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Assessment of the Applicability for Pollution Control Authorities of the Concept "Critical Load" of Long -range Transported Micropollutants in Relation to Aquatic and Terrestrial Ecosystems

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Norwegian Institute for Air Research (NILU)

Norwegian Institute for Nature Research (NINA)

Norwegian Institute for Water Research (NIVA)

Norwegian Polar Institute (NP)

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NATURENS TÅLEGRENSER

Ministry of Environment
Research report no. 92

Naturens Tålegrenser

Programmet Naturens Tålegrenser ble satt igang i 1989 i regi av Miljøverndepartementet. Programmet skal blant annet gi innspill til arbeidet med Nordisk Handlingsplan mot Luftforurensninger og til pågående aktiviteter under Konvensjonen for Langtransporterte Grensoverskridende Luftforurensninger (Genevekonvensjonen). I arbeidet under Genevekonvensjonen er det vedtatt at kritiske belastningsgrenser skal legges til grunn ved utarbeidelse av nye avtaler om utslippsbegrensning av svovel, nitrogen og hydrokarboner.

En styringsgruppe i Miljøverndepartementet har det overordnete ansvar for programmet, mens ansvaret for den faglige oppfølgingen er overlatt en arbeidsgruppe bestående av representanter fra Direktoratet for naturforvaltning (DN), Norsk polarinstitutt (NP) og Statens forurensningstilsyn (SFT).

Arbeidsgruppen har for tiden følgende sammensetning:

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Abstract

The applicability for pollution management and control authorities of the consept " Critical load" of long-range transported micropollutants has been evaluated for sensitive parts of aquatic and terrestrial ecosystems. The background for this assessment is the previous successful use of the critical load approach in international negotiations aiming at the reduction of acid precipitation components. The review of discharge and deposition data for micropollutants showed that this information was inadequate to calculate load with sufficient accuracy except for lead (and possibly cadmium). The reviews about observed effects in Norwegian water, soil, terrestrial and Arctic environments all concluded that effects on population levels from long-range transported persistent organics and metals could not be documented so far. As and alternative approach in international negotiations to reduce spreading of micropollutants it is recommended to use the precautionary principle. It is also recommended to increase efforts in mapping of discharge and deposition of micropollutants and to give high priority to effect studies, in particular with regard to critical doses and critical concentrations in exposure media and organisms

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Assessment of the Applicability for Pollution Control Authorities of the Concept "Critical Load" of Long-range Transported Micro-pollutants in Relation to Aquatic and Terrestrial Ecosystems

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Preface

The present report is sponsored by the Directorate for Nature Management (main contact Else Løbersli) and the Norwegian Pollution Control Authority (main contact Tor Johannesen).

The aim of the project has been to assess whether the knowledge about atmospheric deposition (or ocean current transport), dose/response relationships and critical concentrations of micropollutant is sufficient to make the "critical load" approach an expedient tool for environmental management authorities in international endeavours to reduce long-range, transboundary transport of these contaminants.

The project has been the joint efforts by research institutes representing expertise in aquatic and terrestial pollution ecology as well as transport/load. The following institutions and persons have contributed:

Norwegian Institute for Air Research: J.M. Pacyna
Norwegian Institute for Water Research: J. Knutzen, K. Hylland and T. Källqvist
Norwegian University of Science and Technology/ Dept. of Chemistry: E. Steinnes
Norwegian Institute for Nature Research: T. Nygård
Norwegian Polar Institute: N. Skjegstad, O.E. Henriksen and G. Wing Gabrielsen

The main text is basically arranged as summary chapters based on the sub-reports treating separate aspects of the subject, and annexed in the above order of institutions. For references in the main text one should consult the corresponding annex.

Oslo, December 10, 1997.

Jon Knutzen
Project coordinator

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1. Conclusions

The present knowledge of atmospheric deposition rates of the assumed most hazardous micropollutants (various organochlorines and mercury) is insufficient (cf. ch.3 and annex 1), and the possibilities to calculate with the necessary accuracy such figures in the near future is small. There are also many gaps in knowledge with regard to critical exposure/critical concentrations in the probably most sensitive species/communities in various aquatic and terrestrial environments. Consequently, it appears premature to base argumentation in international negotiations concerning further reductions in long-range transport of micropollutants on the critical load approach.

As an alternative it is recommended to apply the precautionary principle One should also increase efforts to quantify the long-range transport portion of the total load, and give high priority to effect studies in sensitive and vulnerable species.

2. Background and general considerations

The critical load concept has been used as basis for international protocols on the reduction in acid rain (sulfur and nitrogen oxides). To avoid confusion with "critical limits" and other ecotoxicological terms, the use of "critical load" should have the general designation quantity per year. The question to be considered here is whether it is possible with existing information to calculate the long range transport portion of the total load judged critical for sensitive parts of aquatic or terrestial ecosystems.

The basis for the protocols on acidifying compounds of sulfur and nitrogen is a solid documentation of damage to sensitive freshwater systems, primarily the decline or eradication of fish populations. Besides, there is a well founded suspicion that acid rain also causes damage to forest vitality. As to effects on fish, relationships have been established between load and water quality parameters such as reduced acid neutralization capacity, lowered pH, and increased concentrations of toxic species of aluminium. Further, the mechanism of aluminium toxicity has been documented and recognized. Over large land areas the load of acidifying substances is only atmospheric, and it has been quantified with adequate accuracy through many years of measurements in air and precipitation. Since the exposure of the organisms is solely from water, the main steps in the dose/response relation are well documented. Under these conditions the critical load concept has been an expedient tool for management authorities in international negotiations.

In contrast to this, premises for the successful use of critical load of transboundary micropollutants are more dubious. There are several reasons for this:

- Apart from the (partially historical) cases of dieldrin/birds, DDE/birds, mercury/birds and PCB/fish eating mammals and birds, damage to populations is less well documented.

- The hazardous substances occur in all compartments of the environment (e.g. water, sediments and food).

- There are several other sources than atmospheric deposition. This is particularly important in relation to metals (natural load via run-off from catchment areas with large variations in mineralogy, besides local anthropogenic surces). In oceanic environments, transport via currents must be taken into account, and in coastal areas also the load from rivers and possible local waste water effluents.

- Observed damage may be the combined result of two or more contaminants.

- Persistent organic pollutants (POP) disturb several aspects of biochemistry/physiology in animals (hormone balance, immunology. nervous system, reproduction, cancer). Consequently, dose/response relations tend to be complex.

- Several routes of exposure and the high probability of additive/synergistic/antagonistic effects contribute further to the complexity of resolving cause/effect relationships and of calculating the critical load for any single substance (or group). In particular this will be the case for birds and mammals representing high or top trophic levels in complex food webs. (Concerning the latter point it should be possible to focus on the worst case food chain).

Some practical differences between acidification and spreading of micropollutants also deserve mentioning:

- First, acid depositions is in a sense due to "natural" combustion processes which are necessary in about the present scale as long as modern civilization has not solved its energy problem. In comparison, it appears economically far more feasible to prohibit/strongly restrict the use or uncontrolled spreading of hazardous metals and persistent organics to the environment.

- Second, whereas discharge of sulfur and nitrogen oxides below a certain amount will not give irreversible effects, one may claim that practically any additional load of some of the persistent organochorines may have unpredictable consequences because they continue to circulate in the biosphere for decades and for a large part tend to end up in environments far from the sources. It also follows from this, that the **time aspect** is more important with regard to xenobiotic substances (including metals like mercury, cadmium and lead), which in itself is a complication when it comes to assessments of critical load.

Ideally, three conditions should be met in order to justify the use of the critical load concept in international endeavours to reduce long-range transport of hazardous metals and POPs:

1. Damage to populations should be documented in regional scale.

2. The effects should be linked to a critical concentration or dose of the relevant substance(s).

3. The ecosystem load should be mainly from long-range transport, i.e. either from the atmosphere or.
   via ocean currents

An alternative to deriving the critical concentrations from observations of deleterious effects in nature is to use a more theoretical basis for assessment of critical load. This approach is normally used to derive maximum acceptable concentrations in air, water, soil or sediments , known as quality criteria, critical concentrations, target values or ecotoxicological assessment criteria in various regulatory schemes. In the framework of risk assessment, the terms Predicted No Effect Concentration (PNEC) or Maximum Acceptable Toxicant Concentration (MATC) are often used. The latter is used to express that it is often not possible to define a no effect concentration, or in cases where certain effects are judged to be acceptable from a management point of view.

In environmental risk assessments, the risk is expressed by the ratio between PEC (predicted Environmental Concentration) and PNEC (or MATC). The PEC is calculated from the known input of the contaminant from various sources and the physical/chemical characteristics of the substance, which affect its fate and distribution in various compartments of the environment. The same factors have to be considered when assessing critical loads. However, the mode of calculation is reversed so that the critical load is derived from a critical concentration/dose.

The key question is whether it is feasible to use PNECs, MATCs or other derived (not observed) "critical" concentrations as a basis for calculation of critical load. These derived concentrations are estimated from information about toxic effects observed in various experimental studies, in practice often acute and chronic toxicity tests with a limited number of standard test organisms. The extrapolation of results from such laboratory studies to effects in the natural environment involves a considerable uncertainty. This is accounted for by using application or safety factors, which may be as low as 1/1000 when PEC is calculated from short-term toxicity tests with only three species (OECD 1992).

If the results of critical load calculations based on PNEC shall have sufficient credibility in making decisions about restrictions on emission of pollutants, it appears imperative that the uncertainty in PNEC calculations has to be considerably improved. In practice this means that more species have to

be tested under conditions relevant to exposure situations found in nature and with relevant test endpoints for sublethal effects of long-term exposure. If not, the above mentioned safety margins may in many cases result in unrealistically low critical loads. In particular this will be the case for naturally occurring substances such as metals and PAH (polycyclic aromatic hydrocarbons).

For synthetic and other compounds unknown in nature it is more logic to apply such very low critical loads. However, within this group there are several for which there is in principle already wide agreement that they should be phased out, or that any further spreading to the environment should be avoided as far as practically possible (e.g. PCB, dioxins, DDT and other persistent organochlorine pesticides). If this in principle agreement results in international protocols on reduction in use and emissions, the efforts to calculate critical loads seem unnecessary.

Another alternative to manifest ecosystem damage as starting point for the calculation of critical loads, are indications of serious disturbances from biochemical/physiological symptoms (biomarkers) in sensitive and vulnerable organisms. Being the most sensitive among effect variables it would be an advantage to use biomarkers. However, apart from the effect of lead on δ-aminolevulinic acid dehydrogenase in the synthesis of heme, and metabolites of PAH, it is a fundamental problem that - at least for the time being - biomarkers are not sufficiently substance specific. Different groups cause the same response, e.g. induction of the cytochrome P450 system by coplanar PCBs and PAH; metallothionein synthesis by exposure either to cadmium, mercury, copper or zinc, and vitellogenin in male fish provoked by various substances with oestrogen mimicing effect. Accordingly, such broad spectered responses only give basis for calculation of critical load for the sum of groups of substances.

An additional problem with biomarkers is the uncertain relation to effects on the population level. One of the difficulties in this connection is to define a normal interval for biomarker levels or activity, as these vary considerably with biotic factors (sex, age), and also with season/temperature.

If critical limits are to be derived, the selection of methods will depend on the relevant target organism(s). The relevant methods for a polar bear or an arctic food-chain will obviously differ from the methods that should be applied to a freshwater crustacean or a lake ecosystem.

Biological effect methods that are applied at the cellular level are generally more sensitive than methods that may be applied at the population level. The choice of methods that will be used to derive critical limits is largely a political decision: at which level of certainty and at what cost do we want to protect the environment ?

In spite of the above reservations with respect to biomarkers as effect variables for calculation of critical loads, it would be of great scientific interest to investigate the possibility to link biomarker responses first to concentrations in the main medium of exposure and then to critical atomospheric/ocean current loads. In a long-term perspective, management authorities certainly will benefit from the resulting improved knowledge about pollutant transport, fate and effects. In particular this will be the case when the effect of phasing out of substances and other restrictive measures is monitored.

3. Sources, transport and deposition

The following is based on the account by J. Pacyna in Annex 1 (cf. references therein).

Accurate and complete information on sources releases is of vital importance for modeling the atmospheric transport of contaminants on local, regional, and global scale, and for assessing the atmospheric deposition of contaminants. Thus, the information on releases of contaminants is a key parameter when discussing the application of the critical load concept for the emission reduction strategies.

Complete parameterization of models to study long-range transport of contaminants and their atmospheric deposition onto aquatic and terrestrial surfaces is the next parameter in this discussion. Finally, verification of model estimates within the critical load concept through measurements at sites included in a properly designed site network is the third parameter which needs attention.

3.1 Sources and releases of heavy metals

During the last two decades there has been a great progress in defining major anthropogenic and natural sources of Cd, Pb, Hg and other heavy metals in Europe and in other parts of the world (e.g. Pacyna, 1996; Nriagu and Pacyna, 1988).

The first quantitative worldwide estimate of annual industrial input of 16 heavy elements into the air, soil, and water has been published by Nriagu and Pacyna (1988). Pyrometallurgical processes in the primary non-ferrous metal industries are the major source of atmospheric As, Cd, Cu, In, Sb, and Zn, and important source of Pb and Se. Combustion of coal in electric power plants and industrial, commercial, and residential burners is the major source of anthropogenic Hg, Mo, and Se and a significant source of As, Cr, Mn, Sb, and Tl. Combustion of oil for the same purpose is the most important source of V and Ni. Combustion of leaded gasoline is still the major source of Pb. Chromium and Mn are derived primarily from the iron and steel industry. Little information is available on emission of heavy metals from various diffuse (fugitive) sources.

A comparison of the worldwide emissions of heavy metals from natural and anthropogenic sources suggests that human activities generate emissions of heavy metals in such quantities that they significantly exceed emissions from natural sources, except for Hg and soil-derived dust components, such as Cr, Mn, and V.

Most of the emphasis has been on assessing the atmospheric emissions. One of the first attempts to estimate atmospheric emissions of heavy metals from anthropogenic sources in Europe was completed at the beginning of the 1980s (Pacyna, 1984). This work presented information on emissions of as many as 16 heavy metals, including all which are of interest in this document. The survey was later updated, completed, emission gridded and applied in long-range transport models to study deposition processes.

The OSPAR program, studying the atmospheric transport and deposition of pollutants to the North Sea, has collected information on atmospheric emissions of heavy metals since the beginning ot the 1980s. Very recently, an improved inventory of the European emissions has been prepared within the program and should be presented to the wider audience quite soon.

In 1989 the UN ECE established a Task Force on Heavy Metals. One of the major objectives of the Task Force was to collect information on emissions. National response to several requests towards creating a data base for emissions of heavy metals in the UN ECE region was considered

unsatisfactory. Therefore, at the February 1993 meeting of the UN ECE Task Force the member countries were asked to provide by the end of 1993 the information on their emissions in the most recent years, preferably for 1990. At the same time, an update of emission data for Pb, Cu, and Cd for 1989/1990 has been made in connection with the Dutch project on a European Soil and Sea Quality and Atmospheric Deposition of Selected Substances (ESQUAD) (Berdowski et al., 1994).

Recently the International Institute for Applied Systems Analysis (IIASA) has prepared a set of current, past, and future emission estimates (Pacyna, 1996).

The above described work within various international programs and organizations has resulted in a number of important conclusions which can be defined as follows. Collection of information on atmospheric emissions of heavy metals from various countries in Europe within various international programs and organizations, including the UN ECE is not satisfactory. Only some countries report data on total and category by category emissions of agreed heavy metals (particularly priority metals: Pb, Cd, Hg).

The most accurate information has been on emissions of Pb, as two thirds of these emissions originate from combustion of leaded gasoline. Information on consumption of gasoline in various countries is quite accurate, and so is information on the use of Pb additivies. A comparison of various independent estimates for Europe as a whole and for individual countries seems to indicate an accuracy of better than 25 %.

Cadmium and anthropogenic Hg have been also a subject of several emission estimates. Their major sources are fairly well defined and emission factors established. An emission estimation accuracy of 50 % or better can be assigned for these two heavy metals, as proved by the comparison of air concentrations calculated by models using emission data and those measured. The same comparison made for other heavy metals seems to indicate uncertainty of a factor of 2 and more. Quite incomplete is the information on Hg re-emissions from aquatic and terrestrial surfaces, as well as from natural sources of this contaminant in Europe.

For a review of the status regarding emission estimates for heavy metals in Europe, and recommendations to fill gaps in knowledge, it is referred to the report from a recent workshop (WMO, 1997).

3.2 Sources and releases of POPs

Major review of information on POP emissions in Europe has been prepared by Berdowski for the EMEP Workshop in Beekbergen in 1994 (Berdowski, 1994). Information on quantities and spatial distribution of emissions of benzo(a)pyrene and lindane, presented within the ESQUAD project is still the most complete regarding the whole Europe. Since the Beekbergen meeting major concern has been placed on the development of methods for POP emission estimates. The following POPs are the most frequently studied:

- hexachlorocyclohexanes (HCHs) and other pesticides,
- selected polychlorinated biphenyls (PCBs),
- hexachlorobenzene (HCB),
- benzo(a)pyrene (B(a)P) and other policyclic aromatic hydrocarbons, and
- selected dibenzodioxins and dibenzofurans.

Concerning the POP emission estimates in individual European countries, only few countries report these data. It should be admitted that the reported information is incomplete with respect to the list of

priority compounds or compound groups and the list of major emission sources. Accuracy of these estimates is largely unknown.

3.3 Conclusion as regards sources and releases in relation to calculation of critical load

Emission data for Pb and Cd may be considered satisfactory for estimates of critical load in Europe. This is not the case, however, for Hg (e.g. poor information on re-emission and emission from natural sources) and particularly not for POPs.

3.4 Assessment of transport models

Pathways of heavy metals and POPs reaching certain receptors are studied with the use of transport models. Following release into the atmosphere, heavy metals can be either deposited in the vicinity of the emission source or subject to long-range transport via air masses. In most cases (except for Hg and to some extent Se), emission of heavy metals occurs on particles. The size of emitted particles containing heavy metals, as well as the temperature and speed of exhaust gases and the height of the emission source, are the major factors influencing the relative proportion of metals deposited locally and transported over long distances.

The atmospheric residence time of POPs attached to particles is typically a few days. These POPs can be modeled in a similar way as the aerosol bound pollutants, such as heavy metals. However, many POPs are semi-volatile under atmospheric conditions and may occur both in the gas phase and as attached to particles depending on the vapor pressure of the compound. Therefore, transport mechanisms within air masses for heavy metals and POPs are different and they will be considered here separately.

Current status and future directions of numerical models in simulating **the transport of metals** have recently been reviewed by Petersen (1997). The review has produced a strong evidence that long-range transport models, particularly those with relatively simple formulations are now in an operational stage. Some of these models have been developed to simulate transboundary transport of acidifying pollutants and were then modified for heavy metals associated with particles. Concerning Hg, recent progress in understanding physico-chemical processes of this contaminant has permitted to investigate Hg transport and deposition in Europe using the EMEP-type of models and comprehensive Eulerian modeling frameworks.

Petersen (1997) concluded, however, that in spite of significant advancement in knowledge of heavy metal properties and in understanding the processes which determine fate of heavy metals in the atmosphere, our knowledge of heavy metal behavior in the air is far from being complete. The accuracy of estimated concentrations and deposition fluxes could be improved by improving the model input data, primarily emission data and meteorological data. As a result of acceptable emission data, model calculations are quite comparablee (10 to 15 %) with observations for lead. For cadmium, models underestimate the concentrations and depositions fluxes with 50 %. For other heavy metals the comparison of model estimates with observations are even worse. However, air and precipitation measurements of metals in Europe are often few and highly uncertain.

In spite of progress in information on speciation and chemical reactions, mercury values predicted by a model which contains this information agree with observations from the Scandinavian network only within a factor of 2 (e.g. Petersen et al., 1995).

Petersen (1997) also concluded that mesoscale modeling capabilities are necessary to quantify the subgrid scale variability of heavy metal concentrations simulated by regional scale models.

Many **organic pollutants** such as the organochlorine pesticides and the PCBs are susceptible to dispersion on a regional and global scale because they are both persistent in the environment and have a volatility that enables them to move between the atmosphere and the Earth's surface in repeated, temperature-driven cycles of deposition and evaporation (Mackay and Wania 1995). This cycling process has important consequences for the global distribution process of POPs and for any attempt in quantifying their fate with the help of models:

- "grass-hopping" into areas far from emissions such as the polar regions
- net transport from low to high latitudes ("global fractionation", Wania & Mackay, 1993)
- POPs are multimedia components with circulation in the environment for decades

Models can serve as a tool to identify, understand, and quantify the key sources, transport mechanisms and sinks of POPs in the environment. Existing global models for POPs cover a wide range of complexity (e.g. review by Wania et al., 1996). Experience shows that the models with the simplest atmospheric description tend to feature the most sophisticated treatment of atmosphere/surface exchange processes. Also, many required input parameters, specifically emission inventories, usually do not presently exist at the spatial resolution required for complex 3-D models.

A review of results on modeling of the atmospheric long-range transport and deposition of POPs in Europe has been prepared by van Pul et al. (1997). The choice of deposition and degradation parameters has a great impact on the calculated deposition. Two examples were given. Van Jaarsveld et al. (1993) concluded that the uncertainty in calculating B(a)P deposition with the TREND model are with a factor of 2 at remote locations for a total deposition on a yearly basis. Jacobs and Van Pul (1996) carried out a sensitivity analysis of the wet deposition calculated with the EROS model. Variations of about a factor of 2 in the physico-chemical properties of γ-HCH and soil conditions resulted in a variation of the wet deposition of 20-40 % above land and 10-100 % above the sea.

Uncertainties in total emissions can be very high. Berdowski et al. (1994) estimated that the emission estimates in Europe can be with accuracy of a factor of 2 to 5 for γ-HCH and 2 to 4 for B(a)P.

The above mentioned uncertainties result in large differences between model estimates and quite sporadic measurements of POPs in Europe, reaching one order of magnitude (e.g. reported by van Pul et al., 1997).

3.5 Conclusion as regards transport models

Taking into account the status of information on the accuracy of emission data , the accuracy of various parameters related to physico-chemical properties of the contaminants, and the accuracy related to the model concept, the model developemnt and its results are satisfactory for estimates of critical load in Europe for Pb and probably for Cd. For Hg, and particularly for POPs, this modeling is not satisfactory.

3.6 Deposition measurements in Europe

There are international programs aiming at the measurements of air concentrations and atmospheric deposition of heavy metals and POPs, including OSPAR, HELCOM, and AMAP. EMEP is also collecting information on deposition measurements of heavy metals and POPs at the EMEP stations (on a voluntary basis) and at other stations within national networks.

Major revision of measurement programs for heavy metals and POPs in the air and atmospheric deposition in Europe has been made at the recent EMEP workshop in Moscow (WMO, 1997). General conclusion was that ongoing measurement activity on heavy metals is large compared to emission estimates and modeling activity. However, the data comparability for heavy metals is low. Lead seems to be measured most frequently compared to other heavy metals, showing the best comparability.

For POPs only a few species are measured at a very limited number of locations.

