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**Proceedings  
of AMAP Workshop on  
Sources, Emissions and  
Discharges  
Kjeller, 23 - 24 August 2001**

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## Summary

This document provides information on sources, emissions and discharges of contaminants measured in the Arctic environment. This information was obtained at the AMAP Workshop on Sources, Emissions, and Discharges, organized at the Norwegian Institute for Air Research, in Kjeller, Norway from 23 through 24 August, 2001. The purpose of the Workshop was to contribute to the assessment of current and future sources and emissions of chemicals worldwide. Another objective was to review the application of knowledge of emissions and sources of heavy metals in modelling their transport within air masses and water currents to the Arctic region. It can be concluded that sources in Asia may have an important influence on the concentrations of heavy metals in various environmental compartments in the Arctic. The atmospheric transport seems to be the main pathway for Hg entering the Arctic region. The opposite is true for Zn. For Pb and Cd the atmospheric transport and the riverine pathway are equally important. However, one should be cautioned that the above mentioned conclusions need further verification.



# **Proceedings of AMAP Workshop on Sources, Emissions and Discharges**

**Kjeller, 23 - 24 August 2001**

## **1 Introduction**

The need for "estimates and regular reporting by the Arctic countries of contaminant emissions and discharges, including accidental discharges, as well as transport and deposition" has been identified by the 1991 Rovaniemi Ministerial meeting as a priority action for AMAP. Arctic countries were requested to "consider the feasibility of developing national inventories on the production, use, and emissions of persistent organic contaminants (e.g. pesticides) to be collected and made available and summarized in the state of the Arctic environment reports. There were also other approaches recommended for a collection of information on emissions of POPs and other pollutants studied within the AMAP, including: 1) the collection of emission from other international organizations, conventions, and 2) joint research projects.

The above mentioned approaches have contributed to the improvement of our knowledge about sources, source regions, and emissions of pollutants within and outside the Arctic region. However, the following has been concluded in the framework of the AMAP future work, prepared on the basis of the requests of the Alta Ministerial Conference and presented in its Ministerial Declaration concerning source inventories: "In spite of the fact that during Phase 1 the participating countries carried out substantial work on inventories of pollution sources which may impact the Arctic region, the AMAP Assessment has shown that the information available is not yet sufficient. During the next phase, this work should be continued".

The issue of how to perform the assessment of sources and source emissions and discharges during the AMAP II Assessment was further discussed at the AMAP Assessment Steering Group meeting in Reykjavik, Iceland in November 2000. It was agreed that the information on this subject should be covered in each assessment chapter as opposed to producing a separate source and emissions chapter. However, the need for a special workshop on sources and emissions was identified to compile relevant information as input to the various assessments. The Center for Ecological Economics at the Norwegian Institute for Air Research (NILU) agreed to organize such workshop in Kjeller, Norway during the period from 23 through 24 August, 2001.

## 2 Purpose of the Workshop

The purpose of the workshop was to contribute to the assessment of current and future sources and emissions of chemicals measured in the Arctic environment. A Working Document was prepared and distributed prior to the Workshop in order to help discussing the latest information on quantities of emissions from various sources and source regions. This document was revised according to the outcome of discussions at the Workshop. The revised version of the Working Document is attached in Appendix A.

Another objective of the Workshop was to review the application of knowledge of emissions and sources of heavy metals worldwide in modeling of pollution transport to the Arctic region with air masses and water currents.

## 3 Structure of the Workshop

A number of presentations have been given during the two first sessions of the Workshop on sources and emissions, and modeling activities, respectively.

*Jozef M. Pacyna* presented an assessment of the 1995 global emissions of heavy metals from anthropogenic sources to the atmosphere. It was indicated that stationary fossil fuel combustion continues to be the major source of Cr, Hg, Mn, Sb, Se, Sn, and Tl with respect to the coal combustion and the major source of Ni and V with respect to oil combustion. Combustion of leaded, low-leaded and unleaded gasoline continues to be the major source of atmospheric Pb emissions contributing about 74 % to the total anthropogenic emissions of this metal in 1995. The third major source of heavy metals is non-ferrous metal production, which is the largest source of atmospheric As, Cd, Cu, In and Zn. Emissions from sources in Asia are clearly the largest for all metals estimated in current assessments. This can be related to the increase of industrial production in this part of the world. It was also indicated that anthropogenic emissions of Pb and V are by one order of magnitude higher than the natural emissions of these metals on a global scale. Anthropogenic emissions are also a factor of 2 to 3 higher than the natural emissions for Cd and Ni, while they are comparable for Cu, Hg, Mo, Sb and Zn.

*Janina Fudala* presented a structure of emission inventorying in Poland with focus on emissions of Greenhouse gases, other gases, fine particles and organic and inorganic contaminants, including heavy metals and persistent organic pollutants. Current emissions, emission trends from the 1980 and emission projections for the year 2005 were discussed. While the emissions of Pb and Cd have decreased from 1988 through 1999, the Hg emissions are constant. Major source of Hg emissions in Poland is by far the combustion of coal to produce heat and electricity. These emissions are expected to decrease by the year 2005 due to the replacement of coal by other fuels, mainly natural gas and biogas.

*Sergey Kakareka* presented a paper on identification and estimation of sources of POPs in Belarus. Emissions of the following groups of pollutants were discussed: PAHs, dioxins and furans, and polychlorinated biphenyls. The following source categories were reviewed with respect to their contribution to the atmospheric

emissions of the studied pollutants: solid fuel combustion in residential furnaces, ferrous metal industry, open burning of municipal refuse and forest fires. Emission estimates were based on the emission factor approach. The largest emissions of PAHs were estimated for combustion of fuel in residential furnaces and open burning. Leakage from transformers and damaged capacitors were estimated as the main source of PCBs discharge to the Belorussian environment.

*Pieter van der Most* presented activities at the OECD ( a task force) in establishing the Pollutant Release and Transfer Register (PRTR) in various countries. This register can be regarded as a toll for developing environmental policies for the improvement of the quality of the environment. The task force established at the OECD in connection with the development of the PRTR system focuses its activities to create a compendium of release estimation techniques for point and diffuse sources, establish a clearinghouse for these activities, and establish an informal communication relay.

*Slava Gordeev* reviewed the information on heavy metal concentrations in water and suspended matter of main Russian Arctic rivers and estuaries. These concentrations in Ob, Lena, and Yenisey do not exceed the average concentrations on a global scale.

Preliminary assessment of heavy metal fluxes to the Arctic Ocean indicates that the outflow of these pollutants in particulate form is larger than their outflow in dissolved form.

*Lars Asplin* of the Institute of Marine Research (IMR), Norway, presented a paper on “*Modeling of pollutant transport in the marine environment*”. An overview of relevant activities at IMR within the Barents Sea region, was outlined. The focus was on the transport of water masses in the Barents Sea. The issue of validation of marine models was furthermore discussed. Finally some animation of the model predictions was presented.

*Jesper Christensen* of the National Environmental Research Institute (NERI), Denmark gave a presentation on “*Atmospheric modelling activities inside the Danish AMAP program*” about studies of the long-range transport of pollutants in the troposphere to the Arctic. A particular focus was on the various aspects of the Danish Eulerian Hemispheric Model (DEHM) System. This model was originally developed and applied for sulphur, but more recently, the DEHM-system has been further developed and applied for lead, photochemical reactions, and mercury. The future focus will be on improved parameterisations of Arctic chemistry, coupling with GOME measurements pluss a range of other activities.

*Alexey Gusev* of the Meteorological Synthesizing Centre – East (EMEP-MSC-East), Russia, focused on the “*Modeling of air pollutant transport to the Arctic*”. The heavy metals in focus were Hg, Cd and Pb. For POPs, the focus was on PCBs (7 congeners), PAHs (4 species), HCHs, HCB and PCDD/Fs (4 congeners). The hemispheric model which operates on a 2.5° x 2.5° grid system has been tested for Hg, PCBs amd HCHs. Examples for  $\alpha$ -HCH and lead was presented. A comparison with the regional MSC-E HM model was outlined. Model intercomparison campaigns for heavy metals have been undertaken and new ones

are planned. Finally, an analysis of the model uncertainties showed that the most uncertain parameter in the model is the emission dataset.

*Marc Cohen* of the National Oceanic & Atmospheric Administration Air Research Laboratory, USA, gave a presentation on “*Source-Receptor Modeling in North America: Lessons learned from attempts to model the transport and deposition of atmospheric toxics to the Great Lakes*”. In this presentation, selected model results were shown where the impact of various sources of a certain dioxin congeners on the receptors within Great Lakes was highlighted. The results were discussed in terms of approximate atmospheric half-lives and typical travel distances for various chemical species. Cohen emphasised the intrinsic uncertainty embedded in the emission data as the most uncertain input parameter of the whole modelling exercise, and discussed the current problems of emission inventories in general terms.

*Paul Bartlett* of the CBNS, Queens College, City University of New York informed about selected results from the “*Nunavut-project*”. In his presentation, dioxin deposition receptors in Nunavut were studied. Marine and terrestrial receptors were assessed, utilising the North American Dioxin Source Emission Inventories (Canada + USA + Mexico) as input.

*Kevin Jones* of the University of Lancaster, UK, gave an overview over recent studies related to the global fate of POPs. The focus was on the global behaviour of POPs in air and in soils and the role of these compartments in the transfer of POPs to the Arctic. The expected and observed behaviour of POPs in the atmosphere was discussed, along with a discussion around the role of soils as potential sources and sinks of these pollutants. To underpin the discussion, comprehensive monitoring data from around the globe were presented for soils, butter and atmospheric monitoring data.

