mg/l as $CaCO_3$). On the basis of its estimated octanol/water partition coefficient alone (log P = 5,51; value from Veith!), a BCF of circa 9600 would have been predicted. However, HCCP did not bioconcentrate substantially, and therefore deviated from the log P:log BCF relationship shown for most of the 29 chemicals tested.

Spehar (1979) conducted a 30-day early-life stage, flow-through toxicity test at 25°C with the fathead minnow. HCCP residues in the fish after 30 days of continuous exposure to HCCP were <0,01 mg/kg for all concentrations tested (0,78-9,1 μg/l), and the BCF was <11 (0,1 mg/kg in fish divided by 9,1 μg/l in water).

Kotzias (1980) and Freitag (1982, 1984, 1985) examined the bioaccumulation of ^{14}C-HCCP in the goldfish. Following 24 hours of exposure to a concentration of 50 μg/l under static conditions at 20 to 25°C a bioaccumulation factor of 308 (Kotzias 1980) was recorded and of 1230 after three days of exposure (Freitag, 1982, 1984, 1985). There is apparently no equilibrium between uptake and elimination after 24 hours. In this investigation no account was taken of a potential metabolism since only the radioactivity in the fish and in the water respectively was measured in order to determine the concentrations of HCCP.

Lu et al. (1975) studied the fate of HCCP in a model terrestrial-aquatic ecosystem maintained at 26,7°C with a 12-hour photoperiod. They also studied the metabolism of HCCP by the organisms present in the model terrestrial-aquatic ecosystem. They reported that unmetabolized HCCP represented large percentages of the total extractable ^{14}C, being 33% in algae, 50% in snail, 46% in mosquito and 41% in fish. Percent biotransformation was calculated for each organism: 4% for the algae, 10% for the snail, 2% for the mosquito

and 27% for the fish. However, these values may underestimate the extent of metabolism, since acetone extractable polar compounds were not considered in the calculations.

Results on the bioaccumulation potential (BAF) of HCCP by the green alga *Chlorella fusca* were reported by Geyer (1984) and Freitag (1982, 1984, 1985). The concentrations of HCCP in the alga and in water were determined after 24 hours of exposure under static conditions at 20 to 25°C to a concentration of 50 μg/l. The BAF for algae was 1090. In a different study (Kotzias, 1980), the bioaccumulation factor in the green alga, *Chlorella fusca*, obtained under the same experimental conditions was given as 1140.

Discussion of bioaccumulation data

Based on the experimentally derived octanol/water partition coefficient of 5,04, HCCP would be predicted to have a BCF of about 1516 (EPIWIN calculations). The TGD QSAR predicts a value of 3800. Investigations on the bioconcentration factor of HCCP, however, show much lower values. Veith et al. (1979) did found that HCCP does not follow the log Kow:log BCF relationship and measured a BCF of 29 in the fathead minnow. Spehar et al. (1979) reported also a low BCF (<11) in the same species under comparable conditions (flow-through study, test duration about 30 days, no use of radiolabelling).

In ^{14}C studies higher BCF-values were reported (323 and 1297 in Goldfish, Podowski, 1979, 1984 and 1230 in the Mosquitofish, Freitag, 1982, 1984, 1985). Since the body residues as well as the radioactivity in water included products other than HCCP, the calculated bioconcentration ratios (BCF), based on total radioactivity, may not give the precise estimate of bioconcentration of HCCP. If the BCFs were based on parent HCCP and not total extractable radioactivity, the BCFs would likely be smaller. Furthermore, the studies of the biotransformation of HCCP in goldfish are complicated by the fact that HCCP and its metabolites are very reactive, many being very volatile and extremely lipophilic. Therefore, these studies should not be used for any qualitative and quantitative representation of HCCP transformation.