The final conclusion was that harmonized and standarized measurements of heavy **metals and POPs** are needed for evaluation of emission inventories, validation of models, and trend analysis. Missing is also information on POP concentrations in the top soil and surface waters which are important variables in the re-volatilization of POPs.

3.7 Overall conclusion

For the critical load concept to serve well as a basis for international agreements/protocols there is a number of prerequisites to be met. Due to genrally large gaps in emission data, and for most POPs also a lack of measurements in air and depositions, these conditions are at present only met for lead and cadmium.

4. Aquatic Ecosystems - Communities of gill breathing organisms

The below assessment is an excerpt from Annex 2 (by J. Knutzen, K. Hylland and T. Källqvist).

4.1 General overview

Among the substances mentioned in the preparatory papers to the Bad Harzburg workshop (Annex 2, ch.2.3), it is only for PCB, DDT/DDE, dieldrin (drins), and mercury that damage to populations at a regional/global scale has been documented (or made highly probable). In all (partly historical) cases the affected species have been mammals or birds. As to effects on gill breathing organisms, damage to populations/communities is more speculative.

Observations from the 1980 ies in the North Sea and the Baltic indicated that reduced hatching and survival of fry could be correlated to levels of organochlorines like PCB, DDT/DDE and dieldrin in ovaries of certain fish species (Westernhagen et al.,1981, 1989 and Hansen et al., 1985). It is possible that the levels correlated to such effects also have occurred on the Norwegian Skagerrak coast before the general decrease in the concentrations of these organochlorines due to restrictions on use and handling.

Of the other substances mentioned in preparatory papers to the Bad Harzburg workshop, suspected large scale effects on fish reproduction is restricted to polychlorinated dibenzofurans/dibenzo-p-dioxins (PCDF/PCDDs) and/or coplanar PCBs (with dioxin like effects), as reported for salmonids from Lake Ontario, USA/Canada (e.g. Walker et al, 1994 and Smith et al, 1994, with references), and similar symptoms in Baltic salmons (Norrgren et al., 1993). Recent results (Bengtsson et al., 1996) indicate that the phenomenon primarily is correlated with thiamine deficiency, however not excluding micropollutants as partially responsible.

For fish and aquatic invertebrates it is so far only one reasonably well documented case of large scale deletrious effects to populations: imposex in dog whelks (*Nucella lapillus*) caused by tributyl tin (TBT) as antifoulant in ship paint. Apart from increased levels, effects from other micropollutants is limited to biochemical and physiological stress symtoms like enhanced enzyme activity, weakening of the immune system and vitellogenine in male fish. Damage from metals have merely been observed in the vicinity of point sources.

4.2 TBT and imposex

Imposex is the irreversible occurrence of more or less developed male organs in females of snails within the group Neogastropoda, in the later stages causing sterility and death. The symptoms are described by the to indices VDSI (Vas Deferens Sequence Index) and RPSI (Relative Penis Size Index) - cf. Gibbs et al. (1987), reflecting increasing degrees of damage. Mean VDSI above 4 and mean RPSI above 40 % are correlated with sterility in part of the female population

In Norway the situation is as follows:

- Imposex have been observed along the whole coast except in the northermost part (Berge et al.,1997). At about half of the stations VDSI was above 4, including some open coast areas.

- National surveys of TBT in mussels (*Mytilus edulis*) (Knutzen et al.,1995, Følsvik,1997), indicate that in most coastal surface water TBT occurs, at least periodically, in concentrations exceeding 1 ng/l, which is recommended as criterion to protect marine organisms (Moore et al., 1992).

- In contrast to what has been reported from some other countries, there is no direct evidence of eradicated dog whelk populations. On the other hand, environmental monitoring has been far from thorough enough to exclude the possibility. Besides, less conspicuous species than the dog whelk may be affected without notice.

  (With regard to critical limits for conservation of marine life, it should be added that a working group within OSPAR (Oslo and Paris Commissions) has proposed an "Ecotoxicological Assessment Criterion" (below which no damage to marine organisms are expected) of 1/20 of the above mentioned criterion.

  The main toxicity mechanisms appears to be clear. According to Bettin et al. (1996) TBT blocks the normally occurring enzymatic transformation of male hormones to oestrogen in females, resulting in accumulation of androgens. Further, VSDI and RPSI values can be related to the concentration of TBT in dog whelks. (Gibbs & Bryan, 1996, Berge et al., 1997).

Other organotin compounds may have the same effects as TBT, but are usually considerably less potent. Recently, however, Horiguchi et al. (1996) observed that triphenyltin acted as strongly as TBT versus some species. The probably additive effect form other organotins means that critical load calculations should include these compounds.

TBT is easily degradeable by phytoplankton and is - in general - metabolized relatively rapidly by vertebrates, crustaceans and polychaetes, more slowly by mussels and snails. However, this general picture has been modified by some recent investigations, showing unexpectedly high accumulation in some whale species (Iwata et al.,1995; Kannan et al.,1995,1997). The findings of Kannan et al. (1997) also raised the question of even more sensitive species than the hitherto focused snails due to the immunosuppressive effect of TBT.

In the most affected areas the main part of TBT has local sources. Nevertheless, the nearly global occurrence of imposex (eg. Ellis & Pattisina, 1990) and the association with main shipping lanes (Ten-Hallers Tjabbes et al., 1994) show that there is also a diffuse component from mobile point sources and possibly long range transport via ocean currents.

4.3 Conclusion for the aquatic environment

Respecting the applicability of critical load on the TBT problem, the first two of the three prerequisites listed in ch. 2 (damage in regional scale, established case/effect relationship) may with some reserve be regarded as met. The third point, however, a significant contribution to the damaging load by long-range transport, is undocumented, not very probable, and difficult to estimate. Also taking into account the great awareness of the problem and recent improvement in pollution status (Evans et al., 1995) due to widespread restrictions on TBT use, other means than the critical load approach seems more expedient to obtain international agreement on further measures agains this pollutant.

As regards other micropollutants in aquatic environments, none of the three conditions for a profitable use of the critical load concept can be met for the time being.

5. Terrestrial environment - Soil organisms and plants

The following account is based on the contribution by E. Steinnes (Annex 3).

5.1 Sources of heavy metals in the soils and the specific situation in Norway

Heavy metals in soils may originate from the following source categories:

- Natural mineral material present in the soil, varying over several orders of magnitude.
- Atmospheric supply, which can be a significant source *e.g.* for Hg
- Agriculture or forestry practices such as fertilisation, which is a source for Cd.
- Local sources such as smelters, mine tailings, etc.
- Long-range atmospheric transport.

Here the focus will be on transboundary anthropogenic input via the atmosphere. Only for some metals this source will contribute significantly to the total load/exposure.

Most of the international literature on soil pollution is concerned about arable soils. In Norway the arable land constitutes less than 5% of the total land area, whereas most of the remaining is covered by natural soils only little disturbed by anthropogenic activity.

The situation with respect to influence of an additional heavy metal load on surface soils will be strongly different for agricultural and natural soils. With arable land the added metal will be mixed within the entire plough layer (the upper 15-20 cm). In most Norwegian soils it may be strongly concentrated in the upper few cm, in particular in the case of well developed humus layer. Moreover, the **speciation** of the metal and hence its mobility and bioavailibility is likely to be quite different in an agricultural soil with 2-3% organic matter compared to a natural soil with mainly organic matter in the surface horizon. Any work to adapt the critical load concept to soils in Norway must reflect the particular importance of natural soils and the specific properties of these soils.

The transboundary atmospheric transport of heavy metals to Norway and the deposition in different parts of the country has been followed regularly over the last 20 years (air concentration measurements, precipitation monitoring, moss surveys), and the temporal and spatial trends in heavy metal deposition is fairly well known over this period. For most metals typically supplied by long range transport of pollutants (LRTP) the deposition rates have decreased by more than a factor of 2 during this period.

Extensive work has also been done to characterize the soil pollutant load resulting from LRTP and distinguish it from other sources (Allen and Steinnes 1980, Steinnes et al. 1989, Njåstad et al. 1995, Steinnes et al.1997). Methods to define and separate the LRTP component in natural surface soils have been proposed (Steinnes and Njåstad 1995) based on chemical analyses of samples from the humus layer and the underground mineral soil, respectively. (For representative mean metal concentrations in Norwegian natural soil, see annex 2)

Elements for which LRTP is an essential contributing factor to natural surface soils in Norway include **lead, cadmium, arsenic, antimony** and to a lesser extent zinc, mercury and selenium.

Data concerning agricultural soils are more sparse, with a monitoring program just in its beginning. Most research on metals in Norwegian agricultural soils so far has been focused on cadmium from fertilisers.

From the available evidence, it is suggested that work related to critical load of heavy metals to soils in Norway primarily be focused on lead and cadmium.

5.2 Critical load of heavy metals

The work by De Vries & Bakker (1996a) constitute a useful framework for a discussion of the feasibility of the critical load concept for Norwegian conditions.

The critical load concept refers to the most sensitive component of the environment with respect to a given pollutant. When it comes to heavy metals, it is normally difficult to define what that component is. Effects may be observed with soil microorganisms as well as invertebrates, various vascular plants, and mammals and birds after accumulation of metals from food).

From the available literature on heavy metal toxicity to soil organisms it seems that soil microorganisms such as bacteria is the most sensitive group of organisms in general. In several studies the soil microbial activity was shown to be reduced at metal levels not much above natural concentrations (Tyler 1992). On the other hand, bacteria have short generation times and hence may more readily become adapted to moderate increases in metal burden.

To define critical exposure adequately, it is clearly preferable to have some measure of bioavailable fraction, but in most cases this is difficult to define.Moreover, the most sensitive organism /system for a given soil may be different depending on a variety of factors, such as cultivated/non-cultivated, crop/vegetation type, organic matter content and a number of other physical and chemical properties of the soil.

For practical work there is need for a simple approach in order to develop a tool for critical load calculation, if possible. Based on target values (upper range of natural total levels for metals in "standard soil"). De Vriess & Bakker (1996a) applied a steady-state model to calculate the corresponding critical load from atmospheric deposition. It is not evident, however, that this model would be usable for Norwegian conditions:

- The assumption of homogeneity is far from reality in natural soils.
- The not considered biotic component may be important for metal turnover.
- Critical load will depend on background concentration, which are quite variable.
- The soil ecosystems in question may be rather far from steady-state.
- Quality criteria based on the bioavailable fractions would be more meaningful as exposure parameters.

5.3 Organochlorines and PAH

The knowledge about atmospheric deposition and soil contamination levels of POPs in Norway is much more limited than for heavy metals. Regular monitoring of these substances in air started only

very recently (Tørseth and Manø 1997) at Lista, southernmost Norway, and Ny-Ålesund, Spitzbergen. Data on the concentration levels of POP in soils are rather scarce (see some data for selected organochlorines in annex 3, indicating strong north-south gradient in atmospheric load). The geographical ditribution of PAH leaves little doubt that significant amount of PAH are supplied by long-range transport. Topsoil levels in the south are about tenfold higher than in the north. The levels of fluoranthene in the south are almost ten times higher that the Dutch target value for this substance in soil, whereas for benzo(a)pyrene the level is similar to the target value.

A manual has also been prepared for the calculation of critical loads of POP in soils (De Vries & Bakker 1996b) The methods proposed are similar to those for heavy metals. One important difference is evident: whereas volatilisation from the soil is an insignificant pathway for all relevant metals except Hg, this is a very important process to be considered for many POPs. Moreover, chemical degradation of POP must be considered, but is difficult to estimate, depending among other factors on soil temperature, and microbiological activity. Toxic responses to POP are also poorly known for most soil organisms. Altogether it appears that critical load calculations for POP are likely to be even more difficult and uncertain than for heavy metals.

5.4 Conclusion with regard to soil organisms and vascular plants

For the usefulness of the critical load concept, basic information particularly is lacking with regard to deposition rates, degradation and critical concentrations of POPs, but also as regards critical concentration/bioavailable fraction and turnover of metals.

For the usefulness of the critical load concept, basic information particularly is lacking with regard to deposition rates, degradation and critical concentrations of POPs but also as regards critical concentration/bioavailable fraction and turnover of metals.

6. Terrestrial environment - Birds and mammals

The below summary is based on the contribution by T. Nygård (Annex 4).

6.1 General considerations

The "critical load" concept is based on an estimation of a level of exposure below which sensitive elements are not harmed. The critical load implies an exposure which has a time and area factor. Data on persistent organic pollutants (POPs) from top soils and water masses are scarce, preventing the construction of models that could predict factors of bioaccumulation and biomagnification (Moriarty 1988, see also ch. 3 and 5 above).

To prove effects of pollutants on populations often require long time-series to gather a sufficient amount of data, due to the overriding effects of natural variation in population numbers and reproduction. The effects are often complex and subtle, and a system of 'early warning'-signals may therefore be desirable. In this context, the concept of biomarkers in individuals has been introduced.

There are large difficulties involved in trying to connect levels of persistent organic pollutants (POPs) and heavy metals (HM) found in soil and vegetation to levels in free-living terrestrial animals. For these organisms the levels are determined via food only. Therefore, the individual feeding habits of an animal, such as preferred prey species and their availability, position in the food chain, and local mineral composition of the bedrock may be important. In addition, confounding factors such as the age and the health of the individual and local point sources of pollution may be significant.

Notwithstanding the difficulties of quantifying the atmospheric load and the resulting concentrations in soil/water and in succeeding trophic levels of the food chains in question, it is essential for the reasoning leading to critical loads to know the corresponding critical concentrations in the most sensitive species. Consequently, one has chosen to focus on suggested critical levels in bird top predators found in Norway. (Some data is also given for fish-eating birds and birds exposed to lead from plants contaminated by atmospheric deposition. For more information about seabirds and mammalian top levels of aquatic food chains and other mammal top predators, see ch. 7).

In relation to critical load it appears most relevant to use a mean population level. The response to a pollutant may vary greatly among individuals, according to e.g. condition, age, sex, and genetical characteristics.

6.2 Background information

The main effect parameter that was chosen in this review was eggshell thinning. The thinning is caused by inhibition of the enzymatic transfer of calcium from blood to eggs in the oviduct. P,p'-DDE affects hormone-systems, and inhibits enzymes responsible for the transport of calcium from the oviduct to the egg (Lundholm 1987). A reduction of shell thickness of 15-20% has been shown to be sufficient to cause population declines in many raptor populations (Newton 1979). Crick (1992) proposed a formula based on a large data set; % shell thinning = 3.5 + 12.3 log (DDE), wet weight. This implies that a critical level is reached for the population when the average DDE levels in the eggs exceed 10 ppm wet weight. Many species (e.g. falcons) have shown population decline below this level (from 3 ppm). 3 ppm DDE is therefore chosen as the critical level in raptor eggs.

The very toxic cyclodienes, among them dieldrin, have led to high adult mortality among raptors (Newton 1988, Nisbet 1988). Widely used in Europe, they probably have been responsible for the declines of many migratory Fennoscandian raptor species wintering in southwest Europe, and they were also a threat to sedentary raptors preying on migrating birds. Newton (1988) found 1 ppm dieldrin in the liver, and 0.7 ppm in eggs of raptors to be a critical value for the sustainability of populations. A dieldrin level of 0.1 ppm in eggs has been suggested for the bald eagle in the USA (Wiemeyer et al. 1984). Different sensitivity of even closely related species have been documented in several cases (Fyfe et al., 1976; Westlake et al.,1978). Allowing for this, 0.1 ppm dieldrin has been chosen as a critical average population value for harmful effects of dieldrin.

The effects of PCBs in birds have to a large degree been obscured by being highly correlated with DDE levels. As the concentrations of the latter now are decreasing, it has become clear that PCBs have effects of their own (Bowerman et al., 1995). Present data are not good enoughto indicate threshold values for specific congeners. Studies of reproductive success of the bald eagle (*Haliaeetus leucocephalus)* in USA suggest 4 ppm ww. of sum PCBs in eggs as a critical level (Bowerman et al., 1995).

Critical levels of the chlordane group and mercury may be set, respectively, at 3-5 ppm (Stickel et al., 1979, and Blus et al., 1983) and at 0.6 ppm w.w. ((Newton & Haas 1988).

(The above information is summarised in table 1 of annex 4, together with data regarding some other organochlorines and lead).

6.3 Assessment of the situation in Norway

Since the start in the mid 1960s a substantial amount of data on the most widespread organochlorines (except Toxaphene, cf annex 4) and potentially hazardous metals has been collected. A national programme for monitoring of the terrestrial environment was established in 1990, and samples of birds and mammals are routinely collected for analysis. Most of the data relating to raptorial birds is shown in table 1.

Table 1. Levels of pollutants in eggs of raptors from Norway, 1974-94, given as µg/g wet weight (except TCDD and $TEQ_{PCDD/F}$, ng/g). The material was analysed at the National Veterinary Institute (own material and from Holt et al., 1979 and Frøslie et al., 1986), and at the Norwegian Institute for Air Research (PCDF/D). Chlor. = Chlordane, Lind. = Lindane, and Dield. = Dieldrin. Values exceeding assumed critical levels are in bold.

Species		DDE	PCB	HCB	Chlor.	Lind.	Mirex	Dield.	TCDD ng/g	TEQ PCDD/F ng/g	Hg
White-tailed	Mean	**4.429**	**14.321**	.090	1.051	.042	.073	**293**			.216
Sea Eagle	edian	2.720	8.398	.051	.843	.027	.042	**205**			.189
Haliaeetus albicilla	Min.	.141	1.120	.009	.034	.008	.011	.030			.010
	Max.	**60.000**	240.000	1.197	4.286	.255	.273	**1.120**			**.680**
	N	68	68	65	38	39	12	25			68
Golden Eagle	Mean	.388	1.033	.024		.025		.019			.078
Aquila chrysaetos	edian	.208	.560	.018		.020		.010			.039
	Min.	.041	.070	.006		.006		.008			.010
	Max.	2.150	5.163	.130		.084		.083			.360
	N	45	39	29		20		9			43
Goshawk	Mean	**4.368**	**12.613**	.097		.051		**.241**			.359
Accipiter gentilis	edian	**3.412**	**7.900**	.037		.017		**.237**			.316
	Min.	.695	1.264	.009		.012		8			.024
	Max.	**14.536**	**36.348**	.337		.139		**.661**			**1.106**
	N	19	16	14		6		11			19
Osprey	Mean	**5.836**	2665	.023	.010	.040		.012			.157
Pandion haliateus	edian	1.347	2.722	.015	.	.028		.007			.079
	Min.	.126	.379	.009	.010	.017		.005			.040
	Max.	**54.984**	**6.952**	.057	.010	.104		.040			.553
	N	14	14	12	1	6		10			13
Peregrine	Mean	**7.309**	**18.279**	.229		6.28		**1.966**	0.043*	0.185*	.445
Falco peregrinus	edian	**4.640**	**17.600**	.108		.475		**.460**			.460
	Min.	.139	2.161	.038		.156		.079	0.033*	0.179*	.142
	Max.	**28.323**	**34.617**	1.710		1.406		**12.036**	0.052*	0.190*	**.851**
	N	17	17	15		4		9	2*	2*	17

(tabl. cont.)

(table 1 - cont.)

Species		DDE	PCB	HCB	Chlor.	Lind.	Mirex	Dield.	,TCDD ng/g	TEQ PCDD/F ng/g	Hg
Rough-legged	Mean	.165	.827	.021		.028		.021			.105
Buzzard	edian	.094	.347	.014		.017		.007			.023
Buteo lagopus	Min.	.032	.071	.009		.017		.008			.010
	Max.	.569	**5.056**	.066		.055		.079			.553
	N	10	10	10		8		7			9
Sparrowhawk	Mean	**15.888**	**9.129**	.374		.094		**.396**			.183
Accipiter nisus	edian	**14.083**	**4.685**	.090		.096		**.320**			.158
	Min.	1.662	1.049	.019		.052		**.237**			.050
	Max.	**35.200**	**31.200**	1.560		.132		**.600**			.395
	N	10	10	10		4		6			7
Short-eared Owl	Mean	2.464	1.580	.018				.015			39
Asio flammeus	edian	.	.	.				.			.
	Min.	2.465	1.580	.019				.016			.040
	Max.	2.465	1.580	.019				.016			.040
	N	1	1	1				1			1
Eagle Owl	Mean	1.327	**4.186**	.034		.052		.008			.040
Bubo bubo	edian	.336	1.003	.010		.		.008			.021
	Min.	.024	.220	.006		.052		.008			.010
	Max.	**6.083**	**15.800**	.114		.052		.009			.158
	N	8	8	7		1		4			8
Merlin	Mean	**9.1**	1.4	.11	0.11	.16	0.005	**.170**			.25
Falco columbarius	edian	**6.0**	1.0	.04	0.11	.08	0.006	**.170**			.21
	Min.	1.4	.25	.003	0.003	.02	0.0007	.050			.015
	Max.	**39.9**	6.6	1.1	0.28	1.1	0.01	**.290**			**0.88**
	N	43	43	41	37	39	12	2			41
Gyrfalcon	Mean	1.099	2.504	1.683		.234		.007			.153
Falco rusticolus	edian	1265	2356	187		.057		.			.158
	Min.	.289	.883	.002		.017		.008			.130
	Max.	1.577	**4.424**	.295		.630		.008			.171
	N	4	4	4		3		1			3

* M. Oehme & Rune Bergstrøm, unpublished material.

There is considerable geographical variation in atmospheric deposition of pollutants. The regional variation in the concentration of mercury, PCB and DDE in eggs of white-tailed sea eagle is shown in Figures 1-2 in Annex 4. The DDE, PCBs and other POPs as well as mercury declined from south to north (Nygård & Skaare, in press). The declining trends in POPs from south to north is consistent with the deposition pattern of PCBs in Scandinavia (Södergren 1972), and the general pattern of deposition of pollutants to Norway (Tørseth & Semb 1995). The white-tailed sea eagle is a sedentary bird, whereas the merlin is a migrating species. The geographical variation in POP level in merlin eggs was not significant, but there were significantly higher mercury levels to the north (Nygård 1997).

As use and discharges of pollutants have been banned or severely restricted, declining pollutant levels over time are expected. Nygård (1997) found that ecological half-lives for DDE and PCBs in eggs in white-tailed sea eagle were 17 and 19 years, respectively, while the mercury levels were stable.

The most readily observed effect parameter of chemical pollution is eggshell thickness. As it is directly connected to successful reproduction, it is highly relevant. There is a linear relationship between eggshell thickness and log-p,p' DDE, but other compounds may also be involved, such as PCB (synergistic, potentiating (Lincer 1994) and Hg. The available data on shell thickness in Norwegian bird of prey species is shown in Table 2.

Table 2. Eggshell thickness in Norwegian birds of prey before and after 1947 (mm/1000, based on average clutch values), mainly own material (Nygård, 1990, Nygård, 1997).

Species	Mean thickness Before 1947	n	Mean thickness After 1947	n	Diff.	p
White-tailed Sea Eagle	619	29	600	71	-2.2*%	0.045
Golden Eagle	601.8	7	597.6	24	-0.7%	0.84
Sparrowhawk	270.7	2	227.2	6	-15.8%	-
Goshawk	436.6	32	407.9	14	-6.6%	<0.001
Osprey	514.1	18	444.0	14	-13,6%	<0.001
Peregrine	361.4	18	308.2	22	-14.7%	<0.001
Merlin	241.9	23	212.9	52	-12%	<0.001
Eagle Owl	416.7	7	387.5	7	-7.0%	0.086

* Thickness index (Ratcliffe, 1967) was - 6.0%).