*Peter Outridge* of Geological Survey of Canada presented a paper on “*Incidence of above normal blood Hg levels on First Nations people in Canada*”. The occurrence of mercury was studied from a receptor perspective. The role of modern versus pre-industrial levels of Hg in various biological matrices (human and seal hair, beluga tooth) was presented, and the accompanying techniques discussed (isotope techniques).

*JoLynn Carroll* from Akvaplan-NIVA, Norway gave the presentation on “*Fluxes and biogeochemical processes for contaminants in Northern Sea*”. Selected results from the project “*Transport and fate of contaminants in the northern seas*” were presented along with plans for the next phase of this project. A key focus was on geochemical models, needed for the study of sea-surface and sea-floor processes required in marine transport models.

*Srinivasan Venkatesh* of Environment Canada Meteorological Service of Canada (MSC), presented the work of Dr. Yi-Fan Li: “*The beta-HCH story*”. The difference in observed and expected behaviour of beta-HCH in the Arctic was discussed in comparison with previous studies for alfa-HCH. Building upon a recent global source inventory for beta-HCH, the possible pathways of this chemical to the Arctic were discussed. The beta-HCH isomer was shown to have a

relative low concentration in Arctic air, but high concentrations in surface waters. This difference in occurrence and behaviour was attributed to differences in key physical-chemical properties of the two HCH isomers, whereby the beta-HCH isomer is more water-soluble than the alfa-HCH isomer. It was thus anticipated that the ocean current pathway was relatively more important in transporting beta-HCH to the Arctic, and it was hypothesised that the Bering Strait act as a bottleneck for this transfer.

*Benoit Fribourg-Blanc* of OIE/IOW (Office International de l'Eau/International Office for Water) gave a presentation about relevant activities within the European Environment Agency, EEA. These activities are carried out under the European Topic Centers on Waters, a consortium of 10 organisations led by the Water research Centre in the UK. A particular emphasis was on EUROWATERNET, which contains European information on water sources, including discharges to water.

Extended summaries of these presentations are included in Appendix B.

Main discussion on the sources, emissions, discharges, and modeling has been organized in two parallel groups within Session 3. The outcome of these discussions is presented below.

Finally, the meeting agenda is presented in Appendix C, while a list of participants in the Workshop is enclosed in Appendix D.

## 4 The outcome of the Workshop discussions

### 4.1 Discussion in Group A

|                    |   |
|--------------------|---|
| <i>Chair:</i>      | Jozef M. Pacyna   |
| <i>Rapporteur:</i> | Knut Breivik  |
| <i>Members:</i>    | Paul Bartlett, USA<br>Annmari Blom, Sweden<br>Knut Breivik, NILU, Norway<br>Christensen, Jesper, Denmark<br>Fribourg-Blanc, Benoit, France<br>Janina Fudala, Poland<br>Vyacheslav V. Gordeev, Russia<br>Kevin C. Jones, United Kingdom<br>Vitaly Kimstach, AMAP<br>Susanne K.M. Marcy, USA<br>Peter Outridge, Canada<br>Jozef M. Pacyna, NILU, Norway<br>Elisabeth G. Pacyna, NILU, Norway<br>Marina Varygina, Russia<br>Frits Steenhuisen, The Netherlands |

#### 4.1.1 General matters

As a starting point, it was noted that a clarification of source terms is needed, both for the further discussion as well as for the improvement of the Working Document prepared for the Workshop. Specifically, it was agreed that the term *emissions* should be used when discussing atmospheric emissions. Secondly, *discharges* should be used when chemical releases are occurring to water as receptor of the pollutants. Thirdly, that *releases* should only be used when addressing the total environmental releases of pollutants, irrespective of the receiving media.

Another key issue that was raised, has dealt with criteria that comprise a “good” inventory. It was recognised that several criteria are used to define the quality of an inventory. Typically a good inventory is defined as being transparent, comparable and accurate. It was suggested that the 2<sup>nd</sup> AMAP assessment should give an overview and elaborate on the details about the quality of inventories.

It was stressed that the Working Document should be presented in a form of synthesis, aiming at the integration of data representing the current status of knowledge. Besides, it should offer some ideas about the key gaps in knowledge.

The importance of relevant activities within other conventions and programs was mentioned. The Working Document should mention these activities and their relevance for the 2<sup>nd</sup> AMAP assessment.

Finally, it was felt during the discussion and in light of the presentations at the workshop, that it is important to bring out the message that source inventories are

very often the key uncertainty in model predictions. Therefore, improved inventories are considered as a key priority to improve model predictions.

#### **4.1.2 POPs**

It was recognised that there are more data of relevance available for North America than what is presented in the present version of the Working Document. More information from North America should therefore be included. Paul Bartlett may contribute on this particular issue.

The AMAP workshop in Bergen, 1999, made several recommendations related to source inventories. It was suggested that the responsible scientists of each key task assigned at that Workshop may be contacted to provide an update about recent activities related to those recommendations.

The transfer of POPs by rivers to the ocean was considered as a key gap in knowledge in relation to the overall environmental releases of these contaminants.

The data quality of source inventories for POPs is often very limited and efforts should be undertaken to assess and eventually improve the quality of such data.

It was concluded that data on the “new” POPs are scarce, and that more knowledge about the environmental releases are likely to be requested in the future.

It was also recognised that the quality of source-receptor model predictions is often affected by the poor quality of source and emission data. It was, however, pointed out that receptor-based modelling could serve as a policy oriented tool that may provide useful information in spite of the problems with source data.

It was recognised that there has been a progress in a number of related areas since the first AMAP assessment report. The most notable improvements are within the following subjects:

- source characterisation and source location,
- improvement in the estimation methodologies,
- several global datasets are becoming available, and
- substantial progress within various conventions, programs and organisations.

Key gaps in knowledge related to inventories for POPs were considered to be:

- anthropogenic emissions versus re-emissions,
- atmospheric pathway versus the ocean pathway,
- riverine fluxes and discharges,
- future scenarios of emissions, and
- source identification.

### **4.1.3 Heavy Metals**

#### **Hg**

Substantial progress has been made in Hg emission inventorying since the first AMAP assessment report on the sources of Hg. In particular, improved anthropogenic emission data has become available, through the involvement of authorities, research groups and industry.

The term “background concentrations” of Hg was discussed. This term shall represent the Hg concentrations due to past emissions from both natural and anthropogenic sources.

A number of gaps were still identified for emission inventorying of Hg, the most notable ones being:

natural emissions versus anthropogenic emissions,  
the contribution of gold mining to total Hg emission levels, and  
the role of discharges of Hg to the total environmental releases of the element.

A need for the following information was also identified:  
cumulative deposition maps of Hg for the assessment of Hg re-emission, and  
information on historical releases (peat and ice core studies were mentioned as a possible tool for this analysis).

#### **Lead**

It was concluded that Pb can be regarded as a successful case of policy implementation aiming at the reduction of contaminant releases to the environment.

It was pointed out that an update of the 1990 global emission inventory of atmospheric emissions of Pb shall be prepared. This survey shall then be used by modelers to assess the emission and concentration changes of Pb over the past few decades. Thus an assessment of chemical recovery of the environment can be approached, followed by an assessment of concentrations in biotic compartments of the environment.

#### **Cadmium**

It was underlined that no emission inventory has been prepared for Cd on a global scale during the last decade. This is a major gap in knowledge of Cd sources and their impact on the environment. A need for such global inventory was stressed.

## 4.2 Discussion in Group B

|                    |   |
|--------------------|---|
| <i>Chair:</i>      | Mark Cohen  |
| <i>Rapporteur:</i> | Simon Wilson  |
| <i>Members:</i>    | Lars Asplin, Norway<br>JoLynn Carroll, Norway<br>Mark Cohen, USA<br>Salve Dahle, Norway<br>Sergey Gromov, Russia<br>Alexey Gusev, Russia<br>Sergey Kakareka, Belarus<br>Jarle Klungsøyr, Norway<br>Astrid Olsson, Sweden<br>Pieter Van Der Most, The Netherlands<br>Srinivasan, Venkatesh, Canada<br>Simon Wilson, AMAP |

The following is a summary of the conclusions of the Group B discussions regarding the content of the Draft Working Document:

### 4.2.1 General

The document needs to be adjusted to provide a better balance between the Arctic vs. global perspectives. At present the emphasis of the document is on global emissions. Whilst these are of interest from the point of view of ‘contaminant transport into the Arctic’, the document also needs to address (and where possible quantify) emissions and discharges within the Arctic area (as defined by the AMAP boundary) as a regional subset of the global emissions/discharges.

Bearing in mind the information included in the presentations of Mark Cohen and Paul Bartlett regarding the relative importance of numerous small sources close to the Arctic in comparison with large but more distant sources, the document might also need to address this issue in more detail. This issue has potential implications for management and development of emission reduction policies.

Related to the above, model-runs should be performed for the ‘Arctic receptor area’, without inclusion of current emission inventories, in order to establish how far away from the Arctic relevant sources are located – for different contaminant. The resulting information could help focus effort on collection of data on sources relevant to Arctic contamination.

A better definition of ‘natural sources’ needs to be established and consistently used throughout the AMAP assessments. At present there is confusion about whether ‘natural sources’ include only true natural sources (volcanoes, forest fires, etc.) or whether they also include re-emissions of contaminants due to earlier anthropogenic emissions.

An important message to convey from the workshop and also include in the working document is that ‘information needed to derive emission/discharge inventories needs to be public’. The Aarhus process provides a possible way

around some of the current legal restrictions relating to public access to information. Reference should be made in the document to the situation regarding reporting requirements with respect to mandatory and voluntary list of substances covered by the UN ECE agreements.