Although QSAR estimates for the BCF point to a significant bioaccumulation potential, HCCP was found to be rapidly metabolised and eliminated in a number of studies. This is reflected in relatively low experimental BCFs. US-EPA concluded to use the BCF of <11 and adjusted it for lipid content. The weighted average BCF for the edible portion of freshwater and estuarine aquatic organisms was calculated and found to be 4,34 (ATSDR, 1999). The BCF value of less than 11 will be used in EUSES as this steady-state bioconcentration factor was measured in 30-day flow through exposures to constant levels of HCCP.

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3.2.1.1 Toxicity test results

(...)

3.2.1.1.2 Fish

(…)

3.2.1.1.3 Acute toxicity

(....)

3.2.1.1.4 Long-term toxicity

Table 3.18. Long-term toxicity to fish

Species	Method	Duration [days]	Criterion	Value [μg/l]	Endpoint	Reference
Fathead minnow (larvae, <0,1 g) (*Pimephales promelas)*	FT, M	30	NOEC	3,7	survival	Spehar (1977, 1979)

S = static; FT = flow-through; U = unmeasured concentrations; M = measured concentrations

In a 30-day early-life stage flow-through toxicity test with fathead minnows using 1 day old larvae the 96-h LC50 value was 7 μg/l (measured concentration; see section 3.2.1.1.3). The lowest concentration causing 50% mortality was reached within 4 days. Furthermore, HCCP residues found in fathead minnows at the end of the 30-day exposure period were low (< 0,1 μg/g), and a BCF value of < 11 was reported. Based on the

toxicity and growth data it can be concluded that 3,7 μg/l is the highest concentration of HCCP that produces no adverse effects on fathead minnow larvae.

3.2.1.1.5 Aquatic invertebrates

(…)

The following table shows the chronic toxicity data for invertebrates.

Table 3.20. Long-term toxicity to freshwater and marine invertebrates

Species	Method	Duration [days]	Criterion	Value [μg/l]	Endpoint	Reference
Freshwater species						
Daphnia magna	UBA (1984)	21	NOEC	9	Reproduction	Kuehn (1989)
Marine species						
Mysid shrimp (*Mysidopsis bahia)*	FT, M	28	NOEC	0,3	Reproduction	cited in US-EPA (1984)

S = static; FT = flow-through; U = unmeasured concentrations; M = measured concentrations

Kühn reported a 21-day NOEC of 9 μg/l (nominal?) for Daphnia magna. In the unpublished study from US-EPA, groups of 40 mysid shrimp were exposed for 28 days in a flow-through system. Measured concentrations were found to be about one-half of the nominal ones. Mortality occurred in all concentrations except the control, but showed no consistent dose-response relationship. Reproduction, however, was more clearly related to dose (NOEC of 0,3 μg/l).

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3.4 PBT-assessment

Persistence

HCCP is considered to be inherently biodegradable in the risk assessment with a rate constant of 0 h^{-1} and should be regarded as potentially persistent.

When using the three models of BIOWIN, HCCP is predicted to be not fastly biodegradable with the non-linear model and not readily biodegradable with the MITI non-linear model. The ultimate biodegradation timeframe model does not support these results. Only when all three models give the same result a prediction of the biodegradability can be made. Based on this model the biodegradability of HCCP is unclear.

The current information on the abiotic degradation of HCCP, hydrolysis in particular, is insufficient to draw a conclusion on the 'real situation' in the marine environment.

HCCP is potentially persistent. As the B criterion is not met (see below) further testing on P is not considered necessary.

Bioaccumulation

Based on the log Kow-value of 5,04, HCCP would be considered to potentially fulfil the B-criterion. However, as a reliable BCF-value of less than 11 is derived from experimental data, the substance is not expected to fulfil the B-criterion.

HCCP does not meet the B-criterion of the PBT-criteria.

Toxicity

The lowest NOECs for freshwater and marine organisms were found to be 0,0037 and 0,0003 mg/l, respectively. This is clearly under the cut-off value of 0,01 mg/l.

HCCP meets the T-criterion in the PBT-assessment.

Overall, HCCP does not meet the PBT criteria.

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