Mustelids, especially mink, have been reported to be very sensitive to POPs, and PCB in particular. The PCB levels in otter have been monitored in Norway in more than two decades. However, there are no evidence of any anomalies in reproduction connected to the prevalence patterns of PCBs, and the mean levels were from four to eight ppm PCBs on a lipid basis. (Christensen & Heggberget 1995). This is slightly below the suggested No Observed Effect Level (NOEL) for this species (AMAP report, 1997).

Herbivores may be exposed to elevated levels of heavy metals through browse contaminated via the atmosphere. Such metals will not normally bioaccumulate through food-chains, as they are deposited in specific organs, such as kidney, liver and bone. The highest concentrations in mammals are usually found in southern Norway. For lead, this is documented in moose *Alces alces* (Frøslie et al. 1984) common shrew *Sorex araneus*, bank vole *Clethrionomys clareolus* and for mountain hare *Lepus timidus* (Kålås & Lierhagen 1992). There were a good correlations between the lead concentration in liver from moose and the levels in moss from the corresponding sites, indicating air pollution as an important pathway of lead to the moose.

The same regional pattern is seen for cadmium. Small rodents from the south have higher cadmium levels than in the north (Kålås & Jordhøy 1995, Kålås et al. 1995a, 1995b).

6.4 Concluding remarks

As seen from table 1 estimated critical levels of POPs have been exceeded in some or several cases in most of the bird species examined (Tab. 2). For DDE, mean population levels above critical have been detected in oshawk, osprey, peregrine, sparrowhawk and merlin. For sum PCBs, the same has been found for white-tailed sea eagle, peregrine, sparrowhawk and great horned owl. Regarding

dieldrin, the U.S. estimate of 0.1 ppm as a critical level has been exceeded on a mean population level for white-tailed sea eagle, goshawk, peregrine, sparrowhawk and merlin. Using the more conservative estimate for the peregrine from Britain of 0.7 ppm, only the peregrine has levels above critical.

For the other POPs, there is too little data on the possible effect on populations. It is worth noticing, however, that the high levels of chlordanes found in white-tailed sea eagle and merlin may have biological significance.

As regards mercury, no species had average values above the suggested 0.6 ppm w.w. but goshawk, peregrine and merlin had values approaching this level.

It follows from the present review that in several species there is a high risk for effects at the population level. Further, critical concentrations are fairly well established for at least some of the substances in question. Even so, the critical load approach does not seem applicable to higher terrestrial animals, as is it is extremely difficult to relate deposition rates to resulting concentrations in their tissues. This is mainly due to the influence of many environmental and ecological variables acting on the net accumulation rates, such as local soil and bedrock conditions, age and condition of the animals, local food variations and individual preferences, and individual variations in metabolism and release of the contaminants. In addition to this comes that for mercury and organochlorines estimates of deposition rates are very uncertain due to lack of measurements (cf. ch. 2).

The lead level in grouse species will largely be determined by atmospheric deposition on plant foliage. So far negative effects on grouse species are merely speculative, but in this case there is a better basis than for organochlorines/mercury in the future to relate critical concentrations to a corresponding critical atmospheric load.

So far the observed effects of long-range transported metals and persistent organochlorines in Norwegian mainland mammals are limited to elevated levels and a south-north declining gradient particularly for lead and cadmium in certain species. There is no evidence for damage at the population level, but little is known about possible long-term effects of the sublethal levels found.

7. Artic biota

In the main the following is a summary of the assessment by N. Skjegstad, E.O. Henriksen and G.W. Gabrielsen (Annex 5).

7.1 Background, scope and limitations

Because high concentrations of PCBs are found in e.g. polar bear and glaucous gull in Arctic, much of the concern about the pollutional state of the Arctic are focused on this group of contaminants. Dead and dying glaucous gulls are found with high concentrations in their body. Hermaphrodites are found among polar bears, and recently, some cubs of the polar bear are found to be smaller than normal (Derocher, pers. comm.). The suggestion is that **PCB are causing all these effects, and consequently is given the highest priority here**. Nevertheless, also other organochlorines are considered.

In principle this assessment was also supposed to deal with critical loads for heavy metals. Although relatively high concentrations of cadmium and mercury are found in seabirds, seals, whales and polar bear, so far no effect studies regarding metals have been performed within the Antarctic Assessment and Monitoring Programme (AMAP). Due to this lack of effect data heavy metals are omitted from the assessment The same applies to PAH, from which the long-range transported load is not expected to affect polar communities.

According to the suggestions for priorities for the Bad Harzburg Workshop, one of the objectives is to identify receptors for calculation of critical limits (concentrations). The present document focuses on levels and concentrations of PCBs in different arctic/Norwegian biota. As far as possible, the observed levels from different studies are compared with threshold values (critical concentrations) for some types of effects which is reported in the literature.

The data on concentrations related to effects are presented with reservations for uncertainties caused by differences in experimental conditions in the quoted studies (e.g. field vs. laboratory observations, variation in biological parameters such as age, sex, condition, reproductive status, tissues analysed). Due to lack of data, possible additive or synergetic effects from the presence of several contaminants cannot be considered.

The main background material assessing critical concentrations in arctic biota has been taken from the AMAP (1997). This report summarizes levels of a number of POPs in many species in the circumpolar Arctic. Some threshold levels for toxic effects (NOEL) in various animals are also given. These levels are used as basis for the assessment of critical concentrations.

The present focuses on the conditions prevailing in the Norwegian Arctic. However, most of the studies referred to in AMAP are Canadian. The number of effect studies being limited has also necessitated extrapolation among species, not only among areas.

7.2 Mammals

Ranges of measured TEQ_{TCDD} concentrations in arctic marine and terrestrial mammals are shown in Table 1. The concentrations in polar bear both from Canada and Svalbard partly exceed the level (209 pg/g lipid weight (lw)) which are associated with immune suppression in captive harbor seals. On the other hand, they are below the EC_{50} level in mink liver for kit survival (9,500 pg/g lw).

Table 3. Ranges of measured TEQ_{TCDD} concentrations (pg/g lw) (from PCDD/Fs, non-ortho and mono-ortho PCBs) in arctic marine mammals (AMAP report, 1997).

Species	Concentration range (pg/g lw)
Polar bear (Canada)	20-230
Polar bear (Svalbard) (1990-1994)	80-260
Beluga	3-25
Narhwal	9-20
Ringed seal	2-30

Levels of PCDD/PCDF in adipose tissue (58-73 % lipid - Ramsay *et al.*, 1992) in Canadian polar bears sampled in 1983-84 ranged from 2-23 pg TEQ/g wet weight (ww). The mean levels of PCDD/PCDF, non-ortho and mono-ortho PCBs in liver from Canadian bears collected in 1992-1994 were 27, 29 and 172 pg TEQ/g lw, respectively (Letcher *et al.*, 1996). The combined TEQs exceed those associated with immunosuppression in harbor seal (209 pg TEQ/g lw). Norheim *et al.* (1992) found PCB levels in polar bears at Svalbard close to those connected with reproductive disorders in Baltic seals (Helle *et al.*, 1976; Bergman & Olsson, 1985; Olsson *et al.*, 1992). Mono-ortho PCB levels were measured in polar bears from Svalbard (Bernhoft *et al.*, 1997) and the levels were in the range of 42-102 pg TEQ/g ww, or 80-260 pg TEQ/g lw (Table 1). The levels are comparable to or higher than the mono-ortho PCB TEQs (140 pg TEQ/g lw) associated with immunosuppressive effects in harbor seal.

Ranges of $\sum$PCB concentrations in different arctic mammals, and threshold values for mammalian effects for $\sum$PCB concentrations (from the AMAP report), are shown in tables 4 and 5, respectively.

Table 4. Ranges of $\sum$PCB concentrations (ng/g lw) measured in arctic mammals (AMAP report, 1997).

Species	Concentration range (ng/g lw)
Polar bear, Svalbard	7,000 - 90,000
Polar bear, circumpolar (inluding Svalbard)	3,000 - 90,000
Arctic fox, Svalbard	2,000 - 240,000
Beluga	2,000 - 8,000
Narhwal	5,000
Ringed seal	500 -5,000
Walrus	100 - 20,000
Harbor porpoise	2,000 - 25,000
Harp seal	500 - 6,000
Mink liver (LW)	2 - 70

Table 5. Threshold values for mammalian effects in respect to ∑PCB concentrations (ng/g lw) (AMAP report, 1997).

Concentration for effect (ng/g lw)	Mammalian effect	Species
90,000-110,000	EC50 kit survival	Mink liver
90,000	Poor reproductive success	Baltic ringed seal
60,000-70,000	EC50 litter size	Mink liver
40,000	Poor reproductive success	Captive harbor seals, blood
35,000	LOEL, immune effects	Rhesus monkey blood
9,500	NOEL, kit survival	Mink muscle
9,000	NOEL, reproduction	Otter muscle
1,000	NOEL, visual memory	Rhesus monkey offspring
800	LOEL, short-term memory	Human offspring cord blood serum

By comparing the tables it appears that the highest concentrations recorded in polar bear and arctic fox by far exceeds effect thresholds for PCB observed in several other species. This applies to several effect end points but the exceedance of the threshold values are particularly noteworthy as regards neurobehavioral effects and reproduction in sensitive species.

Assuming that polar bear and arctic fox are as sensitive to PCB in food as mink, there is a further warning in the registration that mean levels of ∑PCB in ringed seal blubber from 16 sites in the Canadian and Norwegian Arctic, exceed the diet NOEC in mink related to reproduction (72 ng/g ww, Heaton *et al.*, 1995).

Relatively low levels of ∑PCB are found in different cetaceans in Canadian areas. In beluga the levels of ∑PCB exceed the NOEL and LOEL for initial neurobehavioral effects, but are below the NOEL for otter reproduction and mink kit survival (table 5).

From tables 4-5 it is also seen that PCB levels in harbour porpoise from different parts of the Arctic exceed NOEL/LOEL for neurobehavioral effects; the highest recordings also NOEL for reproduction in otter and mink and approaching thresholds for immunosuppressive and reproductive effects in other species.

7.3 Seabirds

PCB levels in eggs of arctic fish eating birds have been recorded in the range 100-2700 ng/g w.w. (AMAP report, 1997). This is low or moderate compared to effect levels in other bird species (lowest LOEL of 3400 ng/g, AMAP report, 1997) except in white-leghorn chicken. Effect studies with arctic seabirds are lacking and is particularly needed for the glaucus gull, which is a top predator in the Arctic.

7.4 Norwegian effect studies

Studies in the polar bear by Skåre et al. (1994) revealed a significant negative correlation betwen retinol and ∑PCB, and also indicated that thyroid hormones could be used as biomarker for PCB. Letcher et al. (1994a, 1996) demonstrated that the cytochrome P450 system of the polar bear was induced by PCB, and that CYP1A1/2 levels in liver microsomes were correlated to the sum TEQ of coplanar congeners.

In a number of glaucous gulls found dead at Svalbard in 1989 it was recorded mean PCB levels in liver, brain and kidney of $16/15/10x10^3$ ng/g w.w., respectively. As seen from table 5 in annex 5 this is up to several times LOEL and NOEL values for reproduction found in a selection of other species.

7.5 Concluding remarks

With the present status of knowledge it appears beyond reach in the forseeable future to establish critical loads for definite substances or substance groups, which nonetheless represent a very probable threat to the most sensitive arctic species and communities. Besides the unsolved problem of quantifying the loading rate via the atmosphere (cf. ch.. 3) and ocean currents with sufficient accuracy, the reason for this is the complexity of factors determining the relation between load and accumulation/effects. Examples of this is primarily the time aspect of accumulation in the Arctic ecosystem as a whole, but also the inadequate knowledge of interactions among different pollutants with regard to metabolism, bioaccumulation/biomagnification and effects.

The key question is whether critical concentrations for deleterious population effects in polar bear, arctic fox, and possibly sensitive species among sea mammals and seabirds already is exceeded or how close to critical concentrations the present contaminant levels are. Consequently, it seems to be most useful for environmental management authorities to increase efforts to establish critical concentrations for all relevant species, and with a higher degree of certainty than at present.

Annex 1.

An assessment of the applicability of the critical load concept as a basis for international protocols on emission reductions for heavy metals and persistent organic compounds: Sources, transport and deposition.

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An assessment of the applicability of the critical load concept as a basis for international protocols on emisssion reductions for heavy metals and persistent organic compounds:

Sources, transport, and deposition

by
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1. Introduction

Accurate and complete information on sources and sources releases is of vital importance for modeling the atmospheric and riverine transport of contaminants at all distances (on local, regional, and global scale), and for assessing the atmospheric deposition of contaminants. Thus, the information on sources and releases of contaminats from these sources is a key parameter when discussing the application of the critical load concept for the emission reduction strategies.

Complete parameterization of models to study long-range transport of contaminants and their atmospheric deposition onto aquatic and terrestrial surfaces is the next parameter in this discussion. Finally, verification of model estimates within the critical load concept through measurements at sites included in a properly designed site network is the third parameter which needs attention.

The aim of this chapter is to describe the above defined parameters of critical load concept in the context of international protocols on reductions of emissions of heavy metals and persistent organic pollutants (POPs), planned in the UN ECE region. Major concern is placed on priority metals and POPs defined in the scope of the whole document. Information on heavy metals and POPs is discussed separately due to differences in sources and behavior of these two groups of pollutants in the environment.

2. Sources and source releases

There are both anthropogenic and natural sources of heavy metals entering the environment, while most of the POP sources are of anthropogenic origin.

2.1. Sources and source releases of heavy metals.

During the last two decades there has been a great progress in defining major anthropogenic and natural sources of Cd, Pb, Hg and other heavy metals in Europe and in other parts of the world (e.g. Pacyna, 1996; Nriagu and Pacyna, 1988). It was concluded that high temperature processes, such as coal and oil combustion in electric power stations and heat and industrial plants, gasoline combustion, roasting and smelting of ores in non-ferrous metal smelters, melting operations in ferrous foundries, refuse incineration, and kiln operations in cement plants generate various heavy metals, which enter the atmosphere and the aquatic and terrestrial ecosystems. Practically every industry discharges one heavy metal or the other into these ecosystems.

The first quantitative worldwide estimate of annual industrial input of 16 heavy elements into the air, soil, and water has been published by Nriagu and Pacyna (1988). Pyrometallurgical processes in the

primary non-ferrous metal industries are the major source of atmospheric As, Cd, Cu, In, Sb, and Zn, and important source of Pb and Se. Combustion of coal in electric power plants and industrial, commercial, and residential burners is the major source of anthropogenic Hg, Mo, and Se and a significant source of As, Cr, Mn, Sb, and Tl. Combustion of oil for the same purpose is the most important source of V and Ni. Combustion of leaded gasoline is still the major source of Pb. Chromium and Mn are derived primarily from the iron and steel industry. Little information is available on emission of heavy metals from various diffuse (fugitive) sources.

Major sources of anthropogenic heavy metal contamination of aquatic ecosystems, including the ocean are domestic wastewater effluents (especially As, Cr, Cu, Mn, and Ni), coal-burning power plants (As, Hg, and Se in particular), non-ferrous metal smelters (Cd, Ni, Pb, and Se), iron and steel plants (Cr, Mo, Sb, and Zn) and the dumping of sewage sludge (As, Mn, and Pb). The atmosphere is the major route of Pb entry into natural waters and also accounts for over 40 % of the V loading. A comparison of global emission estimates indicates that for most of the heavy metals, the annual anthropogenic inputs into the water exceed the quantities emitted to the atmosphere.

The estimates by Nriagu and Pacyna (1988) suggest that soils are receiving large quantities of heavy metals from disposal of a variety of industrial wastes. The two principal sources of heavy metals in soils worldwide are, however, the disposal of ash residues from coal combustion and the general wastage of commercial products on land. Urban refuse represents an important source of Cu, Hg, Pb, and Zn with notable contributions of Cd, Pb, and V also coming via the atmosphere. The large volumes of wastes associated with animal husbandry, logging, as well as agricultural and food production can affect the heavy metal budget of many soils significantly. Although municipal sewage sludge may not be a particularly important source on a global scale, it can be one of the most important sources of metal contamination of soils on a local scale.

A global assessment of natural sources of atmospheric heavy metals has been made by Nriagu (1989). Biogenic sources can account, on average, for over 50 % of Se, Hg, and Mo, and from 30 to 50 % of the As, Cd, Cu, Mn, Pb, and Zn, released annually to the atmosphere from natural sources. Volcanic emanations can account for 40 to 50 % of the Cd and Hg and 20 to 40 % of the As, Cr, Cu, Ni, Pb, and Sb emitted annually from natural sources. Seasalt aerosols seem to account for <10 % of atmospheric heavy metals from natural sources. Finally, soil-derived dusts can account for over 50 % of the total Cr, Mn, and V emissions, as well as for 20 to 30 % of the Cu, Mo, Ni, Pb, Sb, and Zn released annually to the atmosphere. As the accuracy of emission estimates for natural sources is low, the above mentioned percentage contributions should be considered with caution.

A comparison of the worldwide emissions of heavy metals from natural and anthropogenic sources suggests that human activities generate emissions of heavy metals in such quantities that they significantly exceed emissions from natural sources, except for Hg and soil-derived dust components, such as Cr, Mn, and V.

The natural sources of heavy metals which influence freshwater and terrestrial environment are even more difficult to assess than the atmospheric sources. No information is available to the author on the amounts of these releases of heavy metals.

Information on global emission of heavy metals is important in Europe because a part of these contaminants deposited in some European regions originate outside Europe. However, the key emission information when discussing the deposition of heavy metals in Europe is on sources and releases of these pollutants within Europe.

Most of the emphasis has been on assessing the atmospheric emissions. One of the first attempts to estimate atmospheric emissions of heavy metals from anthropogenic sources in Europe was completed at the beginning of the 1980s (Pacyna, 1984). This work presented information on

emissions of as many as 16 heavy metals, including all which are of interest in this document. Previous works, mostly within the EEC programs have dealt with either a single metal (e.g. cadmium or lead) or a given source category (e.g. combustion of fossil fuels).

The above mentioned European-wide emission survey was later updated, completed, emission gridded and applied in long-range transport models to study deposition processes.

The OSPAR program, studying the atmospheric transport and deposition of pollutants to the North Sea has collected information on atmospheric emissions of heavy metals since the beginning ot the 1980s. Very recently, an improved inventory of the European emissions has been prepared within the program and should be presented to the wider audience quite soon.

The OECD has also been involved in a review of information on emissions of heavy metals, particularly in the member countries. Very little of new information has been obtained through this activity because the basic information on emission factors for major source categories was very similar to that used in the approaches by Pacyna (1984) and Axenfeld et al. (1992).

In 1989 the UN ECE established a Task Force on Heavy Metals. One of the major objectives of the Task Force was to collect information on emissions. National response to several requests towards creating a data base for emissions of heavy metals in the UN ECE region was considered unsatisfactory. Therefore, at the February 1993 meeting of the UN ECE Task Force the member countries were asked to provide by the end of 1993 the information on their emissions in the most recent years, preferably for 1990. At the same time, an update of emission data for Pb, Cu, and Cd for 1989/1990 has been made in connection with the Dutch project on a European Soil and Sea Quality and Atmospheric Deposition of Selected Substances (ESQUAD) (Berdowski et al., 1994).

Recently the International Institute for Applied Systems Analysis (IIASA) has prepared a set of current, past, and future emission estimates (Pacyna, 1996).

The above described work within various international programs and organizations has resulted in a number of important conclusions which can be defined as follows. Collection of information on atmospheric emissions of heavy metals from various countries in Europe within various international programs and organizations, including the UN ECE is not satisfactory. Only some countries report data on total and category by category emissions of agreed heavy metals (particularly priority metals: Pb, Cd, Hg).

It is very difficult to assess the accuracy of currently available emission data for heavy metals in Europe mostly due to limited information on emissions, emission factors, and specific statistical data in various countries.

The most accurate information has been on emissions of Pb, as two thirds of these emissions originate from combustion of leaded gasoline. Information on consumption of gasoline in various countries is quite accurate, and so is information on the use of Pb additivies. A comparison of various independent estimates for Europe as a whole and for individual countries seems to indicate an accuracy of better than 25 %.

Cadmium and anthropogenic Hg have been also a subject of several emission estimates. Their major sources are fairly well defined and emission factors established. An emission estimation accuracy of 50 % or better can be assigned for these two heavy metals, as proved by the comparison of air concentrations calculated by models using emission data and those measured. The same comparison made for other heavy metals seems to indicate uncertainty of a factor of 2 and more. Quite incomplete is the information on Hg re-emissions from aquatic and terrestrial surfaces, as well as from natural sources of this contaminant in Europe.

Compared with atmospheric emissions, much less information is available on emission inventories reporting discharges of heavy metals to the aquatic and terrestrial environment. In one of very few approaches in Europe, discharges of Cu, Pb, Ni, and Zn from major point sources to the aquatic environment were estimated for the Commission of the European Communities (EC) (Daamen et al., 1990). IIASA has been aaproaching to assess releases of Cd, Cu and Zn. Accuracy of these estimates is lower than the accuracy of estimates of atmospheric emissions.

The status of the emission estimates for heavy metals in Europe was reviewed at the 1996 EMEP emission workshop in Moscow (WMO, 1997). It was concluded that more focus should be placed on obtaining the information on:

- emissions of heavy metals on different size particles. Two classes of particles should be considered at the beginning: below and above 2 μm in diameter,
- spatial distribution of emissions within the EMEP grid system, prepared by national emission experts and reported to the EMEP data base,
- location of major point sources with additional information on the height of the source,
- seasonal changes of emissions differentiating between winter (increased heat production) and summer seasons,
- re-emission of Hg from terrestrial and aqatic environments,
- emissions from natural sources, particularly Hg emissions,
- chemical and physical forms of metals emitted from various sources, including emissions of organo-metallic forms,
- past changes of emissions of at least Cd, Pb, and Hg, and
- future trends of emissions of at least Cd, Pb, and Hg.

The above information was found missing but very important in order to estimate transport and deposition of heavy metals in Europe.

2.2. Sources and source releases of POPs

Major review of information on POP emissions in Europe has been prepared by Berdowski for the EMEP Workshop in Beekbergen in 1994 (Berdowski, 1994). Information on quantities and spatial distribution of emissions of benzo(a)pyrene and lindane, presented within the ESQUAD project is still the most complete regarding the whole Europe. Since the Beekbergen meeting major concern has been placed on the development of methods for POP emission estimates. The following POPs are the most frequently studied:

- hexachlorocyclohexanes (HCHs) and other pesticides,
- selected polychlorinated biphenyls (PCBs),
- hexachlorobenzene (HCB),
- benzo(a)pyrene (B(a)P) and other policyclic aromatic hydrocarbons, and
- selected dibenzodioxins and dibenzofurans.

Concerning the POP emission estimates in individual European countries, only few countries report these data. It should be admitted that the reported information is incomplete with respect to the list of priority compounds or compound groups and the list of major emission sources. Accuracy of these estimates is largely unknown.