#### **4.2.2 Emission Inventories**

The general lack of transparency concerning the methods used to construct emission inventories means that it can be difficult to evaluate them in terms of their completeness, accuracy, and usefulness, etc.

The concepts of 'quality' and 'uncertainty' need to be introduced in connection with emission inventories presented in the AMAP assessments. One possible approach would be to employ a 'rating/checklist' similar to that presented by Mark Cohen (attached). A systematic evaluation process is also being addressed under EMEP.

The AMAP workshop on Modelling and Sources (AMAP 1999<sup>1</sup>) presented a number of recommendations concerning emission inventory activities, included the following:

*Recommendation 31:*

**Develop approaches to estimating changes in emissions over time to account for changing levels of economic activity, changes in fuels and fuel use, developments in control technology, etc.**

There is a general need for up-to-date emissions inventories; current work tends to produce inventories with a time lag of 5-10 years relative to the current situation. For North America, a system has been developed to reprocess data and rapidly generate updated gridded emissions inventories. Similar systems need to be developed for inventories on a global scale.

The problem of more rapid updating of emission inventories, to provide 'currently relevant' inventories is still unresolved; it is potentially simple, or at least feasible, but is prevented largely by data access restrictions and issues associated with confidentiality of relevant information which has possible economic value.

The working document should stress the relative importance of emission inventories (in comparison with e.g. meteorological input data) in determining the uncertainty associated with output of long-range transport models. This should specifically refer to information presented at the workshop where a relevant sensitivity analysis had been conducted.

---

<sup>1</sup> AMAP 1999. Modelling and Sources: A Workshop on Techniques and Associated Uncertainties in Quantifying the Origin and Long-Range Transport of Contaminants to the Arctic. Report and extended abstracts of the workshop. Bergen, 14-16 June 1999. AMAP Report 99:4

### **4.2.3 Reporting**

The group agreed that the questionnaire circulated by AMAP to its member countries, to request information on their national reporting commitments under various international fora, yielded useful information for those responsible for preparing inventories of emissions/discharges. Considering the information provided by Canada, Finland and Sweden, the group encouraged AMAP to continue to try to obtain equivalent information from the other 5 Arctic countries that had not yet responded to the questionnaire.

The group agreed that, wherever possible, officially reported data should be used in compilation of inventories of emissions/discharges. However, it was also recognized that quality assurance of 'officially reported' data is often lacking. To a certain extent this is being addressed through use of agreed 'guidelines' and 'handbooks', etc., but there remains a need for increased comparison with scientifically based estimates and observations, etc. to confirm the officially reported data.

The lack of ranges in reported data was identified as a major deficiency – this is a problem in relation to data reported officially and from other sources.

In considering the relative merits of 'officially reported' emissions/discharge data vs. data derived from some sort of 'scientific expert judgment'; it should be remembered that most officially reported data is itself based on a process that involves 'expert judgment' at some levels or other during compilation and calculation of the final "official" estimates.

Problems relating to confidentiality and data access restriction were again raised in connection with data reporting.

The UN 'inventory of inventories' was mentioned.

### **4.2.4 POPs emission inventories**

The working document (and AMAP assessment) should stress the importance of collecting data now on production, distribution, usage, releases, etc., of substances such as current use pesticides and other 'new' POPs. This is to avoid the situation of having to reconstruct these data at some later point in time – as had to be done for substances such as DDT and HCH, etc. At present, because there is no requirement to do this, it is not being done. This issue should also be drawn to the attention of other relevant national and international fora.

The working document should better distinguish Local vs. Regional aspects; the emphasis in the draft document is on global inventories, but local sources such as accidents/spills should also be considered. A text on the relative importance of spills vs. chronic releases, etc. should be included.

Global inventories are needed for long-range transport modeling.

Better use should be made in the working document/AMAP assessments of environmental (atmospheric) half-lives, such as those presented to the workshop

by Mark Cohen, or transfer coefficients in order to identify the spatial extent of the inventories that are required for modeling transport to the Arctic.

In discussing the newly developed PCB inventories (Breivik et al.) there is a need to reflect the large uncertainties that still exist in relation to this work. The ACAP PCB project (*Multilateral Cooperative Project on Phase-out of PCB Use, and Management of PCB-contaminated Wastes in the Russian Federation – Phase 1*) carried-out by AMAP in Russia provides a good example of the usefulness of pilot studies for local sources, that in some cases can be generalized in considering larger sources or other sources.

The following overview of availability of information on sources of atmospheric emissions of POPs was constructed:

|                  | Global/regional  | Local   | Notes   |
|------------------|--|---|---|
| HCH              | Yes, spatially-distributed inventory available, described in AMAP phase 1                    |   | AMAP phase 2 assessment should update the budget model presented in AMAP phase 1 – model runs should be made to compute new budgets for Arctic reservoirs |
| PCBs             | Yes, spatially-distributed inventories of production and usage (but not emissions) available | Russia ( <i>Multilateral Cooperative Project on Phase-out of PCB Use, and Management of PCB-contaminated Wastes in the Russian Federation</i> ) |   |
| DDT              | Yes, Canadian project (Li et al.) expected to provide information by end-March 2002          |   |   |
| DDE              | ?  |   |   |
| Toxaphene        | ?  |   |   |
| Atrazine         | ?  |   |   |
| Dioxins          | Yes, North America<br>?, Europe  |   | Historical spatial trends; missing contribution from zinter production and pesticide production sources   |
| PAH              | Some for Europe  |   |   |
| Current use POPs |  |   | Information collection should be initiated now  |

#### 4.2.5 Heavy metals emission inventories

The following overview of availability of information on sources of atmospheric emissions of heavy metals was constructed:

|         | Global/regional   | Local | Notes  |
|---------|---|-------|--|
| Lead    | Yes, spatially-distributed inventory available for 1990 described in AMAP phase 1; updated inventory for 1995 available but not yet spatially-distributed |       |  |
| Mercury | Yes, spatially-distributed inventories available for 1990 (described in AMAP phase 1) and for 1995  |       | 1995 inventory is speciated (Hg <sup>0</sup> , Hg <sup>2+</sup> , Hg-P and Hg-Tot) and available for 3 emission height classes. Missing electrotechnical (lamp) sources and soil reservoir |
| Cd      | Not yet available   |       |  |
| Others  | ?   |       |  |

The participant from Belarus referred to information on a large soil reservoir source of Hg at a specific location in the FSU due to past local contamination.

#### 4.2.6 Aquatic discharges

Information on riverine inputs is available for Russian rivers for heavy metals (from V. Gordeev) and POPs (from A. Zhulidov/L. Alexeeva); the RAIPON/AMAP PTS project will also yield relevant data/information.

Information on riverine inputs for Europe is available from OSPAR, but only for a limited number of Arctic rivers.

Information for inputs from Canadian rivers should be available (for the Mackenzie and other rivers?). R. MacDonald should be able to provide this information.

Potential sources of information about inputs to the Arctic via other North American (i.e. US/Alaskan) rivers could not be identified by the members of the group.

Resolving net riverine inputs from gross riverine inputs remains an unresolved issue.

Some information is available on river ice transport of heavy metals (J. Hoelmen – concerning studies in the Lena)

Concerning contaminant fluxes with sediments incorporated in sea ice, the following text and associated references were suggested as an addition to the draft working document, to be added just after section 4: Riverine Inputs ...

#### ***4.2.7 Contaminant fluxes with sediments in ice***

Sediment-laden sea ice has been identified as an efficient mechanism for transporting contaminants throughout the Arctic Ocean (Dethleff et al., 1998, 2000; Pfirman et al., 1995; Weeks, 1994; Reimnitz et al., 1990). Contaminant-laden lithogenic materials are entrained into sea ice during the formation of frazil ice in turbid seawater (Rigor and Colony, 1997; Eidsvik, 1996). Contaminants are also deposit directly onto sea ice from the atmosphere. After entrainment, pollutants are transported by sea ice having a possible lifetime of 3-4 years and a potential net travel distance of thousands of kilometres (Rigor and Colony, 1997). Both the Laptev Sea (Pfirman et al., 1995; Dethleff et al., 1998) and the Kara Sea (Pfirman et al., 1997) have been identified as key regions of ice-formation that are important with respect to the supply of ice to adjacent seas and the central Arctic Basin. With the exception of radionuclides (Landa et al., 1997) however, estimates of contaminant incorporation and transport in sea ice are not generally available.

Through a Norwegian government sponsored program, fluxes from the Kara Sea of persistent organic pollutants in sea ice have been made using information on (1) satellite observations of sea ice movement, (2) model estimates of sediment uptake during sea ice formation, and (3) measured concentrations of pollutants in sea ice- or bottom- sediment from the Ob-Yenisey shelf (Carroll et al., 2000). The results were recently supplied to AMAP and will be used further to conduct a comparison with other sources and transport mechanisms controlling contaminant transfer from land to sea (i.e. ocean currents, river fluxes, sea ice).

| <b>Criteria for evaluating air emissions inventories in relation to their potential use as inputs to atmospheric transport models (source M. Cohen)</b> |   |
|---|---|
| <b>1</b>  | <p><b>Inventory should be comprehensive, i.e. all potential source classes should be included.</b></p> <ul style="list-style-type: none"> <li>• Omissions in the inventory should be acknowledged.</li> </ul>   |
| <b>2</b>  | <p><b>Inventory should be complete in its treatment of each source class.</b></p> <ul style="list-style-type: none"> <li>• All emission sources for a given source class should be included; omissions should be acknowledged.</li> <li>• What is level of confidence? e.g., there may be more confidence in an inventory based on frequent, thorough inspections by trained regulators, etc.</li> </ul>  |
| <b>3</b>  | <b>Geographical resolution should be as high as possible.</b>   |
| <b>4</b>  | <b>Temporal resolution should be as high as possible.</b>   |
| <b>5</b>  | <p><b>Estimates should be based on actual measurements to the largest extent possible.</b></p> <ul style="list-style-type: none"> <li>• Statistical summaries and ranges of the measurements should be provided.</li> </ul>   |
| <b>6</b>  | <p><b>Documentation of the use of emission factors should be provided, and if they are used, adequate information should be provided about them.</b></p> <ul style="list-style-type: none"> <li>• Inventory should state when emission factors are used.</li> <li>• Basis of emission factors should be clearly described.</li> <li>• Uncertainties and ranges should be given.</li> <li>• Match between source and emission factor basis should be justified.</li> </ul> |
| <b>7</b>  | <p><b>Details on key characteristics of each source should be provided.</b></p> <ul style="list-style-type: none"> <li>• Throughput; air pollution control equipment.</li> <li>• Process information especially relevant to emissions.</li> </ul>   |
| <b>8</b>  | <p><b>Details about the emissions should be provided.</b></p> <ul style="list-style-type: none"> <li>• Form of pollutant in emission (vapor, particle, particle size).</li> <li>• Temperature, height, velocity of the emissions.</li> </ul>  |
| <b>9</b>  | <p><b>Data should be available in different useful forms.</b></p> <ul style="list-style-type: none"> <li>• Both summary-level and facility-level data are useful.</li> </ul>  |
| <b>10</b>   | <p><b>Inventory should be publicly available (including facility-specific details).</b></p> <ul style="list-style-type: none"> <li>• Public and independent researchers have a right to know.</li> <li>• Quality control/assurance: the more people that critically evaluate an inventory, the more likely that errors will be detected.</li> </ul>   |

## 5 Acknowledgements

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**Appendix A**  
**Working Document**



**Working Document on Sources and Emissions  
for the AMAP Workshop on Sources, Emissions, and Discharges  
at the Norwegian Institute for Air Research, Kjeller, Norway  
23-24 August, 2001**

prepared by  
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## **1. Introduction**

The need for "estimates and regular reporting by the Arctic countries of contaminant emissions and discharges, including accidental discharges, as well as transport and deposition" was identified as a priority action for AMAP already at the 1991 Rovaniemi Ministerial meeting. The Arctic countries were requested to "consider the feasibility of developing national inventories on the production, use, and emissions of persistent organic contaminants (e.g. pesticides)" to be collected and made available and summarized in the state of the Arctic environment reports. There were also other approaches recommended for the collection of information on emissions of POPs and other pollutants studied within the AMAP, including:

- the collection of emission information from other international organizations, conventions, etc., and
- initiation and implementation of joint research projects concerning sources, emissions and discharges.

Although progress has been made in provision and compilation of information on sources, emissions and discharges, much still remains to be done to improve national reporting and delivery of comparable data for source regions both within and outside the Arctic. With reference to AMAP's future work, the Alta Ministerial Conference (as reflected in the 1997 Alta Declaration) concluded the following concerning source inventories: "In spite of the fact that during [AMAP] phase 1 the participating countries carried out substantial work on inventories of pollution sources which may impact the Arctic region, the AMAP Assessment has shown that the information available is not yet sufficient. During the next phase [of AMAP], this work should be continued".

Major gaps in knowledge of sources and emissions of contaminants measured in the Arctic environment and recommendations on how to close these gaps have been defined during the AMAP Workshop on Modelling and Sources: Techniques and Associated Uncertainties in Quantifying the Origin and Long-Range Transport of Contaminants to the Arctic, organized in Bergen in June 1999 (AMAP, 1999).

The issue of how to perform an assessment of sources, emissions and discharges as part of the AMAP Phase II Assessment process was then discussed at the AMAP Assessment Steering Group (ASG) meeting in Reykjavik, Iceland in November 2000. It was agreed that, rather than producing a separate report on

sources and emissions/discharges, information on these subjects should be covered in the various topic related assessments concerning POPs, heavy metals, radioactivity, acidification, etc. However, the ASG also identified the need for a special workshop on sources and emissions in order to bring together experts in these fields to compile relevant information as input to the various topic related assessments. The Center for Ecological Economics at the Norwegian Institute for Air Research (NILU) agreed to organize this workshop in Kjeller, Norway, from 23 through 24 August, 2001.

The current Workshop is aiming at providing the basis for the update and completion of information on sources and emissions and discharges of contaminants to the air and aquatic ecosystem on a global and regional scale, presented in the First AMAP assessment. Thus, the information presented at the Workshop will be used during the Second AMAP assessment, to be completed in 2002.

The following subjects will be discussed during the Workshop:

- updating and completing the inventories of atmospheric emissions and discharges to the aquatic and terrestrial ecosystems that were prepared during the first phase of AMAP assessment for selected heavy metals, POPs, and acidifying substances,
- assessment of emissions of atmospheric contaminants from natural sources that affect the Arctic, particularly for mercury, cadmium, sulfur, and nitrogen components,
- assessment of historical trends of anthropogenic emissions of pollutants to the atmosphere, including source estimates and maps/datasets of spatial distributions, e.g., within the IGBP Global Emission Inventories Activity (GEIA) grid system of 1° by 1°,
- assessment of current and past fluxes of pollutants to the Arctic with suspended solids and waters of the main Arctic rivers, including the Yenisey, Ob, Lena, and Mackenzie (this subject was identified in the AMAP first phase assessment as a significant gap in knowledge),
- assessment of contaminant fluxes with ocean currents and ice (this subject was also identified in the AMAP first phase assessment as a significant gap in knowledge),
- elaboration of a list of hot-spots of current and past emissions/discharges of contaminants that are transported to the Arctic, including detailed information on geographical locations of the sources and quantification of the emission/discharges of contaminants, and
- assessment of future scenarios for emissions/discharges of contaminants reaching the Arctic with the air masses, river waters, and ocean currents, for the years 2010, and 2020.

The purpose of this Working Document is to help discussion during the Workshop through the synthesis of information on sources and emissions of contaminants, particularly heavy metals and POPs, which appeared in the literature after the First AMAP assessment.

## 2. Sources and emissions of persistent organic pollutants (POPs)

Information on the global sources and emissions of persistent organic pollutants (POPs) is needed for the interpretation of contamination levels in remote areas. In particular, modelers need this information to derive source-receptor relationships for POPs to predict environmental concentrations in the Arctic. Furthermore, knowledge about the environmental releases of POPs within individual source categories is needed by policy-makers for the development of strategies for further emission reduction. While major international efforts have been made to improve inventories needed for research on ozone depletion (CFCs), climate change (CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O) and acid deposition (SO<sub>2</sub>, NO<sub>x</sub>), inventories for many organic toxic compounds were less in a focus and considered less reliable and accurate (e.g. Graedel et al. 1993, Pacyna and Graedel, 1995). Improvements in source inventories are considered to be a major research need in the context of environmental pollution and fate of POPs (e.g. Vallack et al. 1998; Jones and de Voogt, 1999). The relative poor quality of most of the current data on POP emissions is obviously mitigating the confidence in model outputs, e.g. the reliability concerning the impact of current or altered emissions on environmental levels within the Arctic (e.g. AMAP, 1999).

When addressing the issue of releases of POPs to the environment, the consequences of major accidents or the problems of local and national pollution are often in focus. Although these are significant problems, the long-term chronic contamination of the environment may be considered to be of equal importance (Schwarzenbach et al. 1993). However, the above implies that much effort have been devoted to address the emissions of POPs at a smaller spatial scale. As a result, most current emission inventories are often incomplete in terms of spatial coverage in order to be a useful tool to study Arctic source-receptor relationships. Secondly, emission inventories should ideally be established as historical time trends, reflecting the potential environmental lifetime of the pollutant in question. For many long-lived POPs, such as PCBs and DDT, this means that inventories ideally should cover several decades to interpret current environmental levels. From this it should be clear that reliable estimates of the historical and current global emissions of POPs are very difficult and in some cases almost impossible to establish (Jones and de Voogt, 1999). In spite of these problems, useful information can still be gained from previous local, regional and global studies. The First AMAP assessment report gives an overview of sources of POPs in circumpolar countries (AMAP, 1998). Since the preparation and completion of the first assessment report, several regional and global studies have been completed.

From a source-perspective, POPs and related organic chemicals may in a simplified manner be classified as members of one or two principal source categories. Some are intentionally produced for one or several specific purposes, while others are formed accidentally. For the latter category, the emission sources in question are often industrial processes or various combustion derived emissions.

Within the group of deliberately produced chemicals, there are several subgroups for which these chemicals may be typically be assigned (solid vertical lines). Frequently, it is common to discriminate between the agro-chemicals or pesticides

(e.g. DDT, HCHs), solvent chemicals (e.g. Trichloroethene;  $C_2HCl_3$ ) and chemicals produced for a number of other applications. The other principal source category is the group of chemical compounds that are formed and released to the environment as unwanted by-products from various anthropogenic activities. Typically, the anthropogenic activities responsible for the major emissions, are activities within industries and various combustion processes. Examples are emissions of the Polychlorinated dibenzo-p-dioxins (PCDDs) and the polychlorinated dibenzofurans (PCDFs) and the Polyaromatic Hydrocarbons (PAHs).