2.3. Do we know enough about sources and source releases of heavy metals and POPs in Europe ?

The above question is addressed here mainly with a view of the application of emission data in models arriving at the current deposition load and then critical load. Taking into account the status of information on accuracy and completeness of emission data available in Europe one may conclude that the emission data for Pb and Cd are satisfactory for estimates of critical load in Europe. This is not the case for Hg (e.g. poor information on re-emission and emission from natural sources) and particularly for POPs.

3. Transport models

Pathways of heavy metals and POPs reaching certain receptors are studied with the use of transport models. Following release into the atmosphere, heavy metals can be either deposited in the vicinity of the emission source or subject to long-range transport via air masses. In most cases (except for Hg and to some extent Se), emission of heavy metals occurs on particles. The size of emitted particles containing heavy metals, as well as the temperature and speed of exhaust gases and the height of the emission source, are the major factors influencing the relative proportion of metals deposited locally and transported over long distances.

The atmospheric residence time of POPs attached to particles is typically a few days. These POPs can be modeled in a similar way as the aerosol bound pollutants, such as heavy metals. However,many POPs are semi-volatile under atmospheric conditions and may occur both in the gas phase and as attached to particles depending on the vapor pressure of the compound. Therefore, transport mechanisms within air masses for heavy metals and POPs are different and they will be considered here separately.

3.1. Atmospheric transport of heavy metals.

During the recent years it has been recognized that the numerical models can be an important tool in formulating effective control strategies for the reduction of atmospheric deposition fluxes of heavy metals to the terrestrial and aquatic environments. Various models have been developed to study the transport of particle-attached metals as well as of mercury and its species. Current status and future directions of numerical models in simulating this transport have been reviewed by Petersen (1997) recently. The development and application of dispersion models for heavy metals has progressed from formulations for inert species to models accounting for the physico-chemical transformations of atmospheric mercury species. The review by Petersen (1997) has produced a strong evidence that long-range transport models, particularly those with relatively simple formulations are now in an operational stage. Some of these models have been developed to simulate transboundary transport of acidifying pollutants and were then modified for heavy metals associated with particles. Concerning Hg, recent progress in understanding physico-chemical processes of this contaminant has permitted to investigate Hg transport and deposition in Europe using the EMEP-type of models and comprehensive Eulerian modeling frameworks.

Petersen (1997) concluded, however, that in spite of significant advancement in knowledge of heavy metal properties and in understanding the processes which determine fate of heavy metals in the atmosphere, our knowledge of heavy metal behavior in the air is far from being complete. This applies also to the simulation of transport mechanisms. This fact is due to incomplete data needed for proper parameterization of this simulation. The accuracy of estimated concentrations and deposition fluxes could be improved by improving the model input data, primarily emission data and meteorological data. It has already been indicated here that information on sources and source

releases of Pb and Cd is acceptable with respect to the accuracy and completeness of these data. This is not the case for other heavy metals. As a result, model calculations are quite comparable (10 to 15 %) with observations for Pb. For Cd, model calculations underestimate the concentrations and deposition fluxes within 50 %. For other heavy metals the comparison of model estimates with observations is even worse than for Cd. However, it should be admitted that air and precipitation measurements of heavy metals in Europe are few and highly uncertain in some cases.

In the case of Hg, a progress has been made in obtaining the information on speciation and mechanisms and kinetics of chemical reactions. These are important parameters for models simulating transport and removal processes of Hg from the atmosphere. Even so, values predicted by a model which contains this information agree with observations from the Scandinavian network only within a factor of 2 (e.g. Petersen et al., 1995). For other regions in Europe this comparison is expected to be worse. Such poor agreement between model estimates and observations for Hg in Europe limits the confidence with which the results of different control options can be predicted.

Petersen (1997) also concluded that mesoscale modeling capabilities are necessary to quantify the subgrid scale variability of heavy metal concentrations simulated by regional scale models. Mesoscale models should be interfaced with large scale Eulerian models to obtain a "nested" modeling system capable to simulate long-range transport induced local effects with a spatial resolution of 20 km or less. The developemnet of such practices in Europe is only in a starting phase now.

3.2. Transport of POPs.

Many organic pollutants such as the organochlorine pesticides and the PCBs are susceptible to dispersion on a regional and global scale because they are both persistent in the environment and have a volatility that enables them to move between the atmosphere and the Earth's surface in repeated, temperature-driven cycles of deposition and evaporation (Mackay and Wania 1995). They are for this reason sometimes refereed to as "semi-volatile". This property sets them apart from irreversibly deposited contaminants such as many heavy metals or acid rain components. This cycling process has important consequences for the global distribution process of POPs and for any attempt in quantifying their fate with the help of models:

1) Facilitated by seasonal temperature fluctuations POPs can be transported in a stepwise fashion ("grass-hopping") into areas far from any emissions such as the polar regions. While irreversibly deposited species rely on specific weather situations that favour long range transport in order to reach remote areas, the transport of POPs over long distances may occur in countless steps, with extended periods of retention and with winding detours.
2) Since warm temperatures favour partitioning from the Earth's surface to the atmosphere, but cold temperatures enhance deposition from the atmosphere, it has been hypothesised that the repeated cycling of POPs will result in a net transport from low to high latitudes referred to as "global fractionation" (e.g. Wania and Mackay, 1993).
3) The time frame of the long range transport of irreversibly deposited species is determined by the time that elapses between emission and deposition, i.e. a period of several days to a few weeks. POPs, on the other hand, can essentially be subject to regional and global transport as long as they persist in the environment, which in many cases may be several decades. While a sulphur dioxide molecule emitted one year ago will have little effect on present atmospheric conditions, a DDT molecule applied in the 1950s may still be cycling in the global atmosphere today.
4) POPs are truly multi-media contaminants, which can occur in the atmosphere, fresh and sea water, sediments, soils and vegetation. Approaches that focus on one medium are unlikely to yield a comprehensive and accurate picture of their global behaviour.

Models can serve as a tool to identify, understand, and quantify the key sources, transport mechanisms and sinks of POPs in the environment. They can play a role of synthesis or integration in which they bring together all the component studies of usage or source estimation, physical chemical properties, transport in media such as the atmosphere and oceans and between media such as wet and dry deposition or evaporation, and the extensive monitoring efforts to provide a coherent, and consistent picture of contaminant fate which can form a foundation for justifying suggested controls. They may also assist in the formulation and testing of hypotheses that there is a "cold condensation" or "global fractionation" effect by which certain substances display a tendency to concentrate in regions of cold climate (Wania and Mackay, 1993).

Existing global models for POPs cover a wide range of complexity (e.g. review by Wania et al., 1996). The global atmosphere has been treated in one, two, and three dimensions. Higher resolution of the atmosphere, however, does not necessarily increase the validity of the model results. The complexity devoted to describe atmospheric transport in the 3-D models can occur at the expense of the treatment of other important processes. Experience shows that the models with the simplest atmospheric description tend to feature the most sophisticated treatment of atmosphere/surface exchange processes. Also, many required input parameters, specifically emission inventories, usually do not presently exist at the spatial resolution required for complex 3-D models.

A review of results on modeling of the atmospheric long-range transport and deposition of POPs in Europe has been prepared by van Pul et al. (1997). It was pointed out that large uncertainties can be involved in POP modeling in Europe due to large uncertainties introduced by the model concept, some specific parameters and emission data. The uncertainties introduced by a model concept are expected to be relatively small reaching 30 %.

The choice of deposition and degradation parameters has a great impact on the calculated deposition. Two examples were given. Van Jaarsveld et al. (1993) concluded that the uncertainty in calculating B(a)P deposition with the TREND model are with a factor of 2 at remote locations for a total deposition on a yearly basis. Jacobs and Van Pul (1996) carried out a sensitivity analysis of the wet deposition calculated with the EROS model. Variations of about a factor of 2 in the physico-chemical properties of γ-HCH and soil conditions resulted in a variation of the wet deposition of 20-40 % above land and 10-100 % above the sea.

Uncertainties in total emissions can be very high. Berdowski et al. (1994) estimated that the emission estimates in Europe can be with accuracy of a factor of 2 to 5 for γ-HCH and 2 to 4 for B(a)P.

The above mentioned uncertainties result in large differences between model estimates and quite sporadic measurements of POPs in Europe, reaching one order of magnitude (e.g. reported by van Pul et al., 1997).

3.3. Are we good enough to model transport of heavy metals and POPs in Europe ?

The above question is addressed here mainly with a view of the application of models to simulate deposition load and then critical load of heavy metals and POPs in Europe. Taking into account the status of information on the accuracy of emission data available in Europe, the accuracy of various specific parameters related to physico-chemical properties of the discussed contaminants, and the accuracy related to the model concept one may conclude that the model developement and its results are satisfactory for estimates of critical load in Europe for Pb and probably for Cd. For Hg, and particularly for POPs, this modeling is not satisfactory.

4. Deposition measurements in Europe

There are international programs in Europe aiming at the measurements of air concentrations and atmospheric deposition of heavy metals and POPs, including OSPAR, HELCOM, and AMAP. EMEP is also collecting information on deposition measurements of heavy metals and POPs at the EMEP stations (on a voluntary basis) and at other stations within national networks in various European countries.

Major revision of measurement programs for heavy metals and POPs in the air and atmospheric deposition in Europe has been made at the recent EMEP workshop in Moscow (WMO, 1997). General conclusion was that ongoing measurement activity on heavy metals is large compared to emission estimates and modeling activity. However, the data comparability for heavy metals is low. Lead seems to be measured most frequently compared to other heavy metals, showing the best comparability.

For POPs only a few species are measured at a very limited number of locations. The final conclusion was that harmonized and standarized measurements of heavy metals and POPs are needed for evaluation of emission inventories, validation of models, and trend analysis. Missing is also information on POP concentrations in the top soil and surface waters which are important variables in the re-volatilization of POPs.

5. Final remarks

Theoretically, the critical load concept can serve well as a basis for international agreements/ protocols on emission reduction of environmental contaminants. However, there is a number of prerequisites to be met in such case. This chapter has reviewed some of these prerequisites related to sources of heavy metals and POPs, their releases, transport, and atmospheric removal processes.

Among several heavy metals and POPs considered for international agreements on emission reductions, Pb and Cd seem to meet the prerequisites connected with the application of critical load concept as a basis for these agreements. Emission inventories of these contaminants from the major sources in Europe are quite complete and accurate. Transport models are developed and are in operational phase for a some time already. These models are quite similar as both metals are emitted mostly on particles.This fact makes the modeling exercises much easier. Also chemical forms of Pb and Cd are quite straightforward resulting in a simple chemical reaction scheme for these metals in the atmosphere. Lead and Cd have the longest and largest record of deposition measurements in Europe which helps very much verification of the model estimates of atmospheric loadings now, as well as in the past.

Mercury has been much less evaluated with respect to its emissions, transport, and removal processes compared to Pb and Cd. There are various reasons for this delay in Hg evaluation. Hg exists in a larger number of physical and chemical forms than Pb and Cd. Bigger is also a list of emission sources. Models desribing Hg transport and behavior in the air are much more complexed compared to relatively simple Pb and Cd models based on modeling of particle transport within air masses. Finally, the development of sampling and analytical methods for Hg is a current issue while these methods for Pb and Cd have existed for decades. This fact results in relatively few measurements of Hg in the air and deposition, performed in a proper way. Taking the above into account one may conclude that it is premature to apply a critical load concept as a basis for international agreements on reduction of Hg emissions in Europe.

Situation for almost all POPs considered within international programs is even worse than for Hg. POP emissions are poorly evaluated, the development of transport models is in a starting phase, and so is the development of a European environmental data base which can be used to validate the model estimates. Two compounds: γ-HCH and B(a)P seem to be in a better position than the others, but still their source assessment, transport modeling and deposition evaluation are far from being satisfactory. Current research programs, particularly projects within the EU hold a promise of improvement for selected POPs. At present, however, no justifiable data exist to support the application of the critical load concept as a basis for international agreements on reduction of POP emissions in Europe.

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Annex 2.

On the applicability for pollution control authorities of the concept "critical load" of long-range transported micropollutants in relation to aquatic ecosystems.

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1. Background and general considerations

The critical load concept has been used as basis for international protocols on the reduction in acid rain (sulfur and nitrogen oxides). To avoid confusion with "critical limits" and other ecotoxicological terms the use of "critical load" should have the general designation "quantity per year". The question to be considered here is whether it is possible with existing, or in the short term available, information to calculate the long range transport portion of the total load on aquatic ecosystems judged to be critical for the most sensitive part of the freshwater localities of defined land areas or larger parts of the marine environment.

The basis for the protocols on acidifying compounds of sulfur and nitrogen is a solid documentation of damage to sensitive freshwater systems, primarily the decline or eradication of fish populations. Besides, there is a well founded suspicion that acid rain also causes damage to forest vitality. As to effects on fish, relationships have been established between load and water quality parameters such as reduced acid neutralization capacity, lowered pH, and increased concentrations of toxic species of aluminium. Further, the mechanism of aluminium toxicity is documented and recognized. Over large land areas the load of acidifying substances is only atmospheric, and it has been quantified with adequate precision through decades-long anlysis of air and precipitation. Since the exposure of the sensitive organisms is solely from water, the main steps in the dose/response relation are well documented, and the critical load concept has been an expedient tool for mangement authorities in international negotiations.

In contrast to this, premises for the successful use of critical load of transboundary micropollutants are more dubious. There are several reasons for this:

- Apart from the (partially historical) cases of dieldrin/birds, DDE/birds, mercury/birds and PCB/fish eating mammals and birds, damage to populations is less well documented.
- The hazardous substances occur in all compartments of the environment (e.g. water, sediments and food).
- There are several other sources than atmospheric deposition. In particular this is important in relation to metals (natural load via run-off from catchment areas with large variations in mineralogy, besides local anthropogenic sources). In oceanic environments, transport via currents must be taken into account, and in coastal areas also the load from rivers and possible local discharges of waste water.
- Observed damage may be the combined result of two or more contaminants.
- Persistent organic pollutants (POP) disturb several aspects of biochemistry/physiology in animals (hormone balance, immunology. nervous system, reproduction, cancer). Consequently, dose/response relations tend to be complex.
- Several routes of exposure and the high probability of additive/synergistic/antagonistic effects contribute further to the complexity of resolving cause/effect relationships and of calculating the critical load for any single substance (or group). In particular this will be the case for birds and mammals representing high or top trophic levels in somewhat variable food webs. (Concerning the latter point it should be possible , however, to focus on the worst case food chain).

Some practical differences between acidification and spreading of micropollutants also deserve mentioning:

- First, the cause of acid depositions is in a sense "natural" combustion processes which are necessary in about the present scale as long as modern civilization has not solved its energy problem. In comparison, it appears economically far more feasible to prohibit/strongly restrict the use of uncontrolled spreading of hazardous metals and persistent organics to the environment.
- Second, whereas discharge of sulfur and nitrogen oxides below a certain amount will not give irreversible effects, one may claim that practically any additional load of some of the persistent organochlorines may have inpredictable consequences because they continue to circulate in the biosphere for decades and for a large part tend to end up in environments far from the sources. Top predators in general and particularly birds and mammals in polar climate appear to be most vulnerable, perhaps also representatives of high trophic levels associated with another plausible sink for POP: deep ocean habitats, about which there is a profound lack of knowledge respecting ocurrence and possible effects of persistent organics. It also follows from this, that the **time aspect** is more important with regard to xenobiotic substances (including metals such as mercury, cadmium and lead) than other contaminants, which in itself is a complication when it comes to assessments of critical load.

Ideally, three conditions should be met in order to justify the use of the critical load concept in international endeavours to reduce long range transport of hazardous metals and POP:

1. Damage to populations/communities should be documented in regional scale.
2. The effects should be linked to a critical concentration or dose of the relevant substance(s)
3. The load should be mainly from long-range transport, i.e. either from the atmosphere or via ocean currents.

An alternative to deriving the critical concentrations from observations of deleterious effects in nature is to use a more theoretical basis for assessment of critical load. This approach is normally used to derive maximum acceptable concentrations in air, water, soil or sediments , known as quality criteria, critical concentrations, target values or ecotoxicological assessment criteria in various regulatory schemes. In the framework of risk assessment, the terms Predicted No Effect Concentration (PNEC) or Maximum Acceptable Toxicant Concentration (MATC) are often used. The latter is used to express that it is often not possible to define a no-effect concentration, or in cases where certain effects are judged to be acceptable from a management point of view.

In environmental risk assessments, the risk is expressed by the ratio between PEC (predicted Environmental Concentration) and PNEC (or MATC). The PEC is calculated from the known input of the contaminant from various sources and the physical/chemical characteristics of the substance, which affect its fate and distribution in various compartments of the environment. The same factors have to be considered in the assessment of critical loads. However, the mode of calculation is reversed so that the critical load is derived from a critical concentration/dose.

The key question is whether it is feasible to use PNECs, MATCs or other derived (not observed) "critical" concentrations as a basis for calculation of critical load. These derived concentrations are estimated from information of toxic effects observed in various experimental studies, in practice often acute and chronic toxicity tests with a limited number of standard test organisms. The extrapolation of results from such laboratory studies to effects in the natural environment involves a considerable uncertainty. This is accounted for by using application or safety factors, which may be as low as 1/1000 when PEC is calculated from short-term toxicity tests with only three species (OECD 1992).

If the results of critical load calculations based on PNEC shall have sufficient credibility in making decisions about restrictions on emission of pollutants, it appears imperative that the uncertainty in PNEC calculations has to be considerably improved. In practice this means that more species have to be tested under conditions relevant for the exposure situation in nature and with relevant test endpoints for sublethal effects of long-term exposure. If not, the above mentioned safety margins may in many cases result in unrealistically low critical loads. In particular this will be the case for naturally occurring substances such as metals and PAH (polycyclic aromatic hydrocarbons).

For synthetic and other compounds unknown in nature it is more logic to apply such very low critical loads. However, within this group there are several for which there is in principle already wide agreement that they should be phased out, or any further spreading to the environment should be avoided as far as practically possible (e.g. PCB, dioxins, DDT and other persistent organochlorine pesticides). If this in principle agreement results in international protocols on reduction in use and emissions, the efforts to calculate critical loads seem unnecessary.

Another alternative to manifest ecosystem damage as starting point for the calculation of critical loads, are indications of serious disturbances from biochemical/physiological symtoms (biomarkers) in sensitive and vulnerable organisms. Being the most sensitive among effect variables it would be an advantage to use biomarkers. However, apart from the effect of lead on δ-aminolevulinic acid dehydrogenase in the synthesis of heme, and metabolites of PAH, it is a fundamental problem that - at least for the time being - biomarkers are not sufficiently substance specific. Different groups cause the same response, e.g. induction of the cytochrome P450-system by coplanar PCBs and PAH; metallothionein synthesis by exposure either to cadmium, mercury, copper or zinc, and vitellogenin in male fish provoked by various substances with oestrogen mimicing effect. Accordingly, such broad spectered responses will only provide a basis for calculation of critical load for the sum of groups of substances.

An additional problem with biomarkers is the uncertain relation to effects on the population level. One of the difficulties in this connection is to define a normal interval for biomarker levels or activity, as these vary considerably with biotic factors (sex, age), and also with season/temperature. Even with clearly abnormal biomarker levels the link to population relevant variables such as frequency of illness, reduced reproduction, change in behaviour, etc. has not been documented for fish and invertebrates (more convincingly so for some fish eating birds/mammals and top predators). In the few cases with a chain of indices from biomarkers to increased cancer frequency or reduced fertility in fish, the symptoms are limited to local recipients (e.g. Krahn et al.,1986; Malins et al.,1987; Myers et al.,1990; Stein et al,1990,1993; Casillas et al.,1991; Theodorakis et al.,1992 and Johnson et al., 1993).

For gill-breathing organisms there are no cases of clear relations between biomarkers and population relevant parameters that have been associated with long range transport of pollutants.

If critical limits are to be derived, the selection of methods will depend on the relevant target organism(s). The relevant methods for a polar bear or an arctic food-chain will obviously differ from the methods that should be applied to a freshwater crustacean or a lake ecosystem.

Biological effect methods that are applied at the cellular level are generally more sensitive than methods that may be applied at the population level. The choice of methods that will be used to derive critical limits is largely a political decision: at which level of certainty and at what cost do we want to protect the environment ?

In spite of the above reservations with respect to biomarkers as effect variables for calculation of critical loads, it would be of great scientific interest to investigate the possibility to link biomarker responses first to concentrations in the main medium of exposure and then to critical

atmospheric/ocean current loads. In a long-term perspective, management authorities certainly will benefit from the resulting improved knowledge about pollutant transport and fate. This will particularly be the case when the effect of phasing out of substances and other restrictive measures is monitored. It is difficult to see, however, that one will be able in the forseeable future to to calculate critical load of micropollutants with an exactness even approaching what has been achieved for acidifying substances. Consequently, if the aim is to establish international protocols against the spreading of micropollutants, biomarkers in fish and aquatic invertebrates can only serve as supporting evidence in strategies representing alternatives to the critical load concept (e.g. the precautionary principle).

2. Fish, aquatic invertebrates and plants

2.1 General

For fish, aquatic invertebrates and water plants in Norway it is so far only one reasonably well documented case of damage to populations on a regional scale : imposex in dog whelks (*Nucella lapillus*) caused by tributyl tin as antifouling agent in ship paint. Apart from increased levels, effects from other micropollutants is limited to biochemical and physiological stress symtoms such as enhanced enzyme activity, weakening of the immunesystem and vitellogenin in male fish. Damage from metals have merely been observed in the vicinity of point sources.

Observations in the Baltic and the North Sea in the 1980 ies of decreased fertility in fish (reduced hatching and survival of offspring) has been correlated statistically to then commonly occurring concentrations of various persistent organochlorines in ovaries. The "critical" levels of respectively PCB and DDE/DDT mentioned for flounder (*Platichthys flesus*) and herring (*Clupea harengus*) by Westernhagen et al. (1981) and Hansen et al. (1985) are 120/20 µg/kg wet wet weight. In later observations in whiting (*Merlangius merlangus*) Westernhagen et al. (1989) assessed critical values to 200, 20 and 10 µg/kg w.w., respectively for PCB, Σ DDT and dieldrin. Possibly, concentrations approaching these may also have occurred on parts of the Norwegian Skagerrak coast before the general decrease in levels as a result of restrictions on use and discharge of PCB and organochlorine pesticides.

Other examples of suspected large scale damage to fish reproduction caused by persistent organochlorines (with the strongest focus on dioxins and/or non-ortho PCBs) is the blue sac syndrome in salmonids from lake Ontario, USA/Canada (Walker et al.,1991,1994;Spitsbergen et al.,1991; Mac & Schwartz, 1992, and Smith et al.,1994) and similar symptoms in Baltic Salmon (e.g. Norrgren et al., 1993). The most recent conclusion about the phenomenon is that it appears to be related to thiamine deficiency (Bengtsson et al., 1996). However, the causal complex remains to be fully resolved, and a contributing role of persistent organohalogens cannot be excluded. A rather wide interval of 55-400 ng TE/kg w.w. inn eggs of various salmonids has been indicated as threshold level(s) for reduced hatching/yolk sac fry survival (Walker et al.,1994 ; Smith et al,1994 and references therein). (TE = Toxicity equivalents = equvalents of 2,3,7,8- tetrachlorodibenzo-p-dioxin (TCDD), taken as a sum from dioxins and non-ortho and other coplanar PCBs).