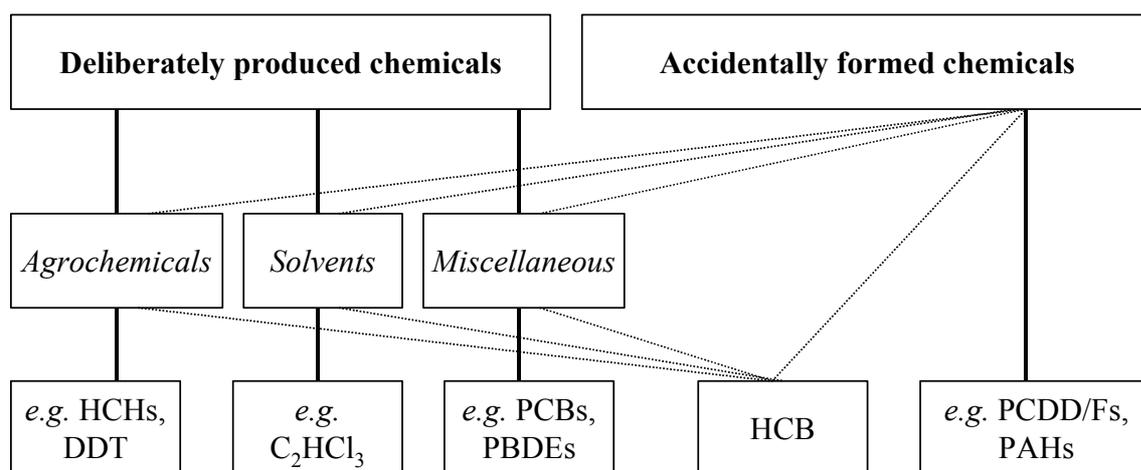


Figure 1: Simplified classification for some important source categories of POPs and related organic chemicals of environmental concern.

For some of these chemicals, there is no overwhelming single source category. One example is Hexachlorobenzene (HCB) that is both deliberately produced and unintentionally formed, as indicated with vertical lines in Figure 1 (see Bailey, 2001). It should also be noted that additional emission pathways are known, which are not indicated in Figure 1. For example, PCBs may be accidentally formed by *de novo* synthesis within various combustion processes (e.g. Brown et al. 1995).

## 2.1 European studies

Considering the current status of European emission data for POPs in general, it is quite clear that progress has been made over the last few years. Emission data for POPs are among other being collected as part of the EMEP programme (Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe). Still, very few Parties are able to submit official emission data for POPs to EMEP (Mylona, 1999; Vestreng and Støren, 2000). So-called expert estimates should not substitute national reported data. However, as long as the proper national bodies are not able to fulfil their obligations to report the national emissions to EMEP, expert estimates may play an important role for source-receptor modellers and for other research purposes. One important example is the European emission inventory of heavy metals and POPs for 1990 that has gained substantial interest (Berdowski et al. 1997). This

study was prepared on the basis of submissions of emission data from the Parties to the OSPARCOM, HELCOM and Convention on LRTAP. For the countries, sources and compounds that were lacking in official submitted data, default emission estimates were prepared to make the inventory complete. One of the major strengths of this study is thus the wide coverage of different priority substances as identified by OSPARCOM and HELCOM. This inventory covers a wide range of deliberately produced and accidentally formed POPs and related organic chemicals, including PAHs, PCBs, PCDD/Fs, HCB, several pesticides and solvent chemicals. Another European emission inventory was recently presented by Pacyna et al. (1999). This study focused on a subset of POPs, notably the PCBs, PCDD/Fs, HCHs, HCB, DDT and Benzo[*a*]pyrene (B[*a*]P). One objective with this study was to estimate the historical trend in European emissions from 1970 to 1995 and to facilitate an analysis of the environmental response to changes in emissions. As individual constituents within groups of compounds (e.g. PCDD/Fs, PCBs, HCHs) may behave quite differently in the environment, a particular emphasis was on the emissions of individual isomers and congeners (Pacyna et al. 1999; Breivik et al. 1999). Similar research efforts have been undertaken at the national level to estimate emissions of other POP constituents. In U.K. there have been studies on the national emissions for selected PCBs (Harrad et al. 1994) and selected PCDD/Fs (Alcock et al. 2001).

## 2.2 Global studies

The best possibilities to derive with consistent and reliable source and emission estimates are for the deliberately produced POPs. Several databases have been established for the global production and usage of such compounds.

Table 1 presents selected values compiled in a recent review (MacDonald et al. 2000) along with updated figures for PCBs. Among the insecticides presented below, most information is available for HCHs (technical HCH and lindane). The global estimates are described in a series of publications (Voldner and Li, 1995; Li et al. 1996; Lie et al. 1998; Li, 1999*a,b*) and recently utilised to model the global distribution and fate of  $\alpha$ -HCH (Wania et al. 1999; Wania and Mackay, 1999).

Table 1: Estimates of the global historical usage or production of selected deliberately produced POPs (modified after MacDonald et al. 2000).

|               | Use           | Period of time | Estimated total global usage / production (Mt) | Reference            |
|---------------|---------------|----------------|--|----------------------|
| DDT           | Insecticide   | 1950-1992      | 2.6  | Voldner and Li, 1995 |
|               |               | 1970-1992      | 0.99   | Voldner and Li, 1995 |
| Toxaphene     | Insecticide   | 1950-1992      | 1.33   | Voldner and Li, 1993 |
|               |               | 1970-1992      | 0.67   | Voldner and Li, 1993 |
| Lindane       | Insecticide   | 1950-1992      | 0.72   | Voldner and Li, 1995 |
| Technical HCH | Insecticide   | 1948-1997      | 10.0   | Li, 1999 a)          |
| Chlordane     | Insecticide   | 1945-1988      | 0.078  | Barrie et al. 1992   |
| Aldrin        | Insecticide   | 1950-1992      | 0.50   | Barrie et al. 1992   |
| Dieldrin      | Insecticide   | 1950-1992      | 0.034  | Barrie et al. 1992   |
| Endosulfan    | Insecticide   | 1956-1992      | 0.057  | Barrie et al. 1992   |
| Total PCB     | Miscellaneous | 1930-1993      | 1.32   | Brevik et al. 2001a  |
| PCB-28        |               | 1930-1993      | 0.057  | Brevik et al. 2001a  |
| PCB-52        |               | 1930-1993      | 0.038  | Brevik et al. 2001a  |
| PCB-101       |               | 1930-1993      | 0.031  | Brevik et al. 2001a  |
| PCB-138       |               | 1930-1993      | 0.025  | Brevik et al. 2001a  |
| PCB-153       |               | 1930-1993      | 0.027  | Brevik et al. 2001a  |
| PCB-180       |               | 1930-1993      | 0.014  | Brevik et al. 2001a  |

A study on the global historical production, consumption and atmospheric emissions of selected PCBs was recently completed. The first part of this study presents the methodology, principal uncertainties and selected results from an inventory, aiming to quantify the global production and consumption of total PCBs as well as 22 PCB congeners (Brevik et al, 2001a). Available data on the historical production of PCBs and the chemical composition of various technical mixtures was compiled from the literature, taking into account the most recent information from the Russian Federation (AMAP, 2000). Production of individual homologues and congeners is addressed by collecting data from the literature on total PCB production as well as on various technical PCB mixtures. Secondly, these data were coupled with the chemical composition of these technical mixtures to estimate the production of individual homologues and congeners. Whenever there were gaps in the data, assumptions were made to come up with quantitative estimates for the homologue and congener production. Information on imports, exports, consumption, as well as restrictions on production and imports, was further compiled for individual countries. These data, along with assumptions on the trade between countries and regions, have been utilised to derive an estimate of the global historical consumption pattern. Although there are substantial uncertainties involved in these estimates, important aspects governing the large scale temporal and spatial patterns are most likely captured. The availability of information for major producing companies and consuming countries around the time of their peak production indicates that the recent data are more accurate than the data from the past.

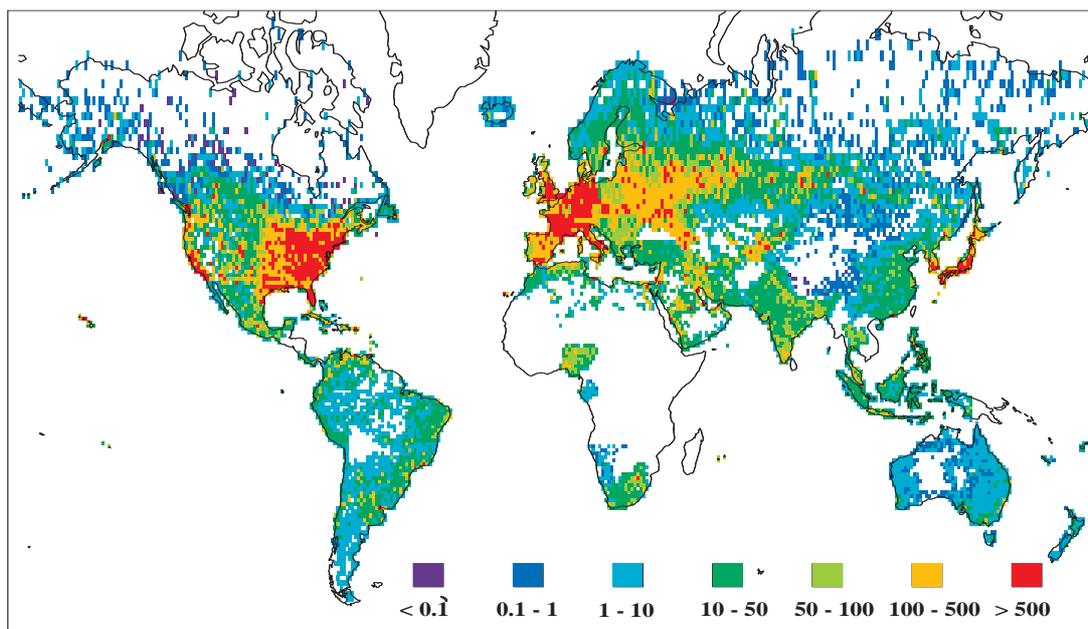


Figure 2: Estimated cumulative global usage of PCBs (legends in tonnes) with  $1^\circ \times 1^\circ$  longitude and latitude resolution.

The estimates account for a reported historical global production of ~1.3 million tonnes PCBs for which more than 70% have been estimated as Tri-CBs, Tetra-CBs and Penta-CBs. The results further suggest that almost 97% of the global historical use of PCBs may have occurred in the Northern Hemisphere. Figure 2 shows the estimated cumulative global consumption pattern for total PCBs (Breivik et al., 2001a).

In an accompanying study, an attempt has been made to estimate the historical emissions of the selected PCBs (Breivik et al., 2001b). It is shown that the diversity of historical usage, disposal and accidental release pathways makes it an extremely difficult task to bridge the gap between consumption and emissions, resulting in an equally complex and diverse true emission pattern. In spite of these uncertainties, the study is considered as a first important step towards the establishment of a global PCB emission inventory with congener resolution. The importance of temperature as a key parameter in controlling and affecting both the absolute value of PCB emissions as well as the PCB emission profile is emphasized. The results suggest that efforts should be directed towards mitigating contaminated materials to be subject to extreme temperatures under uncontrolled conditions, and to better characterize and quantify these potentially important release pathways.

Recently, a global emission estimate for HCB was presented (Bailey, 2001). This study presents a quantitative estimate of the global HCB emissions representative for the mid 1990s. The results suggest that the total current emissions could be around 23,000 kg/yr with a possible range between 12,000 and 92,000 kg/yr (Bailey, 2001). The author suggests that a substantial portion of the HCB measured in the atmosphere can be explained by unidentified sources and by re-volatilization.

For the other POPs, few studies are available which aim preparation of the global emissions. Rough estimates of the global emissions of PCDD/Fs are available from Brzuzy and Hites (1996). In addition, an overview of the current status of national and regional emission inventories for PCDD/Fs globally can be found in UNEP (1999).

### **3. Sources and emissions of heavy metals (HMs)**

An accurate and complete emission inventory for atmospheric trace metals on a global scale is needed for both modeler community and policy makers in order to assess the current level of environmental contamination by these pollutants, major emission sources and source regions, and the contribution of atmospheric pathway to the contamination of terrestrial and aquatic environment. International policy makers would need the information on global emissions in order to properly analyze source contribution to the pollution of a certain region, and then to propose proper abatement strategy. Modelers would need the emission survey to provide with the scientific substantiation material for these strategies through the application of global models of trace metal transport within air masses. Major progress has been made in assessing emissions of trace metals in various countries and even regions, e.g. Europe since the first global emission estimate for these pollutants was made by Nriagu and Pacyna (1988).

These improved national and regional emission inventories have been recently used to assess the global trace metal emissions from anthropogenic sources in the mid 1990's (Pacyna and Pacyna, 2001a). In the contract for the Ecotoxicity Advisory Panel (ETAP) of the International Lead and Zinc Research Organization (ILZRO), the International Copper Association (ICA), and the Nickel Producers Environmental Research Association (NiPERA) the authors have estimated the 1995 emissions of 16 HMs from anthropogenic sources worldwide. The results are presented in Table 2.

Table 2: *Worldwide emissions of trace metals from major anthropogenic sources to the atmosphere in 1995 (in tonnes/year)*

| Source Category                    | As            | Cd           | Cr            | Cu            | Hg           | In        | Mn            | Mo           | Ni            | Pb             | Sb           | Se           | Sn           | Tl           | V              | Zn             |
|------------------------------------|---------------|--------------|---------------|---------------|--------------|-----------|---------------|--------------|---------------|----------------|--------------|--------------|--------------|--------------|----------------|----------------|
| Stationary fossil fuel combustion  | 809           | 691          | 10 145        | 7 081         | 1 475        |           | 9 417         | 2 642        | 86 110        | 11 690         | 730          | 4 101        | 3 517        | 1 824        | 240 084        | 9 417          |
| Vehicular traffic                  |               |              |               |               |              |           |               |              |               | 88 739         |              |              |              |              |                |                |
| Non-ferrous metal production       | 3 457         | 2 171        | -             | 18 071        | 164          | 45        | 59            | -            | 8 878         | 14 815         | 552          | 466          | 319          | -            | 77             | 40 872         |
| Iron and steel production          | 353           | 64           | 2 825         | 142           | 29           | -         | 1 060         | -            | 36            | 2 926          | 7            | 7            | -            | -            | 71             | 2 118          |
| Cement production                  | 268           | 17           | 1 335         | -             | 133          | -         | -             | -            | 134           | 268            | -            | -            | -            | -            | -              | 2 670          |
| Waste disposal                     | 124           | 40           | 425           | 621           | 109          | -         | 511           | -            | 129           | 821            | 272          | 24           | 115          | -            | 23             | 1 933          |
| Other                              |               |              |               |               | 325          |           |               |              |               |                |              |              |              |              |                |                |
| <b>TOTAL</b>                       | <b>5 011</b>  | <b>2 983</b> | <b>14 730</b> | <b>25 915</b> | <b>2 235</b> | <b>45</b> | <b>11 047</b> | <b>2 642</b> | <b>95 287</b> | <b>119 259</b> | <b>1 561</b> | <b>4 601</b> | <b>3 951</b> | <b>1 824</b> | <b>240 255</b> | <b>57 010</b>  |
| <b>1983 emission <sup>*1</sup></b> | <b>18 820</b> | <b>7 570</b> | <b>30 480</b> | <b>35 370</b> | <b>3 560</b> | <b>25</b> | <b>38 270</b> | <b>3 270</b> | <b>55 650</b> | <b>332 350</b> | <b>3 510</b> | <b>3 510</b> | <b>3 790</b> | <b>5 140</b> | <b>86 000</b>  | <b>131 880</b> |

<sup>\*1</sup> Nriagu and Pacyna (1988)

<sup>\*2</sup> Emission of Hg from gold production

These results indicate that stationary fossil fuel combustion continues to be the major source of Cr (69 %), Hg (66 %), Mn (85 %), Sb (47 %), Se (89 %), Sn (89 %), and Tl (almost 100 %) with respect to the coal combustion and the major source of Ni (90 %) and V (almost 100 %) with respect to oil combustion.

Combustion of leaded, low-leaded and unleaded gasoline continues to be the major source of atmospheric Pb emissions contributing about 74 % to the total anthropogenic emissions of this metal in 1995.

The third major source of trace metals is non-ferrous metal production, which is the largest source of atmospheric As (69 %), Cd (73 %), Cu (70 %), In (100 %) and Zn (72 %).

It should be noted again that the information on the emissions of trace metals from waste incineration is clearly incomplete with respect to the number of countries reporting their emissions from this source and inaccurate with respect to the emission factors used to calculate trace metal emissions from waste incineration for countries not reporting their national emissions. In addition, statistical information on the amounts of municipal and sewage sludge wastes is also incomplete. The above mentioned problems result in underestimation of trace metal emissions from this source, particularly for As, Cd, Cr, Mn, Pb, Sb, Se, Sn, and Zn.

Emissions of trace metals released mostly during non-ferrous metal production have decreased by a factor of 2 to 3 between the beginning of the 1980's and the mid 1990's, mostly due to the improvement of emission control efficiency in major smelters in Europe and North America. One cannot rule out another explanation of this change, that the emission factors used to estimate emissions at the beginning of the 1980's were somewhat overestimated for some metals, as already discussed by Skeaff and Dubreuil (1997).

Emissions of trace metals emitted mostly from coal combustion are lower by a factor of 2 in the 1990's compared to the emissions at the beginning of the 1980's while emissions of V and Ni from oil combustion are higher by a factor of 2 to 3. This increase in emissions nicely resembles the factor of 2.2 increase in oil combustion to meet electricity and heat demands between the beginning of the 1980's and the mid 1990's. Indeed, the consumption of oil and the metal content of oil are the main factors affecting the amount of V and Ni emissions to the atmosphere from oil combustion in power plants. Most of the oil-firing power plants are not equipped with any dedusting installations. It can also be assumed that the content of Ni and V in crude oil has not changed significantly in the period of comparison. Thus, the main explanation of more than double increase of V and Ni emissions from oil-firing power plants would be increased production of electricity and heat worldwide.

Results of worldwide emission estimates of anthropogenic trace metals for various continents are presented in Table 3.