Generally, there is a lack of exact knowledge as regards critical concentrations of micropollutants in organs/tissue of vulnerable aquatic organisms. To protect fish eating animals, however, Newell et al. (1987) suggested the following limits for fish muscle (µg/kg): aldrin (22), chlordane (370), ΣDDT (possibly DDT, DDE and DDD separetly, 200), dieldrin (100), endrin (25), hexachlorobenzene (200), Hexachlorocyclohexanes (100), heptachloroepoxide (200), octachlorostyrene (20), ΣPCB (110), TCDD (0.0023). With exception for the proposed TCDD limit, these are rather high levels for fish fillet, even in high fat species, and only occur in the neighbourhood of point sources.

As the only example of (at least indications of) regional damage in Norway, the case of TBT is described in more detail below. For other micropollutants the contribution to possible negative effects on aquatic organisms via ocean currents has hardly been taken into considerations. Instead of repeating the difficulties/impossibility of applying critical load in such cases, a general assessment of the hitherto observed effects on gill-breathing organisms and water plants of the substances mentioned in preparatory documents for the Bad Harzburg workshop is given.

2.2 TBT and imposex in dog whelks.

Imposex is the irreversible occurrence of more or less developed male organs in females of snails within the group Neogastropoda. The symptoms are described by the to indices VDSI (Vas Deferens Sequence Index) and RPSI (Relative Penis Size Index) - cf. Gibbs et al. (1987). In individual females VDSI has seven stages designated by the numerals 0-6 and corresponding to increasingly developed male organs, from the first signs of a vas deferens (1) to the stages 5/6 characterized respectively by overgrowth of the genital papilla and aborted egg capsules. VDSI is given as population mean of individual values, and at population values above 4 an increasing part of the females are sterile (stage 5/6). RPSI expresses the mean population ratio (mean female penis length)^3/(mean male penis length)^3 x 100 %. Registrations in populations has shown that some of the females will be sterile when RPSI > 40%..

As to TBT/imposex in Norway the situation is as follows:

- Apart from four localities in the northernmost part of the country (Finnmark) varying degrees of imposex have been recorded 1993-1995 at the other 28 observeded sites (Berge et al.,1997). At about half of the stations VDSI was above 4 (part of the female population sterile), including some open coast areas.
- From national surveys of TBT in mussels (*Mytilus edulis*) (Knutzen et al.,1995, Følsvik,1997), and assumed bioconcentration factors (ratio of TBT in mussels (wet weight basis): TBT in water of 10^4 - $5x10^4$, cf. references in Knutzen et al., 1995) it is probable that in most of Norwegian coastal surface water TBT occurs, at least periodically, in concentrations exceeding 1 ng/l, which is recommended as criterion to protect marine organisms in Canada (Moore et al., 1992).
- There is no direct evidence of eradicated dog whelk populations in Norway, in contrast to what has been recorded in some other countries, notably in the vicinity of marinas and pleasure craft harbors in Great Britain (Gibbs & Bryan,1987). On the other hand, environmental monitoring has been far from thorough enough to exclude the possibility. Besides, less conspicuous species than the dog whelk may be affected without notice.

(With regard to critical limits for conservation of marine life, it should be added that a working group within OSPAR (Oslo and Paris Commissions) has proposed an "Ecotoxicological Assessment Criterion" (below which no damage to marine organisms are expected) of 1/20 of the above mentioned Canadian criterion. The basis for this very low limit was results from growth tests with larvae of two oyster species and two application factors (each 1/10) on LOEC (Lowest Effect Concentration) from the tests.)

VSDI and RPSI values can be related to the concentration of TBT in dog whelks. VDSI >4 and RPSI > 40 % (and thus part of the female population probably sterile) corresponds to >100-200 µg TBT/kg dry weight (Gibbs & Bryan, 1996, Berge et al., 1997).

The main toxicity mechanisms appears to be clear. According to Bettin et al. (1996) TBT blocks the normally occurring enzymatic transformation of male hormones to oestrogen in females, resulting in accumulation of androgens. Other organotin compounds may have the same effects as TBT, but are usually considerably less potent. Recently, however, Horiguchi et al. (1996) observed that triphenyltin acted as strongly as TBT versus some species. The probably additive

effect of of other organotins means that critical load calculations should include these compounds. For the time being it is difficult to assess what this means in practice (depending on the quality of information about the use of these compounds and the relative size of their sources)

TBT is easily degradeable by phytoplankton and is -in general - metabolized relatively rapidly by vertebrates, crustaceans and polychaets, more slowly by mussels and snails. However, this general picture has been modified by some recent investigations, showing unexpectedly high accumulation in some whale species (Iwata et al.,1995; Kannan et al.,1995,1997). Further, these studies showed higher body burden in whales compared with fish from the same areas, indicating less efficient metabolism in whales. The whale material was stranded dead animals, which caused the speculation that the immunosuppressive effect of TBT might be a causal factor behind mass deaths of dolphins (Kannan et al.,1997). This again rises the question of even more sensitive organisms than the hitherto focused snails and mussels.

The observations of Stäb et al. (1996) indicated that triphenyltin (which in small amounts is added to some antifouling paints, but more commonly used in fungicides) is less degradeable than TBT and consequently with a higher risk for accumulation from prey to their predators. In accordance with this these authors recorded high levels of triphenyltin in cormorants, however not in other birds.

Whereas the halflife of TBT in seawater with photosynthesing organisms is merely a few days, and probably even less in most species of fish, the degradation in sediments is considerably slower. Here the halflife may be several months (Lee, 1996), and in dark and cold environments years.

The main source of TBT is ship traffic, pleasure boating and associated activities (shipyards, sandblasting, marinas, etc.). Significant amounts may also be discharged with municipal waste water (Fent, 1996).

From this follows that in the most affected areas the main part of TBT has local sources. Nevertheless, the nearly global occurrence of imposex (eg. Ellis & Pattisina, 1990) and the association with main shipping lanes (Ten-Hallers Tjabbes et al., 1994) show that there is also a diffuse component from mobile point sources and possibly long range transport via ocean currents. Atmospheric transport is less probable (apparently no studies conducted).

Looking at the three premises (ch.1) for a profitable use of the critical load concept, the two first conditions (documented damage in regional scale and knowledge of the critical concentration of the causal substance) may be regarded as fulfilled for practical purposes in the case of TBT. In contrast , the third prerequisite (solely or mainly loading via long range transport) is not met. In a rather special sense diffuse loading may be claimed to play a role in regional/global contamination but its contribution to damage of snail populations or other suspected effects is difficult to resolve and most probably insignificant. To quantify this international component of contamination appears impractical as it will require calculations with large uncertainties: leakage from submerged hull area of the world's ship traffic and transport via ocean currents, thereby taking into account varying speed of these currents and varying rates of leakage, degradation of TBT and its removal from surface water by sedimentation. In addition come the complicating factors of additive effects from other organotin compounds. It should also be mentioned that restrictions in many countries from the late 1980 ies on the use of TBT on boats shorter than 20-25 m have resulted in many cases of reduced imposex frequency and generally improved conditions ('Evans et al., 1995)

In conclusion it may be stated that it is possible that the crical load concept in theory can be used

for the purpose of attaining international agreements about further restrictions on TBT use but that the awareness of the problem, and resulting improvements so far, and the technical difficulties in calculating the diffuse part of the load all speak for other approaches.

2.3 Effects in aquatic environments of substances mentioned in preparatory papers to the Bad Harzburg workshop.

In these introductory documents the following substances or substance groups are listed (cf. note by T. Johannesen 19970520):
POPs, priority 1: HCB, PAH (benzo(a)pyrene, benzo(ghi)perylene, benzo(b,k)fluoroanthene,, fluoroanthene, indeno/1,23-cd)pyrene) and PCDF/PCDD (dioxins); priority 2: PCB, pentachlorophenol and lindane; priority 3: endosulfan, fenthione, chlordane, nitrofene,quitozene, Toxaphene, trichlorethylene and xylene; priority 4: aldrin, chlorodecone, DDT, dieldrin, endrin, heptachlor and Mirex; priority 5: others.
Metals, priority 1: cadmium,lead, mercury; priority 2: arsenic, chromium, copper, nickel, selen ad zinc.

Only for DDT(DDE), dieldrin, PCB, PCDF/PCDD and mercury there are documented or suspected regional/global damage on population level, and all such cases (except the somewhat questionable effects on salmonids in the Baltic and Lake Ontario) concern lung breathing animals.

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Annex 3.

Applicability of the critical load term on heavy metals and persistent organic pollutants in soil.

Eiliv Steinnes.

Applicability of the critical load term on heavy metals and persistent organic pollutants in soils.

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1. HEAVY METALS

1.1. Sources of heavy metals in soils.

Heavy metals in soils may originate from the following source categories:

A. Natural mineral material present in the soil. The naturally derived heavy metal concentrations in soils may vary over several orders of magnitude, depending on the minerals present in the soil itself and its immediate surroundings.

B. Atmospheric supply due to natural cycling processes, which can be a significant source *e.g.* for Hg from crustal degassing and Se from biogenic processes in the marine environment.

C Agriculture or forestry practices such as fertilisation. This has been shown to constitute a significant source *e.g.* for Cd to agricultural land from phosphate fertilisers.

D. Local pollution sources such as metal smelters, mine tailings, etc.

E. Long range atmospheric transport from domestic and foreign source regions.

In the present work the focus will be on anthropogenic input from the atmosphere, with emphasis on transboundary pollution.

1.2. The specific situation in Norway.

Most of the international literature on soil pollution is concerned about arable soils, *i.e.* agricultural land, gardens, parks, etc. In Norway the arable land constitutes less than 5% of the total land area, whereas most of the remaining land (forests, heathland, mires, impediment) is coveredby natural soils only little disturbed by anthropogenic activity. This is in sharp contrast to the situation in some other countries such as the Netherlands where most of the soils are on arable land.

The situation with respect to influence of an additional heavy metal exposure to surface soils will be strongly different for agricultural and natural soils. In the former case the added metal will be mixed within the entire plough layer (the upper 15-20 cm). In the latter case it may be strongly concentrated within the upper few cm, in particular in the case of a well developed humus layer. Moreover the speciation of the metal and hence its mobility and bioavailibility is likely to be quite strongly different in an agricultural soil with 2-3% organic matter compared to a natural soil with mainly organic matter in the surface horizon. Any work to adapt the critical load concept to soils in Norway must reflect the particular importance of natural soils in this country and the specific properties of these soils.

1.3. Long range atmospheric transport as a source of heavy metals in Norwegian soils.

The transboundary atmospheric transport of heavy metals to Norway and the subsequent deposition in different parts of the country has been followed regularly over the last 20 years (air concentration measurements, precipitation monitoring, moss surveys), and the temporal and spatial trends in heavy metal deposition is fairly well known over this period. For most metals typically supplied by long range transport of pollutants (LRTP) the deposition rates have decreased by more than a factor of 2 during this period.The followimg bulk deposition values (mg m^{-2} yr^{-1}) were calculated for 1990 based on precipitation sampling at Birkenes, southern Norway: V, 0.36; Zn, 4.6; As, 0.23; Mo, 0.048, Cd; 0.061; Sb, 0.062; Tl, 0.011; Pb, 1.9; Bi, 0.010 (Berg et al. 1994).

Extensive work has also been done in the same period to characterize the soil pollutant load resulting from LRTP and distinguish it from the fraction due to other sources (Allen and Steinnes 1980, Steinnes et al. 1989, Njåstad et al. 1995, Steinnes et al.1997). Methods how to define and separate the LRTP component in natural surface soils have been proposed (Steinnes and Njåstad 1995) based on chemical analyses of samples from the humus layer and the underground mineral soil, respectively. Representative mean values for some heavy metals in natural surface soils in different parts of Norway (Steinnes et al. 1997; Hg values from Steinnes and Andersson 1991)) are given in Table 1.

Table 1. Mean values for some heavy metals in natural surface soils in different parts of Norway (ppm).

	Cu	Zn	As	Se	Cd	Sb	Hg	Pb
Southernmost Norway, 0-60 km from the coast	13.6	84.1	5.87	2.02	1.38	2.40	0.18	111.3
Middle Norway, inland	9.5	39.6	1.18	0.33	0.20	0.28	0.17	14.6
Northernmost Norway, inland	8.7	46.7	1.10	0.23	0.18	0.22	0.15	8.5
Environmental quality objectives, Netherlands	36	140	-	-	0.8	-	0.3	85

Elements for which LRTP is an essential contributing factor to natural surface soils in Norway include **lead, cadmium, arsenic, antimony** and to a lesser extent **zinc, mercury** and **selenium.** Additional elements for which the atmospheric deposition data indicate LRTP as a potentially important contributor to surface soils are **vanadium, silver, tin, tellurium, thallium,** and **bismuth.** Data for the geographical distribution of these elements in Norwegian surface soils will be available

in the near future from a collaboration project between the author's laboratory and Norwegian Institute for Air Research. For metals such as **beryllium, chromium, manganese, nickel** and **copper** the contributions from LRTP to natural surface soils are probably insignificant.

As far as agricultural soils are concerned, similar data do not exist so far. The research organization JORDFORSK is now starting a monitoring program for heavy metals in agricultural soils that may improve the knowledge considerably with respect to LRTP contributions. Most research on metals in Norwegian agricultural soils so far has been focused on cadmium from fertilisers.

From the available evidence, it is suggested that work related to critical load of heavy metals to soils in Norway be concentrated on **lead** and **cadmium** in the first case.

1.4. Calculation of critical loads.

A manual for calculating critical loads of heavy metals for soils has been published by two Dutch workers (De Vries and Bakker 1996a). Their report constitutes a useful framework for a critical discussion of the feasibility of the critical load consept for Norwegian conditions.

The critical load concept refers to the most sensitive component of the environment with respect to a given pollutant. When it comes to heavy metals, it is normally difficult to define what that component is. Effects may be observed with:

I. Soil microorganisms (reduced microbial biomass and/or species diversity, affecting microbially mediated processes such as soil respiration).

II. Soil fauna, especially some invertebrates (decrease in abundance, diversity and biomass).

III. Vascular plants including trees (various phytotoxic effects).

IV. Terrestrial fauna and humans (accumulation followed by possible effects to specific organs).

From the available literature on heavy metal toxicity to soil organisms it seems that soil microorganisms such as bacteria is the most sensitive group of organisms in general. In several studies (cf. Tyler 1992) the soil microbial activity was shown to be reduced at levels of given heavy metals not far exceeding those observed in natural topsoils in southern Norway. Combined effects of two or more metals present simultaneously have been studied only to a very limited extent, and possible additive or even synergistic effects are conceivable. On the other hand organisms such as bacteria have very short generation times and may hence more readily than longer-lived organisms become genetically adapted to moderate increases in heavy metal burden. Therefore it is not sure that the most sensitive component with respect to a heavy metal is always found among the microorganisms.

Ideally, it is not sufficient only to know which system or organism is the most sensitive with respect to a given metal. It is also necessary to define some parameter describing as far as possible the actual exposure. In most cases the total concentration of a metal in the soil does not necessarily reflect very well the exposure. Some measure of bioavailable fraction is clearly preferable, but is in most cases difficult to define, even with respect to a given organism.

Moreover, the most sensitive organism /system for a given soil may be different depending on a variety of factors, such as cultivated/non-cultivated, crop/vegetation type, organic matter content and a number of other physical and chemical properties of the soil.

For practial work, therefore, there is need for a more simple approach in order to develop a tool for critical load calculation, if at all applicable. In the Dutch manual (De Vries and Bakker 1996a) so-called target values (upper range of natural levels) for total metal concentrations in a «standard soil» were selected as quality criteria. These values were adjusted for differences in organic matter and clay content from that of the «standard soil» according to a simple formula, reflecting the estimated influence of these factors on the bioavailability. A steady-state model was then applied to calculate the critical load, *i.e.* the highest atmospheric deposition rate that would lead to an equilibrium level of a given metal in the soil not exceeding the calculated value. The mass balance equation takes into consideration a number of processes in the soil/plant system, such as mineralisation of organic matter, weathering, foliar uptake, surface runoff, root uptake, leaching, etc. In order to estimate the extent of these processes it is necessary to describe the equilibrium partitioning of the metal between the soil solution and the solid phase.

It is far from evident that this method would be usable for Norwegian conditions. A few rather evident problems are listed in the following:

- The model assumes a homogeneous soil phase. This situation is approached in a cultivated soil, but is far from reality in a natural situation.

- The model does not take into account the soil biotic component, which may be very important for the turnover of metals in natural soils.

- The calculated critical load will depend on the background concentration of metal in the soil (as with any model dealing with total concentrations).

- The soil ecosystem in question may be rather far from a steady-state situation.

It seems quite obvious that a quality criterion based on a property more closely related to bioavailability, such as the concentration in soil solution or some appropriate extractable fraction, would be more meaningful as an effect parameter than the total metal concentration in the soil. However, calculations of critical load based on soil solution concentration of metal would probably be associated with additional uncertainty.

2. PERSISTENT ORGANIC POLLUTANTS

The groups of persistent organic pollutants (POP) that have raised most concern internationally are the polynuclear aromatic hydrocarbons (PAH) and some of the persistent chlorinated organic compounds (often named orgasanochlorines, OC), such as the DDT-group, PCB (polychlorinated biphenyls), HCB (hexachlorobenzene) and HCH (isomers of hexachloro- cyclohexane including lindane, gamma-HCH). The knowledge about atmospheric deposition and soil contamonation levels of POP in Norway is much more limited than for heavy metals. Regular monitoring of these substances in air started only very recently (Tørseth and Manø 1997) at Lista, southernmost Norway and Ny-Ålesund, Svalbard. Data on the concentration levels of POP in soils are also rather limited in Norway, but some data exists as indicated below.

A manual has also been prepared for the calculation of critical loads of POP in soils (De Vries amd Bakker 1996b) The methods proposed are similar to those for heavy metals. One important difference is evident: whereas volatilisation from the soil is an insignificant pathway for all relevant metals except Hg, this is a very important process to be considered for many POP. In particular some of the lighter organochlorines are fairly volatile, and may be selectively concentrated by atmospheric transport in cold places such as the Arctic (Wania and Mackay 1993). Moreover chemical degradation of POP is an additional factor that must be considered, but is difficult to estimate because it depends

on soil temperature, microbiological activity and other factors. Altogether it appears that critical load calculations for POP are likely to be even more difficult and uncertain than for heavy metals. Toxic responses to POP are also poorly known for most soil organisms, and the selection of a most sensitive organism/system is not likely to be any easier than for heavy metals.

2.1. PAH

PAH in the atmosphere originate mainly from incomplete high-temperature oxidation of organic matter, and are therefore naturally present in the air and soil e.g. from forest fires. Yet industrial activities have strongly enhanced the atmospheric emissions of PAH, and the geographical distribution in natural soils in Norway (Aamot et al. 1996) leaves little doubt that PAH are also supplied by LRTP. Topsoil levels in the south are about tenfold higher than in the north. The levels of fluoranthene in the south are almost ten times higher that the Dutch target value for this substance in soil, whereas for benzo(a)pyrene the corresponding level is similar to the target value.

2.2. Organochlorines

Data for OC in natural surface soils (Lead et al. 1997; unpublished data) include values for selected PCB congeners, DDT/DDE, lindane, and HCB. Examples of levels are given in Table 2.

Table 2. Concentration levels of selected OC in organic surface soils (ng kg^{-1})

	PCB-52	PCB-153	PCB-180	HCB	Lindane
Southern Norway	195	2840	2040	8080	4860
Northern Norway	390	350	370	3700	3640
Svalbard	160	29	5	1440	220
Dutch target values	-	4000	-	-	50

It is likely that most of the OC found in topsoils in Norway are of LRTP origin, but lack of temporal air concentration data makes it difficult to estimate the contribution more exactly. These substances are also more likely to be transported in vapour phase than heavy metals and PAH, in particular the lighter members of the group. From Table 2 it is evident that there is a very strong north - south gradient for the heavier compounds, whereas the lighter ones show rather high levels even in the high Arctic.

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Annex 4.

A contribution to an assessment of the applicability of the critical load concept for persistent organic compounds and heavy metals, based on data from birds and mammals in mainland Norway.

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A contribution to an assessment of the applicability of the critical load concept for persistent organic compounds and heavy metals, based on data from birds and mammals in mainland Norway

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Introduction

The 'critical load' concept is based on an estimation of a level of exposure below which sensitive elements are not harmed according to present knowledge. The critical load implies an exposure, which has a time and area factor. Data on persistent organic pollutants (POPs) from top soils and water masses are scarce. This prevents the construction of models that could predict factors of bioaccumulation (combined intake from food and water) and biomagnification (increase in concentration of a pollutant in animal tissue in successive members of a food chain) (Moriarty 1988).

To prove effects of pollutants on populations often require long time-series to gather a sufficient amount of data, due to the overriding effects of natural variation in population numbers and reproduction. The effects are often complex and subtle, and a system of 'early warning'-signals may therefore be desirable. In this context, the concept of biomarkers has been introduced. These are manifest at the individual level, and include key enzyme systems (cytochrome P450), vitamin A, thyroid hormones and, porphyrins, metallothioneins, etc..

There are large difficulties involved in trying to connect levels of persistent organic pollutants (POPs) and heavy metals (HM) found in soil and vegetation to levels in free-living terrestrial animals. Here, unlike in aquatic environments, the levels are determined via food only. Therefore, the individual feeding habits of an animal, local conditions such as preferred prey species and their availability, position in the food chain, local mineral composition of the bedrock may be important. In addition, confounding factors such as the age and the health of the individual and local point sources of pollution may be significant. I therefore chose to focus on the concept of 'critical limits' for birds and mammals found in Norway.

Some information is available in the literature, although a vast majority of the information available is dealing with levels of pollutants rather than effects. The effect on individuals and organs is most often documented in laboratory studies, while studies concerning effects on the population level are fewer. Nevertheless, for some compounds critical population levels have been proposed. I will here make use of these in the evaluation of whether critical levels have been reached for some of the in some species.

Approach and methods

It is probably more meaningful to use a mean population level as a critical value in this specific context. The response of an animal to a pollutant may vary greatly between individuals, according to e.g. condition, age, sex and genetic background.

As a background, a review of relevant data on the occurrence and concentrations of POPs and HM (mainly mercury) in selected species was carried out. In this connection, it was necessary to find out whether there were sufficient data to stipulate critical pollutant levels, i.e. levels in organs or eggs, which may represent a risk to populations.

Birds.

The birds of prey, Accipitridae, is a group of bird that has been given much attention, both on the national and the international scale both due to their susceptibility and to their position at the top of the food chain. This allows them to accumulate high levels of certain pollutants, especially the lipophilic persistent organochlorines. I have also tried to find references to studies on well-documented dose-response relationships for the various pollutants in question.

The main effect parameter that was chosen in this review was eggshell thinning. The thinning is caused by inhibition of the enzymatic transfer of calcium from blood to eggs in the oviduct. P,p'-DDE affects hormone-systems, and inhibits enzymes responsible for the transport of calcium from the oviduct to the egg (Lundholm 1987). A reduction of shell thickness of 15-20% has been shown to be sufficient to cause population declines in many raptor populations (Newton 1979). Crick (1992) proposed a formula based on a large data set; % shell thinning = 3.5 + 12.3 log (DDE), wet weight. This implies that a critical level is reached for the population when the average DDE levels in the eggs exceed 10 ppm wet weight. Many species (e.g. falcons) have shown population decline well below this level (from 3 ppm). I have therefore chosen 3 ppm DDE as a critical level (no observed adverse effect level, NOAEL) in raptor eggs.