Table 3: Worldwide emissions of trace metals from major source categories to the atmosphere in 1995 on the continent by continent basis (in tonnes/year)

| Continent           | As           | Cd           | Cr            | Cu            | Hg           | In        | Mn            | Mo           | Ni            | Pb             | Sb           | Se           | Sn           | Tl           | V              | Zn            |
|---------------------|--------------|--------------|---------------|---------------|--------------|-----------|---------------|--------------|---------------|----------------|--------------|--------------|--------------|--------------|----------------|---------------|
| Europe              | 607          | 362          | 3 353         | 2 245         | 313          | 8         | 2 339         | 531          | 20 417        | 28 091         | 273          | 838          | 863          | 311          | 57 144         | 7 689         |
| Africa              | 324          | 172          | 847           | 2 031         | 389          | 2         | 738           | 207          | 10 690        | 11 349         | 66           | 300          | 425          | 78           | 28 930         | 2 353         |
| Asia                | 2 416        | 1 463        | 6 234         | 12 979        | 1 121        | 20        | 4 482         | 1 115        | 41 228        | 51 212         | 694          | 1 982        | 1 676        | 771          | 101 314        | 34 886        |
| North America       | 658          | 482          | 3 284         | 2 841         | 215          | 6         | 2 670         | 579          | 11 236        | 17 015         | 375          | 1 086        | 531          | 564          | 26 660         | 5 859         |
| South America       | 925          | 452          | 623           | 5 453         | 84           | 8         | 497           | 128          | 11 092        | 9 118          | 101          | 225          | 426          | 1            | 25 443         | 5 353         |
| Australia & Oceania | 81           | 52           | 389           | 366           | 113          | 1         | 321           | 82           | 624           | 2 474          | 52           | 170          | 30           | 99           | 764            | 870           |
| <b>TOTAL</b>        | <b>5 011</b> | <b>2 983</b> | <b>14 730</b> | <b>25 915</b> | <b>2 235</b> | <b>45</b> | <b>11 047</b> | <b>2 642</b> | <b>95 287</b> | <b>119 259</b> | <b>1 561</b> | <b>4 601</b> | <b>3 951</b> | <b>1 824</b> | <b>240 255</b> | <b>57 010</b> |

Emissions from sources in Asia are clearly the largest for all metals estimated in this work. This can be related to the increase of industrial production in this part of the world. However, progress in combating environmental pollution does not always follow industrial growth, particularly in the Asian countries.

Concerning the contribution of individual countries to the total emissions of anthropogenic trace metals from stationary fossil fuel combustion, the largest emissions have been estimated for Asia, accounting for 40 to 60 % of the total emissions depending on the metal of concern. The only exception is emission of Cd. The data provided by the US EPA indicate that the combustion of fuels in the United States alone contributed as much as 241.6 tonnes to the total global emissions in 1990 (US EPA 1993). It was assumed that similar emissions can be assigned as valid for the mid 1990's for the United States due to the similar amounts of coal and oil combustion in various years at the beginning of the 1990's. No major improvement of emission control technology in the US power plants was also assumed between 1990 and 1995. It should be admitted that the emission of Cd in the United States reported by the US EPA is unusually high, almost 2.5 times higher than in China, the second largest emitter country for this metal from fossil fuel combustion in stationary sources.

The Asian and South American countries are also the largest emitters of As and Cd from non-ferrous metal industry. High emissions are also estimated for the U.S. smelters.

Unfortunately, no data exist on the emissions of trace metals from individual countries or even continents for the beginning of the 1980's. Therefore, the development of emission geography between the beginning of the 1980's and the mid 1990's is impossible to review.

There are also no other data in the literature on global emissions of several trace metals, which could be compared with the estimates presented above.

The global emission inventory presented above is based on both emission estimates by national experts in several countries in Europe and North America, and author estimates for the remaining countries. National estimates are some times based on emission measurements (in non-ferrous metal smelters) which are considered a reliable source of information when preparing emission inventories. Indeed, more emission measurements should be carried out in order to further improve emission inventories. Emission measurements should be suggested for at least major point sources of emissions, such as power plants, smelters, waste incinerators, and cement kilns.

Most of the trace metals are emitted on fine particles, while a portion of Hg and Se in a gas phase. Entering the atmosphere on fine particles and in a gaseous form, trace metals can be transported with air masses on long distances, e.g. a couple of thousand kilometers up to the Arctic region. There is a fairly solid scientific evidence of such transport. It is very important to be able to conclude on to what extent concentrations measured in remote locations are of anthropogenic origin and what is the contribution of emissions from natural sources. Such information is crucial for preparing proper strategies on emission reduction with

respect to emissions from anthropogenic sources. The quality of this information is directly related to the accuracy and completeness of emission inventories for both anthropogenic and natural sources.

Concerning the emissions from natural sources, the information on the subject is quite poor. A new study on the estimates of trace metal emissions from natural sources is now being carried out at the Geological Survey of Canada (Robert Garrett, personal communication). The results of this study are expected in the near future. However, at least some comparison of the estimates of trace metal emissions from anthropogenic sources, presented above with the older estimates for natural sources available from the literature can be approached. An important contribution to describing the significance of natural sources of trace metals in the environment is that by the Geological Survey of Canada (GSC 1995), while the widest quantitative assessment of emissions from these sources is still the one by Nriagu (1989). Obviously, there are also other emission estimates, but they are mainly limited to a certain trace metal or to a single emission source category.

A comparison of global anthropogenic emission estimates in this work with the global natural emission estimates by Nriagu (1989) is presented in Table 4.

*Table 4: A comparison of estimated global anthropogenic emissions of trace metals in the mid 1990's with emissions from natural sources (in thousand tonnes/year)*

| <b>Trace metal</b> | <b>Anthropogenic emissions</b> | <b>Natural emissions: median values</b> | <b>Anthropogenic/national emission ratios</b> |
|--------------------|--------------------------------|---|---|
| As                 | 5.0                            | 12.0                                    | 0.42  |
| Cd                 | 3.0                            | 1.3                                     | 2.3   |
| Cr                 | 14.7                           | 44.0                                    | 0.33  |
| Cu                 | 25.9                           | 28.0                                    | 0.93  |
| Hg                 | 2.2                            | 2.5                                     | 0.88  |
| Mn                 | 11.0                           | 317.0                                   | 0.03  |
| Mo                 | 2.6                            | 3.0                                     | 0.87  |
| Ni                 | 95.3                           | 30.0                                    | 3.2   |
| Pb                 | 119.3                          | 12.0                                    | 9.9   |
| Sb                 | 1.6                            | 2.4                                     | 0.67  |
| Se                 | 4.6                            | 9.3                                     | 0.49  |
| V                  | 240.0                          | 28.0                                    | 8.6   |
| Zn                 | 57.0                           | 45.0                                    | 1.3   |

This comparison suggests that anthropogenic emissions of Pb and V are by one order of magnitude higher than the natural emissions of these metals on a global scale. Anthropogenic emissions are a factor of 2 to 3 higher than the natural emissions for Cd and Ni, while they are comparable for Cu, Hg, Mo, Sb

and Zn. The comparison in Table 4 also indicates that global natural emissions of As, Cr, and Se are larger than the global anthropogenic emissions of these elements by a factor of 2 to 3. Finally, the natural sources are by far more significant than the anthropogenic sources for Mn.

There are a few interesting observations which can be made on the basis of the comparison in Table 9. In general, there are no major surprises in this comparison for the mid 1990's compared to our previous findings with respect to the significance of anthropogenic sources of trace metals to the total budget of these contaminants. Perhaps the only exception is for As, often regarded as very much an anthropogenic trace metal. One should add that the As emission from natural sources considered in Table 4 is quite high and probably overestimated. For Hg, anthropogenic sources seems to be as important as natural sources. It should be underlined that the suggestions presented above are valid when we discuss the significance of anthropogenic versus natural sources on a global scale. This significance can be entirely different when discussing the situation on a local scale, e.g. around a waste incinerator or a volcano and even on regional scale, e.g. in Central Europe or in the Mediterranean Sea basin.

More detailed studies with respect to the spatial distribution of emissions and chemical forms of a given pollutant in emission plume have been carried out for mercury in connection with the Second AMAP assessment (Pacyna and Pacyna, 2001b).

Information on the chemical speciation of mercury emitted from various sources is crucial for the assessment of environmental and human health risks and effects, as well as description of mercury transport, fate, and behavior in the environment. Atmospheric and more general environmental chemistry of mercury is also very much dependent on the chemical and physical speciation of the element in during its emission.

For the first time global emissions of three different chemical species of mercury have been estimated: gaseous elemental mercury, gaseous bivalent mercury, and particulate mercury. These species are the most important species of Hg emitted from various sources. Emission profiles of these chemical species of mercury for various source categories are presented in Table 5.

Table 5: Emission profiles of Hg from anthropogenic sources, 1995.

| Emission profiles of Hg from Anthropogenic Sources , 1995 . |                 |                  |                |                   |                    |          |            |              |                |       |         |
|---|-----------------|------------------|----------------|-------------------|--------------------|----------|------------|--------------|----------------|-------|---------|
| SPECIE  | Coal Combustion |                  | Oil Combustion | Cement production | Non-Ferrous Metals |          | Pig & Iron | Caustic Soda | Waste Disposal | Other | Average |
|   | Power Plants    | Residential Heat |                |                   | Lead               | Zinc     |            |              |                |       |         |
| Hg° (gas)   | 0,5             | 0,5              | 0,5            | 0,8               | 0,8                | 0,8      | 0,8        | 0,7          | 0,2            | 0,8   | 0,64    |
| Hg2+  | 0,4             | 0,4              | 0,4            | 0,15              | 0,15               | 0,15     | 0,15       | 0,3          | 0,6            | 0,15  | 0,285   |
| Hg (particulate)  | 0,1             | 0,1              | 0,1            | 0,05              | 0,05               | 0,05     | 0,05       | 0            | 0,2            | 0,05  | 0,075   |
| Codes   | 11              |                  | 11             | 64                | 61                 | 61       | 50         | 32           | 14             |       |         |
|   | 11              |                  | 11             | 67                | 631, 632           | 641, 642 | 51, 54     | 83           | 16             |       |         |

The profiles were prepared on the basis of data on emissions of mercury in exhaust gases from various sources, collected by the authors for the last 2 decades. An interim report of this collection of data was prepared already at the beginning of the 1990's (Axenfeld et al., 1991). Major progress on the subject has been made only recently through the EU projects MOE (Munthe, 2000) and MAMCS (Pirrone, 2000).