Not only DDE, but also a suite of other potent and persistent chemicals have acted on reproductive success and survival of birds of prey. The very toxic cyclodienes, among them dieldrin, have led to high adult mortality among raptors (Newton 1988, Nisbet 1988). Widely used in Europe, they probably have been responsible for the declines of many migratory Fennoscandian raptor species wintering in southwest Europe, and they were also a threat to sedentary raptors preying on migrating birds. Newton (1988) found 1 ppm dieldrin in the liver, and 0.7 ppm in eggs of raptors to be a critical value for the sustainability of populations. A dieldrin level of 0.1 ppm in eggs has been suggested for the Bald Eagle in the USA (Wiemeyer et al. 1984). Different sensitivity of even closely related species have been documented in several cases (Fyfe et al., 1976). Westlake et al. 1978, Allowing for this, I have chosen 0.1 as an average population value for a 'critical effect' of dieldrin.

The PCBs (polychlorinated biphenyls) are a group of very persistent organochlorine chemicals primarily used in industry. They were first discovered as a pollutant in a White-tailed Sea Eagle *Haliaeetus albicilla* from the Baltic (Jensen 1966). For many years their effects have been obscure, as they were highly correlated with DDE in organs of wildlife. Now, as DDE levels are decreasing more rapidly than PCBs, it is becoming clear that PCBs have environmental effects of their own (Bowerman et al. 1995). Present data are not sufficient to give threshold values for each individual PCB congener. There is often a rather good correlation between the different congeners in different biota, but there is also evidence of differential accumulation and metabolisation between congeners. I have chosen to treat PCBs as the sum of congeners for this purpose. Studies of reproductive success of the Bald Eagle *Haliaeetus leucocephalus* in USA suggest 4 ppm ww. PCBs in eggs as a critical level (NOAEC).

Technical chlordane is a mixture of chlorinated hydrocarbons. Two metabolites —heptachlor epoxide and oxychlordane- have been identified as causing mortality in experimental birds given diets containing technical chlordane (Stickel et al. 1979), and corresponding levels (3-5 ppm ww.) have been detected in birds of prey found dead in the field (Blus et al. 1983)

In Sweden, seed-eating birds and their predators were severely affected by mercury poisoning through alkyl-mercury treated grain (Borg et al. 1969). After the ban on these compounds in Sweden in 1967/68, there was a rapid drop in mercury levels in terrestrial birds of prey, among them the Goshawk *Accipiter gentilis* (Westermark et al. 1975). Seed dressing with alkyl-mercury compounds was prohibited in Norway in 1968, and they were replaced by less toxic compounds based on alkoxy-alkyl mercury. The Norwegian wild fauna has been monitored for mercury content since 1965 (Holt 1969, Holt 1973, Holt et al. 1979, Frøslie et al. 1986). Mercury levels increase in successively higher levels in the food chain. A bioaccumulation factor of up to 1:1000 from water plants to fish-eating birds (via plankton and fish) has been found (Särkkä et al. 1978). Mercury is known to occur in naturally high levels in some marine organisms, such as fish and fish-eating predators (Koeman et al. 1975), whereas mercury levels in terrestrial herbivore birds generally are very low (Fimreite 1979). Newton & Haas (1988) established a critical level of 0.6 ppm ww. as a threshold concentration of mercury in eggs of Merlin, above which reproduction would be reduced. I have chosen this value as a critical level of mercury in raptor eggs. This level is supported by evidence from laboratory and field research (Tab. 1).

Table 1. Selected pollutants and some of their known effects at various levels of exposure, or as found in different organs.

Compound	Concentration	Organ	Species	Effect	Source
ΣPOPs	Not stated		Herring Gull *Larus argentatus*	Supernormal clutches in birds	(Fox & Boersma 1983, Fry et al. 1987)
Dieldrin	0.1 ppm	Egg	Bald Eagle *Haliaeetus leucocephalus*	Egg lethality	Wiemeyer et al. 1984
Dieldrin	0.7 ppm ww	Egg	Peregrine *Falco peregrinus*	Population decline	Newton 1988
Dieldrin	1 ppm ww	Liver	Sparrowhawk *Accipiter nisus,* European Kestrel *Falco tinnunculus*	Population decline	Newton 1988
Heptachlor epoxide	>1.5 ppm ww	Egg	American Kestrel *Falco sparverius*	Reduced productivity, adult mortality	Henny et al. 1983
Heptachlor epoxide	>8 ppm ww	Brain	Birds spp.	Lethal	Stickel et al. 1979
Lead	50 ppm	Diet	American Kestrel *Falco sparverius*	No effect on survival, egg laying, fertility or eggshell thickness	Pattee, O. 1984
Mercury	0.6 ppm ww	Egg	Merlin *Falco columbarius*	Reproductive output	Newton & Haas 1988

Methylmercury	0.5	Diet	Mallard *Anas platyrhynchos*	Reproduction, behavioural responses	Heinz 1979
Methylmercury	0.5 ppm	Diet	Mallard *Anas platyrhynchos*	4 % Shell thinning	Heinz 1980
Methylmercury	1.3-2 ppm	Egg	Pheasant *Phasanius colchicus*	Hatchability	Borg et al. 1966
Methylmercury	10 ppm	Diet	Goshawk *Accipiter gentilis*	Death	Borg et al. 1970
Methylmercury	3.7 ppm	Egg	Common tern *Sterna hirundo*	Hatchability	Fimreite 1974
Metoxychlor	Unknown			Estrogenic	in Fox 1992
Mirex	Unknown			Estrogenic	in Fox 1992
o,p'-DDT	in ovo, 2-5 ppm	Egg	Herring Gull *Larus argentatus*	Feminization in male embryos	in Fox 1992
Oxychlordane	>5 ppm ww	Brain	Birds spp.	Lethal	Stickel et al. 1979
Oxychlordane	3-4 ppm ww.	Brain	Red-shouldered Hawk *Buteo lineatus* and Great horned Owl *Bubo virginianus*	Lethal	Blus et al. 1983
p,p'-DDE	> 12 ppm ww	Egg	Bald Eagle *Haliaeetus leucocephalus*	Reduced reproduction	Nisbet & Risebrough 1994
p,p'-DDE	> 2 ppm ww	Egg	Prairie Falcon *Falco mexicanus*	Reduced reproduction	Fyfe et al. 1976
p,p'-DDE	> 3 ppm ww	Egg	Bald Eagle *Haliaeetus leucocephalus*	Reduced reproduction	Wiemeyer et al. 1984
p,p'-DDE	> 6 ppm ww	Egg	Merlin *Falco columbarius*	Reduced reproduction, reduced nest defence	Fyfe et al 1976
p,p'-DDE	in ovo, 20-100 ppm	Egg	Herring Gull *Larus argentatus*	Estrogenic	in Fox 1992
p,p'-DDE	> 25 ppm ww	Egg	White-tailed Sea Eagle *Haliaeetus albicilla*	Reduced reproduction	Helander 1994
p,p' DDE	40 ppm dw.	Diet	Mallard *Anas platyrhynchos*	21 % shell thinning, 145 % redaction of Ca-content in mucosa cells	Lundholm & Bartonek 1992
PCBs	> 4 ppm ww	Egg	Bald Eagle *Haliaeetus leucocephalus*	Egg lethality	Bowerman et al. 1995
PCBs	10 ppm	Egg	Cormorant *Phalacrocorax carbo*	Reduced hatching success	Dirksen et al. 1994

PCBs	10 ppm	diet	American Kestrel *Falco sparverius*	Potentiating shell thinning effect of DDE	Lincer 1994
Sum m-o PCBs	0.14 ppm ww *	Egg, (yolk sac)	Cormorant *Phalacrocorax carbo*	Incr. EROD levels, yolk sac size, incr. liver weights, reduced plasma thyroxin levels	van den Berg et al. 1994
TCDD	Not quantified		Rat *Rattus norvegicus*	Feminization at the anatomical, physiological and behavioural level	in Fox 1992
TCDD	Not quantified		Great Blue Heron (Ardea herodias)	Elevated EROD levels, subcutaneous oedema, growth retardation, changes in brain	in Fox 1992
PCDD	0.09 ppb ww *	Egg, (yolk sac)	Cormorant *Phalacrocorax carbo*	Head length of embryo, reduced plasma thyroxin levels	van den Berg et al. 1994
TCDD-eq	0.007 ppb ww	Egg	Bald Eagle *Haliaeetus leucocephalus*	Egg lethality	White & Setinak 1994

* Average value of a population with elevated levels compared to control.

Mammals.

Air pollution can change the quality of the food available to mammals. This can be caused by a change of the mineral and metal content of the food. Increased content of heavy metals may affect growth, development and behaviour of offspring, and the function of internal organs. Heavy metals (mercury, cadmium and lead) have no known function in organisms. Increases above normal background levels may therefore cause toxic effects.

Metals can be deposited directly on plant foliage from the atmosphere and be taken up from the soil (Harrison & Johnston 1987), and their soil availability may increase at low pH caused by acid rain. This will generally result in increased uptake of metals via plant roots, as there is a broad general relationship between metals in plants and in the soil (Davies et al. 1987).

The effects in mammals of metals and organochlorines will vary according to species, sex and age. Few effect studies of free-ranging species are available. Most of the studies merely document temporal and geographical differences in organ levels.

Heavy metals are normally found in the highest concentration in specific organs, such as kidneys, liver and bone, whereas persistent organochlorines accumulate in fatty tissue, e.g. liver. Many vital body functions are regulated by these organs. Consequently, this review focus on levels in relevant organs.

Norwegian data

Birds.

Since the analyses of chemical pollutants in eggs of wild birds in Norway started in the mid 1960's a substantial amount of data has accumulated. In the present material of POPs and heavy metals from Norway, the following substances have been analysed for: 33 PCB-congeners (SPCB), IUPAC nos. (after Ballschmiter & Zell 1980): -28, -47, -52, -56, -66, -74, -87, -99, -101, -105, -110, -114, -118, -128, -132, -137, -138, -141, -149, -151, -153, -156, -157, -170, -180, -183, -187, -189, -194, -196, -199, -206 and —209, 5 DDT components and metabolites: P,p'-DDT, p,p'-DDE, p,p'-DDD, o,p'-DDT and o,p'-DDD, chlordanes (heptachlor, heptachlor epoxide, cis-chlordane ,trans-chlordane, cis-nonachlor, oxychlordane and trans-nonachlor), TCDD, (and other dioxin and furans), hexachlorocyclohexane-isomers (a-HCH, b-HCH and g-HCH) and hexachlorobenzene (HCB) and mirex,

Systematic (grid-based) or random sampling of pollutants of higher taxa is difficult to achieve, due to their distribution patterns, insufficient numbers and protection status. A national programme for monitoring of the terrestrial environment in Norway (TOV) is established, and samples of birds and mammals are routinely collected for analysis. Their populations and productivity are monitored yearly in seven different areas in the mountainous regions of Norway and one in Svalbard (Løbersli 1989, Direktoratet for naturforvaltning 1997). However, data from top predators are difficult to obtain from such a system, due to their low breeding density. Therefore, data from the whole country have been utilised for birds of prey. The majority of material is reported in Table 2. Figures exceeding estimated critical levels are in bold letters.

Table 2. Levels of pollutants in eggs of raptors from Norway , 1974-94, given as wet weight values. The material was analysed at the National Veterinary Institute (own material and from Holt et al. 1979 and Frøslie et al. 1986) and at the Norwegian institute for Air Research (PCDF/D). Chlord. = Chlordane, Lind. = Lindane, and Dield. = Dieldrin. Figures exceeding estimated critical levels are in bold.

Species		DDE µg/g	PCB µg/g	HCB µg/g	Chlord. µg/g	Lind. µg/g	Mirex µg/g	Dield. µg/g	2,3,7,8 TCDD ng/g	TE PCDD/F ng/g	Hg µg/g
White-tailed	Mean	**4.429**	**14.321**	.090	1.051	.042	.073	**293**			.216
Sea Eagle	edian	2.720	8.398	.051	.843	.027	.042	**205**			.189
Haliaeetus albicilla	Min.	.141	1.120	.009	.034	.008	.011	.030			.010
	Max.	**60.000**	240.000	1.197	4.286	.255	.273	**1.120**			**.680**
	N	68	68	65	38	39	12	25			68
Golden Eagle	Mean	.388	1.033	.024		.025		.019			.078
Aquila chrysaetos	edian	.208	.560	.018		.020		.010			.039
	Min.	.041	.070	.006		.006		.008			.010
	Max.	2.150	5.163	.130		.084		.083			.360
	N	45	39	29		20		9			43
Goshawk	Mean	**4.368**	**12.613**	.097		.051		**.241**			.359
Accipiter gentilis	edian	**3.412**	**7.900**	.037		.017		**.237**			.316
	Min.	.695	1.264	.009		.012		8			.024
	Max.	**14.536**	**36.348**	.337		.139		**.661**			**1.106**
	N	19	16	14		6		11			19
Osprey	Mean	**5.836**	2665	.023	.010	.040		.012			.157
Pandion haliateus	edian	1.347	2.722	.015	.	.028		.007			.079
	Min.	.126	.379	.009	.010	.017		.005			.040
	Max.	**54.984**	**6.952**	.057	.010	.104		.040			.553
	N	14	14	12	1	6		10			13
Peregrine	Mean	**7.309**	**18.279**	.229		6.28		**1.966**	0.043*	0.185*	.445
Falco peregrinus	edian	**4.640**	**17.600**	.108		.475		**.460**			.460
	Min.	.139	2.161	.038		.156		.079	0.033*	0.179*	.142
	Max.	**28.323**	**34.617**	1.710		1.406		**12.036**	0.052*	0.190*	**.851**

	N	17	17	15		4		9	2*	2*	17
Rough-legged	Mean	.165	.827	.021		.028		.021			.105
Buzzard	edian	.094	.347	.014		.017		.007			.023
Buteo lagopus	Min.	.032	.071	.009		.017		.008			.010
	Max.	.569	**5.056**	.066		.055		.079			.553
	N	10	10	10		8		7			9
Sparrowhawk	Mean	**15.888**	**9.129**	.374		.094		**.396**			.183
Accipiter nisus	edian	**14.083**	**4.685**	.090		.096		**.320**			.158
	Min.	1.662	1.049	.019		.052		**.237**			.050
	Max.	**35.200**	**31.200**	1.560		.132		**.600**			.395
	N	10	10	10		4		6			7
Short-eared Owl	Mean	2.464	1.580	.018				.015			39
Asio flammeus	edian	.	.	.				.			.
	Min.	2.465	1.580	.019				.016			.040
	Max.	2.465	1.580	.019				.016			.040
	N	1	1	1				1			1
Eagle Owl	Mean	1.327	**4.186**	.034		.052		.008			.040
Bubo bubo	edian	.336	1.003	.010		.		.008			.021
	Min.	.024	.220	.006		.052		.008			.010
	Max.	**6.083**	**15.800**	.114		.052		.009			.158
	N	8	8	7		1		4			8
Merlin	Mean	**9.1**	1.4	.11	0.11	.16	0.005	**.170**			.25
Falco columbarius	edian	**6.0**	1.0	.04	0.11	.08	0.006	**.170**			.21
	Min.	1.4	.25	.003	0.003	.02	0.0007	.050			.015
	Max.	**39.9**	6.6	1.1	0.28	1.1	0.01	**.290**			**0.88**
	N	43	43	41	37	39	12	2			41
Gyrfalcon	Mean	1.099	2.504	1.683		.234		.007			.153
Falco rusticolus	edian	1265	2356	187		.057		.			.158
	Min.	.289	.883	.002		.017		.008			.130
	Max.	1.577	**4.424**	.295		.630		.008			.171
	N	4	4	4		3		1			3

* M. Oehme & R Bergstrøm, unpublished material.

Norway receives pollutants transported by long-range transport, which have proven very detrimental to our wildlife resources (80,000 km^2 in Norway have damaged fish stocks due to acid rain; NIVA 1993). Considerable geographical variation in pollutant dose received by different parts of the country is expected, due to gradients in precipitation and distance from pollution sources. The regional variation in the concentration of different pollutants in eggs of White-tailed Sea Eagle is shown in Fig.1. The DDE, PCBs and other POPs as well as mercury declined from south to north (Nygård & Skaare in press). The declining trends in POPs from south to north is consistent with the deposition pattern of PCBs in Scandinavia (Södergren 1972), and the general pattern of deposition of pollutants to Norway (Tørseth & Semb 1995). The White-tailed Sea Eagle is a sedentary bird, whereas the Merlin is a migrating species. The geographical variation in POP level in Merlin eggs was not significant, but there were significantly higher mercury levels to the north (Nygård 1997). The differences in mercury levels among regions (Fig. 2) may be explained by differences in local environmental concentrations, but the actual levels will be determined by the levels in prey prior to egg-laying. The merlin continues to prey on partly migrating bird species after returning to Scandinavia in the spring. The migrants themselves represent a vehicle for transboundary pollutant transport.

Steinnes et al. (1995) found elevated mercury content in moss collected in the northernmost part of Finnmark, probably as a result of dry deposition. It is, however, difficult to establish a link between that gradient and our results. The Merlin eggs from Finnmark came primarily from the inland, where levels of mercury are moderate to low. The most plausible cause for the high levels of mercury is mercury-rich prey items in the north, possibly in combination with the foraging patterns in the winter quarters. As the eggs are produced at the site of breeding, a major contribution will likely come from local food sources.

As use and discharges of pollutants have been banned, use has been severely restricted and abatement measures have been introduced, declining pollutant levels over time are expected. Nygård (1997) found that the expected half-times for DDE and PCBs in eggs in White-tailed Sea Eagle were 17 and 19 years, respectively, while the mercury levels were stable.

The most readily observed effect parameter of chemical pollution is eggshell thickness. As it is directly connected to successful reproduction, it is highly relevant. There is a linear relationship between eggshell thickness and log-p,p' DDE, but other compounds may also be involved, such as PCB (synergistic, potentiating (Lincer 1994) and Hg (Table 1.). The available data on shell thickness in Norwegian bird of prey species is shown in Table 3.

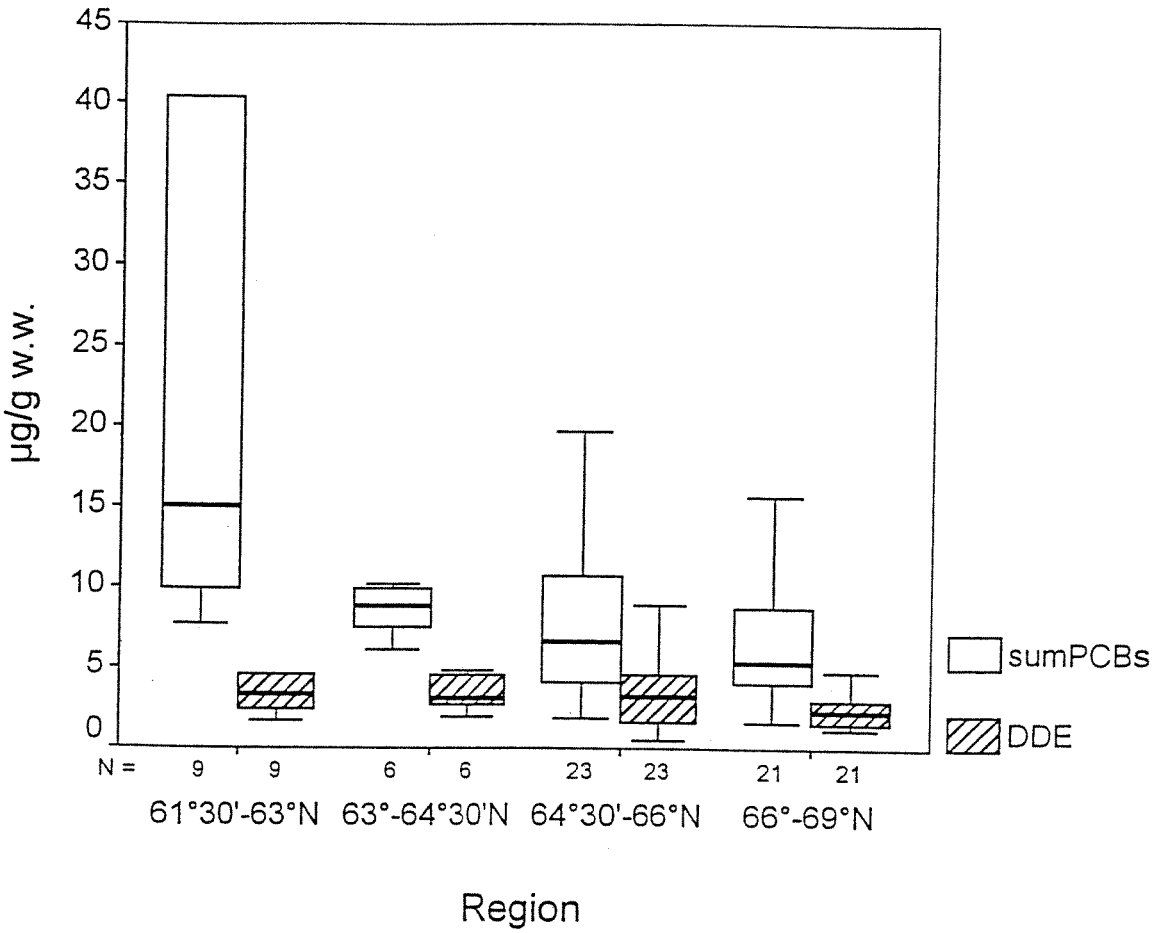


Figure 1. Geographical variation in organochlorine concentration in eggs of White-tailed Sea Eagle in Norway 1974 - 1994. The values are given as (µg/g) on fresh weight basis, based on clutch averages.

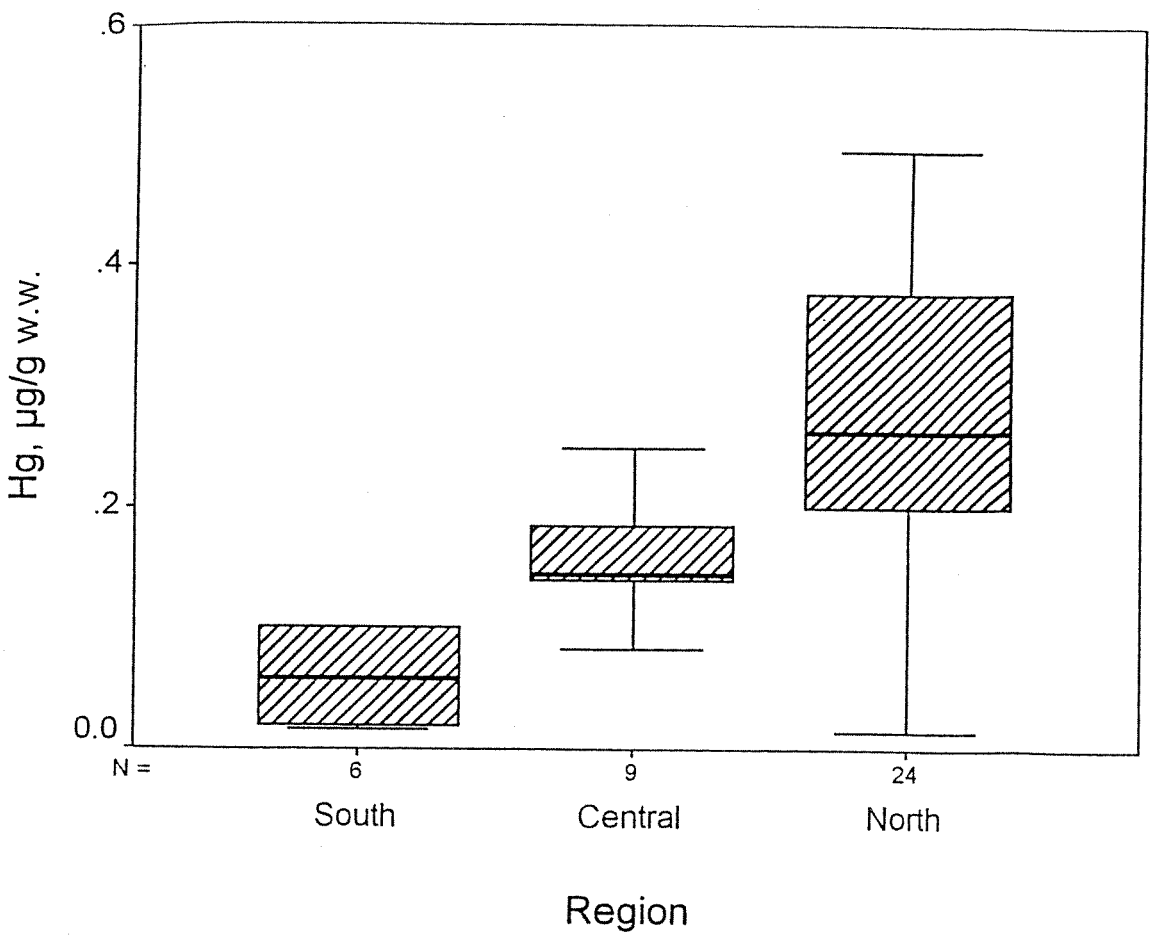


Figure 2. Mercury in eggs of merlin in Norway by region. Values are on a fresh weight basis, based on clutch averages. The difference between regions is significant, using ANOVA, controlling for time-effects (Nygård, 1997).

Table 3. Eggshell thickness in Norwegian birds of prey before and after 1947 (mm/1000, based on average clutch values), mainly own material (Nygård 1990, Nygård 1997).

Species	Mean thickness Before 1947	n	Mean thickness After 1947	n	Diff.	p
White-tailed Sea Eagle	619	29	600	71	-2.2*%	0.045
Golden Eagle	601.8	7	597.6	24	-0.7%	0.84
Sparrowhawk	270.7	2	227.2	6	-15.8%	-
Goshawk	436.6	32	407.9	14	-6.6%	<0.001
Osprey	514.1	18	444.0	14	-13,6%	<0.001
Peregrine	361.4	18	308.2	22	-14.7%	<0.001
Merlin	241.9	23	212.9	52	-12%	<0.001
Eagle Owl	416.7	7	387.5	7	-7.0%	0.086

* Thickness index (Ratcliffe 1967) was —6.0%

The Willow Ptarmigan *Lagopus lagopus*, a bird with a wide distribution in Norway, has tested as a biomonitor of environmental metal levels (Wren et al. 1994). It has been reported to have very high levels of Cadmium in kidney and liver in many mountain regions of Norway, especially in South Norway (Herredsvela & Munkejord 1988, Pedersen et al. 1995), indicating an effect of long-range transport. High levels have been reported from several other areas (Kålås & Lierhagen 1992, Myklebust 1992, Wren et al. 1994), often above suggested critical levels in kidneys (13-15 ppm, Eisler 1985, Pedersen & Nybø 1990) The Cd levels seem to be governed primarily by two factors, age of the bird (increasing levels with age) and the mineral content in the local bedrock (reflected in the local browse plants). Therefore, it is difficult to use cadmium in Willow Ptarmigan for evaluation of critical load. However, lead in liver from different grouse species seems to follow the general pattern of atmospheric deposition of this metal (Direktoratet for naturforvaltning 1997). Grouse species are important prey species for two top predators in mountainous regions in Norway; Gyrfalcon *Falco rusticolus* and Golden Eagle *Aquila chrysaetos*. The acid rain problem in southern Norway has made it necessary to put focus on aquatic birds in their polluted environments.

The Dipper *Cinclus cinclus* takes its food directly from running freshwater, such as insect larvae, gastropods, crustaceans and small fish. Eventual effect from long-range transport of pollutants are to be expected in watershed most affected by acid rain and pollutants transported by precipitation. A comparative study of heavy metal concentration in Dipper by Nybø et al. (1995) showed that eggshells were 6% thinner in the polluted area (southern Norway) vs. control (central Norway). It was hypothesised that this was due to a calcium-deficient diet in the acidified streams.

Mammals.

In case of the organochlorines, the species at high trophic levels are more at risk through bio-accumulation effects. This, however may not be the case for heavy metals, as only certain organic forms such as alkylmercury have molecular properties that allow them to bio-accumulate. Very few data on OCs in Norwegian mammals from the terrestrial environment are available.

Mustelids, especially mink, have been reported to be very sensitive to POPs, and PCB in particular. The PCB levels in Otter have been monitored in Norway in more than two decades. However the are no evidence of any anomalies in reproduction connected to the prevalence patterns of PCBs, and the mean levels were from four to eight ppm PCBs on a lipid basis. (Christensen & Heggberget 1995). This is slightly below the suggested No Observed Effect Level (NOEL) for this species (AMAP 1997).

Herbivores may be exposed to elevated levels of heavy metals through browse effected by air-pollution. Such metals will not normally bioaccumulate through food-chains, as they are normally deposited in specific organs, such as kidney, liver and bone. The highest concentrations in mammals are usually found in southern Norway (Fig. 3). For lead, this is documented in moose *Alces alces* (Frøslie et al. 1984) common shrew *Sorex araneus*, bank vole *Clethrionomys clareolus* and for mountain hare *Lepus timidus* (Kålås & Lierhagen 1992). There were a good correlations between the lead concentration in liver from moose and the levels in moss from the corresponding sites, indicating air pollution as an important pathway of lead to the moose. However, this was not as clearly seen in the wild reindeer *Rangifer tarandus*, probably due to the fact that this species is not present in the southernmost (air-pollution effected) areas of the country (Frøslie et al. 1984). However, reindeer from central mountain plateau in South Norway generally have higher lead levels than those from Finnmark (northernmost Norway) (Sivertsen et al. 1991).

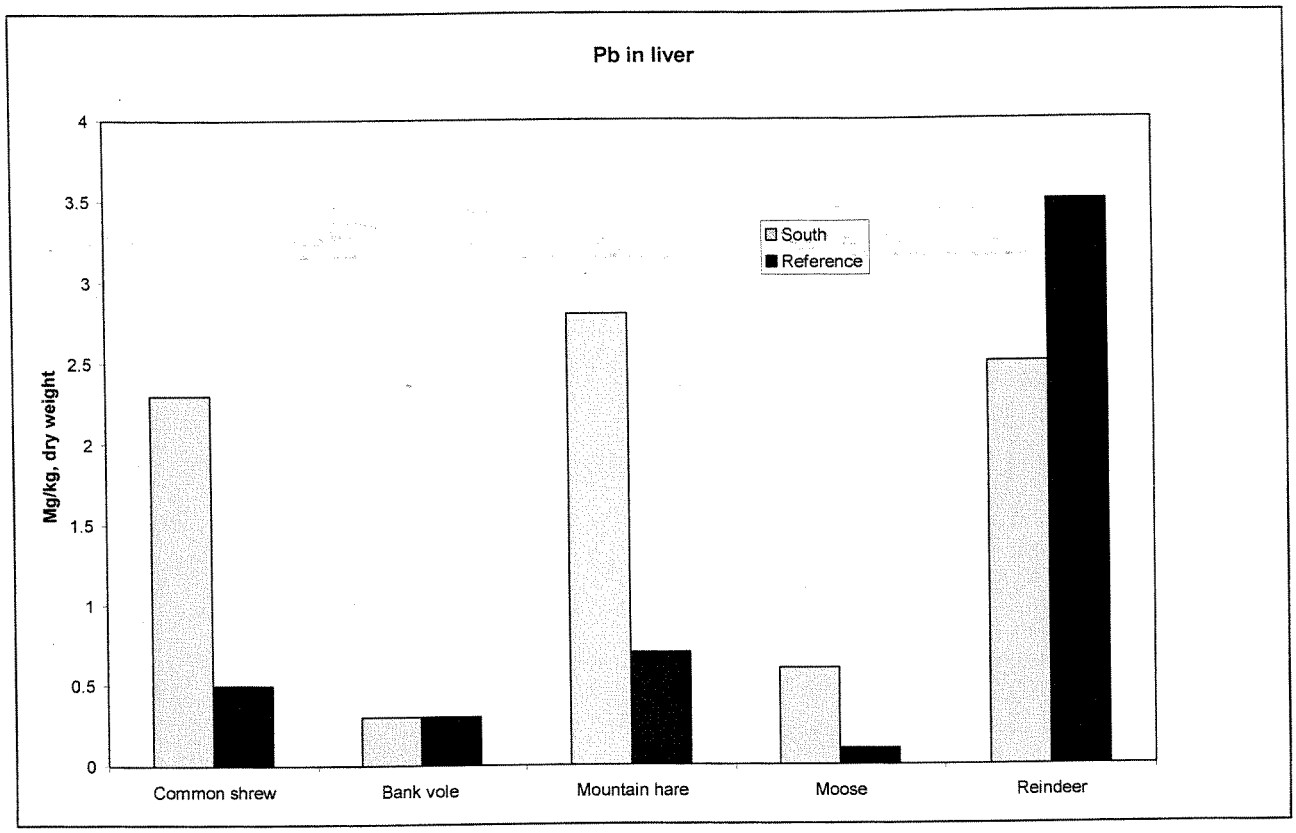
The same regional pattern is seen for cadmium. (Frøslie et al. 1986, Holt & Frøslie 1987). Small rodents from the south also have higher cadmium levels than in the north (Kålås & Jordhøy 1995, Kålås et al. 1995a, 1995b).

Few studies on mercury levels in terrestrial mammals from Norway have been made, but they all indicate low levels and no clear geographical gradients, except for reindeer, where the highest levels are found in the south (Frøslie et al. 1984, Sivertsen et al. 1991).

Heavy metals have a tendency to increase with the age of the animal. This is particularly prominent for cadmium, where kidney levels will increase with the age in mammals (Frøslie et al. 1986, Holt & Frøslie 1987). Lead will tend to accumulate in bone (Kålås & Myklebust 1994). An age-effect of mercury in mammals is not evident. Also, higher levels of heavy metals have been found in male moose than in female ones (Kålås & Myklebust 1994).

The Cd levels in livers of mammals from Norway normally range from 1 to 10 ppm dry weight. This is below what is considered to be indications of Cd pollution (10 ppm wet weight, Eisler 1985).

Little is known about toxic levels of Pb to free-ranging mammals. Laboratory experiments, however, have shown abnormal behaviour of animals given as low doses as 0.1 to 0.5 ppm Pb in diet for extended periods resulting in elevated tissue concentrations. There are differences in sensitivity between species, and young individuals are more susceptible than older ones (Eisler 1988).



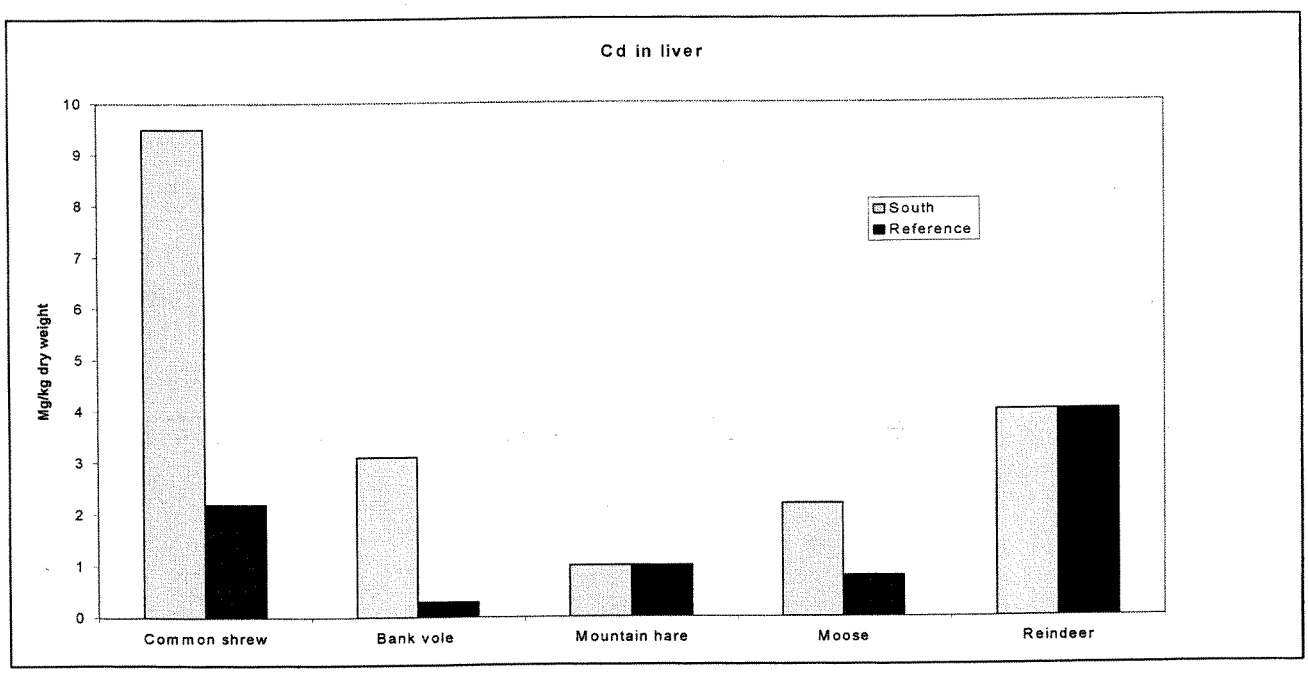


Figure 3. Regional differences in Pb and Cd in liver of some species of mammal from different geographical areas in Norway.

Discussion

Birds.

Estimated critical levels of POPs have been exceeded in some or many cases in most of the species examined (Tab. 2). For DDE, mean population levels above critical have been detected in Goshawk, Osprey, Peregrine, Sparrowhawk and Merlin. For sum PCBs, the same has been found for White-tailed Sea Eagle, Peregrine, Sparrowhawk and Great horned Owl. Regarding dieldrin, the U.S. estimate of 0.1 ppm as a critical level has been exceeded on a mean population level for White-tailed Sea Eagle, Goshawk, Peregrine, Sparrowhawk and Merlin. Using the more conservative estimate for the Peregrine from Britain of 0.7 ppm, only the Peregrine has levels above critical. For mercury, no species had average values above the suggested 0.6 ppm ww. but Goshawk, Peregrine and Merlin had values approaching this level. Most species have some individual specimens exceeding estimated critical concentration of some or one of the listed chemicals. The species living primarily of rodents and herbivorous birds are the ones least affected, such as the Golden Eagle, Rough-legged Buzzard, Short-eared, Tengmalms and Ural Owls.

For the other POPs, there is too little data on the possible effect on populations. I still want to point out the high levels of chlordanes found in White-tailed Sea Eagle and Merlin, which may have biological significance.

The POPs dealt with in this paper are believed primarily to be brought to Norway by long-range transport, as a part of the global large circulatory systems driven by atmospheric movement, precipitation by rain and snow, and by oceanic currents. The gradient in POPs shown in Sea Eagle eggs seems to support this hypothesis.

The cold condensation theory predicts higher mercury levels to the north (Wania & Mackay 1993), The higher mercury levels found in Merlin eggs from Finnmark than elsewhere in the country seem to be in concordance with this, but there are many ecological variables that could be playing a role, e.g. food choice (Nygård 1997). The mercury levels in Norwegian birds of prey are still elevated compared to background levels (approximately doubled, Nygård 1997). Similar results were found in several species of seabirds from the north east Atlantic Ocean (Thompson et al. 1992) and the Baltic Sea and Kattegat (the strait between Denmark and Sweden/Norway; Appelquist et al. 1985), indicating that the increase of mercury in marine environments in this part of the world is real. Slemr & Langer (1992) reported an increase in atmospheric levels over the North Atlantic between 1977 and 1990. However, Iverfeldt et al. (1995) found a decrease of gaseous mercury in the air over southwest Scandinavia of ca. 15% between 1980-84 and 1990-92.

Analyses of lead in moss in country-wide surveys have shown a clear pattern of elevated levels in the south, explained by long-range pollutants in Norway (Steinnes et al. 1994). The lead levels in Grouse species will largely be determined by the lead on the foliage that originates from atmospheric deposition, as the uptake via plant roots is limited (Kabata-Penidas & Penidas 1984, Kålås & Lierhagen 1992). For lead, there may therefore be a potential for connecting critical limits in birds and environmental critical loads that should be explored.

Mammals.

The question of critical limits to marine mammals have been dealt with elsewhere (Skjegstad et al., annex 5), and will not be discussed here, as it seems to be a problem more pressing in the high Arctic than on mainland Norway. There is evidence that the Reindeer *Rangifer tarandus* population is affected by uptake of long-range transported heavy metals from lichens, their main winter-food (Strand et al. 1995). Elevated levels due to uptake via air-polluted browse also seem to be the case for other mammals, such as moose, mountain hare and small rodents. Very little is known about critical levels for exposure to these species, especially regarding ecological effects of long-term sublethal doses.

Conclusions

The critical load concept does not seem applicable to higher terrestrial animals, as is it is extremely difficult to relate deposition rates to resulting concentrations in their tissues. This is mainly due to the influence of many environmental and ecological variable acting on the intake rates, such as local soil and bedrock conditions, age and condition of the animals, local food variations and individual preferences. In addition, there are species-specific accumulation rates and sensitivities.

However, the concept of critical limits of POPs and heavy metals seem to be applicable to birds of prey. Eggshell thinning, reduced viability of eggs, resulting in lowered reproductive output are parameters that easily quantifiable, relatively easy to interpret, and universally accepted as critical factors for any population of birds. It is well documented that POPs, especially DDE and PCB are active and harmful compounds in this respect, and they have to be handled on an international level. Data on heavy metals are less conclusive, except perhaps for methylmercury, which seem to behave in many respects as the POPs, and is still found at elevated levels in this part of the world.

For terrestrial mammals, there is no evidence that critical levels are reached in any species of mammals in Norway, but little is known about possible long-term effects of the elevated, but sublethal levels found.

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Annex 5.

Assessment of the applicability of the term critical load for persistent organic pollutants in actic biota.

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Assessment of the applicability of the term critical load for persistent organic pollutants in arctic biota

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Objective

The objective of the present document is to assess the applicability of the concept *critical load* for persistent organic pollutants in Norwegian/arctic marine and terrestrial mammals, and seabirds, with emphasis on polychlorinated biphenyls (PCB). The purpose of this assessment is:

1) To give an analysis of the applicability of the term *critical load* with respect to persistent organic pollutants.
2) To provide advice to environmental protection authorities for discussions in Norwegian and international fora about critical loads.

Because high concentrations of PCBs are found in e.g. polar bear and glaucous gull in Arctic, a lot of concern are focused around this group of pollutants. Dead and dying glaucous gulls are found with high concentrations in their body. Hermaphrodites are found among polar bears, and recently, some cubs of the polar bear are found to be smaller than normal (Derocher, pers. comm.). The suggestion is that PCB are causing all these effects.

This assessment was also supposed to deal with critical loads for heavy metals. Although relatively high concentrations of cadmium and mercury are found in seabirds, seals, whales and polar bear, no effect studies regarding this group of pollutants in various biota in Arctic are performed during the AMAP programme. We defined heavy metals as beyond the scope of this report because of too little information to base the conclusions on. Thus, within the limited time available for this work, PCB had priority.

The critical load concept

To derive the deposition levels at which effects of acidifying air pollutants start to occur, the *critical load* concept was introduced (Nilsson, 1986). The UN/ECE Working Group on Nitrogen Oxides has defined the critical load for an ecosystem as: "A quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge" (Nilsson & Grennfelt, 1988).

In this connection *critical load* for an ecosystem is given as an amount input of substances/matter per area or volume and time unit (kg/km^2/year or kg/m^3/year). It is conceptually different from the term *critical concentrations* or other terms which are referring to levels and not input. Critical concentration is not related to time as critical load is, and refer to concentrations in organs.

According to the suggestions for priorities for the Bad Harzburg Workshop, one of the objectives is to identify receptors for calculation of critical limits (concentrations). The present document focuses on levels and concentrations of PCBs in different arctic/Norwegian biota. As far as possible, the observed levels from different studies are compared with threshold values (critical concentrations) for some types of effects which is reported in the litterature.

Limitations

There are problems associated to assessment of national critical loads which should be considered. These problems relate to the country or region in which the studies are performed, on which species are investigated, and on whether the studies are performed in the field or in a laboratory. In addition to this, one has to consider many biological and physiological parameters that might differ between the studies and certainly influence the results. The parameters that might affect the response to different POPs is species and even strain specific, and dependent on dose, season, tissue, condition, age, sex, reproductive status etc. One also has the problem that there are different effects at different doses of different pollutants. These aspects make it difficult to generalize which levels should be set as critical loads.

A good reason for caution in extrapolating results from controlled experiments to natural conditions is that they are usually focused on single pollutants, and rarely on mixtures which are present in the wild biota. Relatively few studies assessing synergistic and antagonistic effects of chlorinated organic pollutants have been performed. Relatively little knowledge exists about long-term effects of persistent organic pollutants (POPs).

Effect assessments include many uncertainties. A main problem is how to extrapolate results from laboratory conditions to field conditions. For example, the assessments assume that animals in their natural environment have approximately the same sensitivity as the animals that were used in the toxicological studies. In reality, they could be more sensitive or less so. Thus, when POP levels in arctic biota reach biological effect thresholds determined from animals observed in various studies, it should be interpreted as a warning signal rather than as evidence that such effects actually do occur in the Arctic (from the AMAP report, 1997).

The main background material assessing critical loads in arctic biota has been taken from the AMAP (Arctic Monitoring Assessment Programme) report (1997). This report summarizes levels of a number of POPs in many species in the circumpolar Arctic. Some threshold levels for toxic effects (NOEL) in various animals are also given. These levels are used as basis for our assessment of critical loads in arctic biota.

In general, acute NOEL has been determined by examining the data and setting the no-effect concentration as the highest concentration that does not result in greater than 10 % effect. The effects can be mortality, immobilization, reduced cell count (algae), or behavioral observations. This endpoint is not determined statistically, however, and a standarized approach for determining the no-effect concentration is needed (Hoffman *et al.*, 1995).

The present assessment is supposed to be based on the conditions prevailing in the Norwegian Arctic. However, most of the studies referred to in AMAP are Canadian, and only a few are Norwegian, which makes it difficult not to refer to the Canadian studies. This means that we have extrapolation, not only among areas, but also among species.

Toxicological endpoints and biomarkers

Toxicological endpoints are values derived from toxicity tests resulting from specific measurements made during or at the conclusion of the test. In most of the cases these endpoints are calculated in the laboratory. Endpoints which are referred to in the following chapter are EC50 (the concentration that has an effect on 50 % of the organisms tested), no-observed effect level or -concentration (NOEL/NOEC)) and low-observed effect level or -concentration (LOEL/LOEC).

No biomarker are yet fully developed to be applicable for use in environmental monitoring. Nevertheless, biomarkers indeed have a future potential as a tool in assessment of biological effects of pollutant exposure and thus deserve some attention here. Very few studies of the species of

concern in the Arctic have been dealing with biomarkers. However, the cytochrome P450 system and thyroid hormones have been studied in polar bear.

Biological effects can be measured at different levels of biological organization, from the molecular to the ecosystem level. At a molecular level bio-indicators respond at an early stage (e.g. induction of the cytochrome P450 system) after exposure but are not readily interpreted ecologically. On the other hand, parameters of ecological relevance, such as population decline or reduced reproductive rates respond too late to have diagnostic or preventative value.

The following focuses on levels of PCB, TEQ_{TCDD} concentrations and effects reported in different studies with emphasis on AMAP-results. The chapter is divided into arctic mammals (seals, cetaceans, polar bear and arctic fox) and sea birds.

When known, lipid weight (lw) is given to the levels which are summarized in the following. When lw is unknown only wet weight is given.

Arctic mammals

As already pointed out, in most cases there is a need for comparing different species in regard to threshold values for effects. Of course, this might give a wrong picture of the situation for the actual species. However, as thorough effect studies of every persistent organic pollutant in every species is virtually impossible, extrapolation among species is inevitable.

Polar bear

Ranges of measured TEQ_{TCDD} concentrations in arctic marine and terrestrial mammals are shown in Table 1. The concentration levels in polar bear both from Canada and Svalbard partly exceed the level (209 pg/g lipid weight (lw)) which are associated with immune suppression in captive harbor seals. On the other hand, however, they are below the EC_{50} level in mink liver for kit survival (10,000 pg/g lw).

Table 1. Ranges of measured TEQ_{TCDD} concentrations (pg/g lw) (from PCDD/Fs, non-ortho and mono-ortho PCBs) in arctic marine mammals (AMAP report, 1997).

Species	Concentration range (pg/g lw)
Polar bear (Canada)	20-230
Polar bear (Svalbard) (1990-1994)	80-260
Beluga	3-25
Narhwal	9-20
Ringed seal	2-30

Levels of PCDD/PCDF in adipose tissue (58-73 % lipid - Ramsay *et al.*, 1992) in Canadian polar bears sampled in 1983-84 ranged from 2-23 pg TEQ/g wet weight (ww). The mean levels of PCDD/PCDF, non-ortho and mono-ortho PCBs in liver from Canadian bears collected in 1992-1994 were 27, 29 and 172 pg TEQ/g lw, respectively (Letcher *et al.*, 1996). The combined TEQs exceed those associated with immunosuppression in harbor seal (209 pg TEQ/g lw). Norheim *et al.* (1992) found PCB levels in polar bears at Svalbard close to those connected with reproductive disorders in Baltic seals (Helle *et al.*, 1976; Bergman & Olsson, 1985; Olsson *et al.*, 1992). Mono-ortho PCB levels were measured in polar bears from Svalbard (Bernhoft *et al.*, 1997) and the levels were in the range of 42-102 pg TEQ/g ww, or 80-260 pg TEQ/g lw (Table 1). These levels are comparable to or higher than the mono-ortho PCB TEQs (140 pg TEQ/g lw) associated with immunosuppressive effects in harbor seal.

Assuming that marine mammals are as sensitive to PCB as mink, mean levels of ∑PCB in ringed seal blubber from 16 sites in the Canadian and Norwegian (Svalbard) Arctic, exceed the dietary (mink intake) NOEC concentration for reproduction of 72 ng/g ww (Heaton *et al.*, 1995). This is below both the US EPA guidelines (regarding PCBs) for protecting aquatic wildlife (250 ng/g ww) and the International Joint Commision guidelines (100 ng/g ww) (AMAP report, 1997).

Ranges of ∑PCB concentrations in different arctic mammals, and threshold values for mammalian effects for ∑PCB concentrations (from the AMAP report), are shown in Table 2 and Table 3 respectively.

Table 2. Ranges of ∑PCB concentrations (ng/g lw) measured in arctic mammals (AMAP report, 1997).

Species	Concentration range (ng/g lw)
Polar bear, Svalbard	7,000 - 90,000
Polar bear, circumpolar (inluding Svalbard)	3,000 - 90,000
Arctic fox, Svalbard	2,000 - 240,000
Beluga	2,000 - 8,000
Narhwal	5,000
Ringed seal	500 -5,000
Walrus	100 - 20,000
Harbor porpoise	2,000 - 25,000
Harp seal	500 - 6,000
Mink liver (LW)	2 - 70

Table 3. Threshold values for mammalian effects in respect to ∑PCB concentrations (ng/g lw) (AMAP report, 1997).

Concentration for effect (ng/g lw)	Mammalian effect	Species
90,000-110,000	EC50 kit survival	Mink liver
90,000	Poor reproductive success	Baltic ringed seal
60,000-70,000	EC50 litter size	Mink liver
40,000	Poor reproductive success	Captive harbor seals, blood
35,000	LOEL, immune effects	Rhesus monkey blood
9,500	NOEL, kit survival	Mink muscle
9,000	NOEL, reproduction	Otter muscle
1,000	NOEL, visual memory	Rhesus monkey offspring
800	LOEL, short-term memory	Human offspring cord blood serum

Arctic Fox

No studies of biological effects have been performed on arctic fox (AMAP report, 1997). So far, levels of OCs have only been studied for individuals on Svalbard. On a wet weight basis, OC concentrations in arctic fox liver are similar in specimens collected in 1983-84 and 1993-94. Assuming a lipid content of 6 % (AMAP report), ∑PCB levels in arctic fox liver from 1993-94 range from 2,250-236,700 ng/g lw (Table 2), which are higher levels than found in polar bears. Mean levels were 30,100-63,600 ng/g lw and exceed the NOEL and LOEL for subtle neurobehavioral effects, the NOEL for kit survival in mink, levels known to cause poor reproductive success in harbor seals and the EC50 for reduced litter size (Table 3). Some individuals exceed the levels associated with poor reproductive success in ringed seals and the EC50 for kit survival in mink as well. The lowest adverse effect level for immunosuppression is 21,000 ng ∑PCB/g lw in rhesus monkeys. The mean ∑PCB levels in arctic fox from 1993 and 1994 are above this level.

As arctic fox from Svalbard also eat ringed seal, the dietary intake assessment given above for polar bear is probably also valid. This means that ∑PCB levels in ringed seal blubber from all sites exceed

the dietary NOEC for reproductive effects as well as all guidelines for protecting aquatic wildlife. Arctic fox from Svalbard are therefore at risk for the immunosuppressive effects of PCBs and probably also dioxin-like substances, as well as the reproductive and developmental effects of PCBs.

Cetaceans

Relatively low levels of ∑PCB are found in different cetaceans in Canadian areas (Table 2). In beluga the levels of ∑PCB exceed the NOEL and LOEL for initial neurobehavioral effects, but are below the NOEL for otter reproduction and mink kit survival (Table 3). Experiments with harbor seals fed Baltic herring, indicated immunosuppression at total TEQ levels of around 210 pg/g lw in blubber (Ross *et al.*, 1995). The combined TEQs in beluga range from 4.2-25.3 pg/g ww in blubber, which is usually about 90 % lipid. Thus, the TEQ levels in beluga are considerably lower than those associated with immunosuppresive effects in seal. This is the main picture also for narwhal and minke whale.

In blubber from harbor porpoise from West-Greenland the mean ∑PCB level was 2600 ng/g ww. In animals from the southern Barents Sea the mean ∑PCB level was 24,500 ng/g ww. The NOEL and LOEL for neurobehavioral effects is exceeded in harbor porpoise from both sites. The ∑PCB levels in S. Barents Sea porpoises also exceed the NOEL for otter reproduction and mink kit survival, the levels associated with immunosuppression as well as approaching the levels associated with poor reproductive success in harbor seal. No biological effect studies have been carried out on harbor porpoise.

Seals

The mean ∑PCB levels in Arctic harp, ringed, harbor and grey seals were in the range 241-5700 ng/g ww in blubber (85-95 % lipid), which exceed the NOEL and LOEL for initial neurobehavioral effects, but are below the NOEL for otter reproduction and mink kit survival (AMAP report, 1997). The levels in the previous mentioned seals are also considerably lower than those associated with poor reproductive success in harbor and ringed seal (Table 3). Ringed and harp seal from Svalbard, Barents Sea, Greenland Sea and several sites in Canada have been analyzed for PCDD/PCDF or non-ortho PCB TEQs. The levels were somewhat lower than the sum of PCDD/PCDF and mono-ortho PCB TEQs (69 pg TEQ/g lw) associated with immunosuppressive effects in harbor seal .

Seabirds

Threshold values of ∑PCB (ng/g ww) for avian effects are shown in Table 5. Unfortunately, we do not have such effect levels for glaucous gull, a top predator in the Arctic. As a top predator the glaucous gull is more at risk for being exposed to POPs due to biomagnification through the food chains.

Table 4. Ranges of ∑PCB (ng/g ww) in eggs of Arctic birds (AMAP report, 1997).

Species	Concentration range (ng/g ww) in eggs
Murres	200 - 900
Black guillemot	100 - 500
Common guillemot	400 - 100
Kittiwake	500 - 1,200
Glaucous gull	200 - 2,200
Herring gull	700 - 2,700

Table 5. Threshold values of ∑PCB (ng/g) for effects in different bird species (AMAP report).

Concentration for effect (ng/g ww)	Avian effect	Species
11,000	NOEL reproduction	Black crowned night heron
8,000	NOEL reproduction	Common tern, Great blue heron
6,000	LOEL deformities	Doble crested cormorant
5,400	LOEL egg mortality, egg production	Falcon, herring gull
3,400	LOEL egg mortality	Doble crested cormorant
2,200	NOEL hatching success	Forster's tern
1,400	LOEL hatching success	White-leghorn chicken
600	NOEL hatching success	White-leghorn chicken
200	NOEL deformities	White-leghorn chicken

Ranges of ∑PCB in eggs of Arctic birds are shown in Table 4, and the effect levels are summarized in Table 5. The levels measured in glaucous gull (200-2,200 ng/g ww), are below LOEL for egg mortality in double-crested cormorant, herring gulls and falcons. The levels are also far below the NOEL for reproduction in black crowned night heron, great blue heron and common tern. However, in a ranking of species according to sensitivity toward PCB 77, white leghorn chicken appeared to be the most sensitive, while herring gull was the least sensitive (Bosveld, 1995). Different sensitivities between different species may be important when comparing levels/effects (Kennedy *et al.*, 1996).

Norwegian level/effect studies

Very few Norwegian studies are performed by correlating levels of OCs to some kind of effect. Thus we summarize the most relevant studies which have measured levels of OCs in Norwegian arctic animals. In most of the studies, only levels are measured and no effect parameters. In such cases, we can only compare the levels with NOELs/LOELs for effects in relevant species found in previously mentioned studies. However, some studies with biomarkers are performed and results from some of these are summarized here as well.

Polar bear

As dose effect relationships and critical concentrations are not known in polar bear, we have to use data from other species. All ∑PCB levels in polar bears from Svalbard, sampled from 1990 to 1994, exceed the NOEL and LOEL levels found for subtle neurobehavioral effects in offspring if polar bear is as sensitive as mink (AMAP report, 1997). The ∑PCB levels (Table 2) also exceed the NOEL for kit survival in mink (9,500 ng/g lw) at four of the sites in which it was sampled - among them at Svalbard. LOEL for immunosuppression is 21,000 ng ∑PCB/g lw in rhesus monkeys. ∑PCB levels in polar bear from Svalbard, among other sites, are at or above this LOEL.

Some studies with biomarkers in polar bear have been performed. Skåre *et al.* (1994) investigated levels and effects of PCB in polar bear from Svalbard. The study tried to reveal effects on the hormone balance, vitamin status and reproduction. The young bears had the highest levels of retinol and thyroid hormones, while the lowest levels were found in adult males. The study also revealed a significant negative correlation between retinol and ∑PCB, even after correction for the influence of age and sex. The investigation suggests that retinol and thyroid hormones may be used as biomarkers for exposure and effects of PCBs in polar bear.

Recently, the cytochrome P450 system of the polar bear was characterized by Bandiera *et al.* (1995). Studies performed by Letcher *et al.* (1994a) revealed that CYP1A1, CYP1A2 and CYP2B1 were induced by PCBs in polar bear. The mean cytochrome P450 content in polar bear liver, measured as

EROD activity, was about two times higher than in beluga liver (White *et al.*, 1994) and 10 times higher than activity in male hooded seals (Goksøyr *et al.*, 1985). Assuming that the interlaboratory avtivities are comparable, and it is possible to compare species, this means that the cytochrome P4501A mediated activity was elevated in polar bear. P4501A1 protein levels were ten times higher than 1A2 levels.

CYP1A1 and CYP1A2 content in male polar bear hepatic microsomes was correlated with levels of coplanar PCBs (non-ortho and mono-ortho-substituted) expressed as TEQ. EROD activities in polar bear liver correlated with total TEQ to a concentration of 350 pg/g lipid (Letcher *et al.*, 1996). The strong positive correlation of cytochrome P4501A with non-ortho and mono-ortho PCBs in polar bear liver suggests that these compounds may be responsible for the EROD activity.

Seabirds

Murvoll (1996) studied effects of PCB on vitamin A- and thyroid hormon status in double-crested cormorant. Mean ∑PCB in chicks from three nesting sites along the coast of the Southern Norway (Rogaland, South- and North-Trøndelag) was 1390 ng/g ww or 17933 ng/g lw. The concentrations based on wet weight approached the LOEL for hatching success in white-leghorn chicken, but were far below NOEL for reproduction in black-crowned night heron, common tern and great blue heron. The concentrations were also below LOEL for egg mortality for falcons, herring gulls and double crested cormorant. The study did not reveal any correlation between PCB exposure and vitamin A level, nor between PCB and thyroxin levels.

Common eider, kittiwake and glaucous gull from the Barents Sea area were sampled in 1991 (Savinova et al., 1995). Ranges of ∑PCB in fat and liver are presented in Table 6. The content of PCBs in fat from kittiwakes and glaucous gull approached or exceeded the NOEL for reproduction in black-crowned night heron. The content in liver for the same species were beyond LOEL for hatching success in white-leghorn chicken. In some individuals the levels were also beyond NOEL for hatching success, but exceeded NOEL for deformities in white-leghorn chicken. In common eider the range of ∑PCB in fat appeared to be between NOEL for deformities in white-leghorn chicken and LOEL for hatching success.

Table 6. Ranges of ∑PCB (ng/g ww) in fat and liver in common eider, kittiwake and glaucous gull sampled from the Barents Sea area in 1991 (Savinova et al., 1995).

	Range ∑PCB (ng/g ww)	
Species	**Content in fat**	**Content i liver**
Common eider	105 - 730	3 -24
Kittiwake	22 - 11,607	119 - 729
Glaucous gull	12,875	398 - 1,843

Henriksen *et al.* (1996) collected female kittiwakes in a North-Norwegian colony. Mean and ranges of ∑PCB varied between organ and tissues (Table 7.) The levels found in livers of kittiwakes were all below NOEL for reproduction in black-crowned night heron, common tern and great blue heron. The leves were also below LOEL for egg mortality and egg production in falcons and herring gulls.

Table 7. Total liver concentrations of ∑PCB (ng/g ww) in different organs or tissues in female kittiwakes from Northern Norway (Henriksen et al., 1996).

Organ/tissue	Range concentration (ng/g ww)	Mean ± SD
Liver	12 - 285	85 ± 80
Brain	21 - 337	103 ± 77
Fat	10 -21	14 ± 3

In 1989, a number of glaucous gulls were found dead near a seabird cliff at south Svalbard (Gabrielsen *et al.*, 1995). In an effort to elucidate the cause of death, 12 individuals were sent for autopsy and analysis of chlorinated pesticides and PCBs in different organs. Results of the ∑PCB analysis are given in Table 8. The mean levels inliver, brain and kidney exceeded, or are just below the NOEL for reproduction in black crowned night heron. For some of the individuals the levels in the mentioned organs were very high compared to threshold values of ∑PCB for effects in birds (Table 5).

Table 8. Concentrations of ∑PCB (ng/g ww) in different organs of glaucous gulls (Gabrielsen et al., 1995).

	∑PCBs (ng/g ww)		
	Fat %	**Range**	**Mean ± SD**
Liver	0.4 ± 0.5	800 - 32,300	16,000 ± 10,000
Brain	7.1 ± 0.8	900 - 29,500	15,000 ± 10,000
Kidney	1.5 ± 1	400 - 21,400	10,000 ± 8,000
Muscle	0.6 ± 0.7	500 - 6,000	3,000 ± 2,000

If the objective is to have a numerical value for a *critical load* for POPs in the arctic environment, or any environment as well, we are, as far as we can see, far from having such a number. According to the definition of critical load the task seems very difficult, at least based on present knowledge.

Concerning calculations of critical load for an ecosystem, one of the main problems is all the different components which are involved, that is different species in different trophic levels, affected by different factors and so on. Such a calculation have to give the yearly amount of e.g. PCBs deposited per area unit which will give a critical concentration. To accomplish this, one has to consider accumulation rates, biomagnification and metabolism of the pollutants, which are influenced by a number of factors. How is it possible to calculate how much more releases we might accept, in addition to the load which already exists in the environment, before the limits are reached? Some of the existing load will remain in the environment for decades, and must be considered in a calculation of a critical load.

The previous summary of some studies of levels and/or effects of organic pollutants in Arctic, shows high concentrations of PCBs in several arctic species, especially in polar bear, arctic fox, harbor porpoise and glaucous gull. These results suggest that we are at, or that we already might have passed the limit for what individuals can handle of persistent organic pollutants "without detrimental impact". To calculate a critical load for an ecosystem appeares impossible without a considerable increase in research effort. Possibly, modelling might be able to give a very uncertain estimate, provided input from deposition measurements and a programme for analysis of POP levels in the prey of susceptible species. We know too little about concrete effects in many species. We also know very little about interactions between different pollutants, and also how biotic and abiotic factors might influence the response to a pollutant.

Nevertheless, the referred results indicate that we are near the critical load in the Arctic. No matter how much we want to have quantitative expressions in relation to this pollution problem, an estimated critical load will be of limited value due to the many uncertainties connected to the calculations. Doing such uncertain calculations will not bring us any closer to a solution of the pollution problem. According to national policy, the releases should be reduced, and the problems with POPs will be

tried minimized according to the intentions from the last North-Sea Conference. No matter what critical load that might be calculated, this policy should be continued.

Recommendations

As a final conclusion, we can not see that the endeavour to define the *critical load* will be of a practical, near future value for environmental protection authorities in restricting the spreading of POPs. What would be of value is to strenghten the research effort regarding effect studies in susceptible species. That is, it seems necessary to provide more knowledge about the biochemical, physiological and immunological responses to persistent organic pollutants in a various of arctic species which are of special concern. Such an approach will have to include more experimental studies. In spite of a lot of differences between the field and the laboratory, this is the only way to get valueable information of what you might expect to find in the wildlife. It will also be of great value to strenghten the monitoring aiming at quantification of the load and levels in key media, particularly top predators and prey, and further research with relevance to the establishment of critical limits.

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List of species including *Latin name* which are mentioned in the report:

Polar bear	*Ursus maritimus*
Arctic fox	*Alopex lagopus*
Mink	*Mustela vison*
Otter	*Lutra lutra*
Narwhal	*Monodon monoceros*
Beluga whale	*Delphinapterus leucas*
Minke whale	*Balaenoptera acutorostrata*
Harbor seal	*Phoca vitulina*
Harbor porpoise	*Phocena phocena*
Ringed seal	*Phoca hispida*
Walrus	*Odobenus rosmarus*
Harp seal	*Phoca groenlandica*
Grey seal	*Halichoerus grypus*
Hooded seal	*Cystophora cristata*
Glaucous gull	*Larus hyperboreus*
Herring gull	*Larus argentatus*
Kittiwake	*Rissa tridactyla*
Black guillemot	*Cephus grylle*
Common guillemot	*Uria aalgea*
Common eider	*Somateria mollissima*
Double-crested cormorant	*Phalacrocorax aristotelis*
Black-crowned night heron	*Nycticorax nycticorax*
Great blue heron	*Ardea hierodias*
Peregrine falcon	*Falco peregrinus*
Common tern	*Sterna hirundo*
Forster`s tern	*Sterna forsteri*

ANNEX 6.

List of reports within the Norwegian research programmme on critical load ("Naturens tålegrenser")

Naturens Tålegrenser - Oversikt over utgitte rapporter

1 Nygaard, P. H., 1989. Forurensningers effekt på naturlig vegetasjon en litteraturstudie. Norsk institutt for skogforskning (NISK), Ås.

Uten nr. Jaworovski, Z., 1989. Pollution of the Norwegian Arctic: A review. Norsk polarinstitutt (NP), rapportserie nr. 55. Oslo.

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5 Pedersen, H. C. & Nybø, S. 1990. Effekter av langtransporterte forurensninger på terrestriske dyr i Norge. En statusrapport med vekt på SO2, NOx og tungmetaller. Norsk institutt for naturforskning (NINA), Utredning 005.

6 Frisvoll, A. A., 1990. Moseskader i skog i Sør-Norge. Norsk institutt for naturforskning (NINA), Oppdragsmelding 018.

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10 Pedersen, U. 1990. Ozonkonsentrasjoner i Norge. Norsk institutt for luftforskning (NILU), OR 28/90.

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11b Reuss, J. O., 1990. Critical loads for soils in Norway. Analysis of soils data from eight Norwegian catchments. Norsk institutt for vannforskning (NIVA), O-89153.

12 Amundsen, C. E., 1990. Bufferprosent som parameter for kartlegging av forsuringsfølsomhet i naturlig jord. Universitetet i Trondheim, AVH (stensil).

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14 Frisvoll, A.A., & Flatberg, K.I., 1990. Moseskader i Sør-Varanger. Norsk institutt for naturforskning (NINA) , Oppdragsmelding 55.

15 Flatberg, K.I., Bakken, S., Frisvoll, A.A., & Odasz, A.M. 1990. Moser og luftforurensninger. Norsk institutt for naturforskning (NINA) , Oppdragsmelding 69.

16 Mortensen, L.M. 1991. Ozonforurensning og effekter på vegetasjonen i Norge. Norsk landbruksforsk. 5:235-264.

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32 Amundsen, C.E. 1992. Sammenligning av relativ forsuringsfølsomhet med tålegrenser beregnet med modeller, i jord. Norges geologiske undersøkelse. NGU-rapport 92.294.

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