The type of emission source is a very important parameter determining the chemical speciation and physical form of mercury because it indicates the chemical status of mercury in raw materials, e.g. in coal, ores, or wastes, as well as the industrial technology employing different temperatures in the source, e.g. in a coal-fired power plant or a waste incinerator. However, more detailed information is often needed on these industrial technologies, because the combustion temperature would differ in tradition combustion boiler compared to pulverized coal boiler. Fluidized bed combustion would employ different combustion temperatures compared to more traditional combustion mode.

Chemical composition of raw materials also affects the formation of various chemical species of mercury. For example, larger availability for chlorine in coal or wastes causes larger formation of gaseous bivalent mercury compared to elemental mercury. Higher concentrations of fine particles in exhaust gases would improve conditions for gas-to-particle conversion of gaseous mercury, thus larger emissions of particulate mercury.

The type of emission control devices, particularly different FGD techniques would result in various chemical forms of mercury.

The results of emission estimates for major source categories using the above mentioned profiles are presented in Table 6.

Table 6: Global emissions of gaseous elemental mercury (A), gaseous bivalent mercury (B) and particulate mercury (C) from major anthropogenic sources in 1995 (in tonnes). Emissions from "Other Sources" in Tables 2 and 3 are not included.

**A**

| Continent           | Stationary combustion | Non-ferrous metal production | Pig iron and steel production | Cement production | Waste disposal | Total          |
|---------------------|-----------------------|------------------------------|-------------------------------|-------------------|----------------|----------------|
| Europe              | 92.8                  | 12.3                         | 8.2                           | 20.9              | 2.4            | 136.6          |
| Africa              | 98.6                  | 6.3                          | 0.4                           | 4.2               |                | 109.5          |
| Asia                | 430.3                 | 69.9                         | 9.7                           | 65.4              | 6.5            | 581.8          |
| North America       | 52.4                  | 20.1                         | 3.7                           | 10.4              | 13.3           | 99.9           |
| South America       | 13.5                  | 20.3                         | 1.1                           | 4.5               |                | 39.4           |
| Australia & Oceania | 50.0                  | 3.5                          | 0.2                           | 0.7               |                | 54.4           |
| <b>TOTAL</b>        | <b>737.6</b>          | <b>132.4</b>                 | <b>23.3</b>                   | <b>106.1</b>      | <b>22.2</b>    | <b>1 021.6</b> |

**B**

| Continent           | Stationary combustion | Non-ferrous metal production | Pig iron and steel production | Cement production | Waste disposal | Total        |
|---------------------|-----------------------|------------------------------|-------------------------------|-------------------|----------------|--------------|
| Europe              | 74.2                  | 2.4                          | 1.6                           | 4.0               | 7.6            | 89.8         |
| Africa              | 78.8                  | 1.2                          | 0.1                           | 0.8               |                | 80.9         |
| Asia                | 344.1                 | 13.1                         | 1.8                           | 12.3              | 19.6           | 390.9        |
| North America       | 41.9                  | 3.8                          | 0.7                           | 1.9               | 39.5           | 87.8         |
| South America       | 10.7                  | 3.8                          | 0.2                           | 0.8               |                | 15.5         |
| Australia & Oceania | 39.9                  | 0.7                          | 0.1                           | 0.1               | 0.1            | 40.9         |
| <b>TOTAL</b>        | <b>589.6</b>          | <b>25.0</b>                  | <b>4.5</b>                    | <b>19.9</b>       | <b>66.8</b>    | <b>705.8</b> |

**C**

| Continent           | Stationary combustion | Non-ferrous metal production | Pig iron and steel production | Cement production | Waste production | Total        |
|---------------------|-----------------------|------------------------------|-------------------------------|-------------------|------------------|--------------|
| Europe              | 18.5                  | 0.7                          | 0.4                           | 1.3               | 2.4              | 23.3         |
| Africa              | 19.6                  | 0.4                          |                               | 0.2               |                  | 20.2         |
| Asia                | 86.0                  | 4.4                          | 0.6                           | 4.1               | 6.5              | 101.6        |
| North America       | 10.5                  | 1.2                          | 0.2                           | 0.6               | 13.3             | 25.8         |
| South America       | 2.7                   | 1.3                          | 0.1                           | 0.2               |                  | 4.3          |
| Australia & Oceania | 10.0                  | 0.2                          |                               |                   |                  | 10.2         |
| <b>TOTAL</b>        | <b>147.3</b>          | <b>8.2</b>                   | <b>1.3</b>                    | <b>6.4</b>        | <b>22.2</b>      | <b>185.4</b> |

The major chemical form of mercury emitted to the atmosphere is gaseous elemental mercury, contributing with about 53 % to the total emissions, followed by gaseous bivalent mercury with 37 %. The Hg emissions on particles contribute only about 10 % to the total emissions. Again, Asia contributes about 50 % to the total emissions of all individual chemical forms of mercury. The main source of these emissions is combustion of coal to produce electricity and heat, particularly in China. Chinese emissions from power plants and small industrial and commercial furnaces account for about 500 tonnes per year contributing more than 25 % to the total global emissions of this contaminant.

The 1995 global Hg emission data, including the total, gaseous elemental, gaseous bivalent and particulate mercury are now being spatially distributed within the 1° by 1° grid system, identical with the GEIA grid system. In this way emission maps will be generated and used by the modelers to assess the transport and atmospheric deposition of anthropogenic Hg in the Arctic region. Natural emissions of Hg will be also distributed spatially after completion of their assessment by the Geological Survey of Canada.

Global emissions of other trace metals shall also be spatially distributed for further application in models estimating the air concentrations and atmospheric deposition in the Arctic. The emissions of Cd and Pb shall be given priority in this activity.

#### **4. Riverine inputs of contaminants to the Arctic region.**

Information on riverine inputs of contaminants to the Arctic region is very limited. In addition to the data already presented in the First AMAP assessment, some information became available for Hg and other metal discharges with three major Arctic rivers in Russia: Lena, Yenisey and Ob. This information was obtained from the Joint Russian-French-Dutch Scientific Program on Arctic and Siberian Aquatic Systems (SPASIBA), briefly described in Kimstach et al (1998). It was concluded that Lena, Yenisey and Ob are less affected by anthropogenic impact compared to rivers in other regions of Russia. For example, the Hg concentrations in water in these rivers are between 0.8 and 5.4 nM/liter. These concentrations are significantly lower than the mean global values (Martin and Windom, 1991).

The Hg concentrations measured during the SPASIBA project, as well as during several national Russian projects have been reviewed by Gordeev and Tsirkunov (cited in Kimstach et al., 1998) in order to estimate riverine fluxes of dissolved and particulate mercury to the Arctic Ocean. The following results have been obtained:

- Ob: 0.2 tonnes Hg/ year dissolved and 0.8 tonnes Hg/ year in particulate form,
- Yenisey: 0.2 and 0.3, and
- Lena: 0.4 and 4.0.

Thus, about 5.9. tonnes of Hg enter the Arctic Ocean with these three Russian rivers annually.

More information became also available on the concentrations of other heavy metals in waters of the above mentioned rivers and the metal fluxes of these metals to the Eurasian Arctic in a dissolved form and with particles. The concentrations of various metals in the river waters are presented in Table 7 (after various authors), while metal fluxes are given in Table 8 (after Gordeev, 2001).

*Table 7: Mean concentrations of some heavy metals in waters of the biggest Arctic Russian Rivers (in µg/l), after revision by Gordeev (2001)*

| River   | Cu    | Zn     | Ni   | Pb     | Fe      |
|---------|-------|--------|------|--------|---------|
| Ob'     | 0-27  | 0-129  | -    | -      | 0-3120  |
|         | av. 3 | av. 16 | -    | -      | av. 560 |
|         | 6     | 2      | 1    | 2      | 650     |
|         | 2.3   | -      | 1.32 | 0.014  | 25.8    |
| Yenisey | 0-66  | 0-292  | -    | -      | 10-1520 |
|         | av. 8 | av. 36 | -    | -      | av. 170 |
|         | 6     | 35     | -    | 2      | 650     |
|         | 1.63  | -      | 0.53 | 0.0055 | 14.3    |
| Lena    | 0-11  | 0-159  | -    | -      | 0-820   |
|         | av. 2 | av. 12 | -    | -      | av. 120 |
|         | 4     | 12     | 1    | 1      | 210     |
|         | 0.60  | 0.35   | 0.30 | 0.016  | 22.9    |

*Table 8: Total river flux of dissolved and particulate heavy metals and iron to the Eurasian Arctic (in 10<sup>3</sup> tonnes/year) after Gordeev (2001).*

| River basin           | type of flux | Cu   | Pb    | Zn   | Ni   | Cd     | Hg     | Fe   |
|-----------------------|--------------|------|-------|------|------|--------|--------|------|
| Ob                    | diss.        | 0.85 | 0.006 | 0.16 | 0.53 | 0.0003 | 0.0002 | 12   |
|                       | part.        | 0.84 | 0.26  | 1.7  | 0.63 | 0.0033 | 0.0008 | 940  |
|                       | total        | 1.6  | 0.266 | 1.86 | 1.16 | 0.0036 | 0.001  | 952  |
| Yenisey               | diss.        | 1.0  | 0.004 | 0.82 | 0.34 | 0.001  | 0.0002 | 10   |
|                       | part.        | 0.65 | 0.18  | 1.3  | 0.45 | 0.0136 | 0.0003 | 320  |
|                       | total        | 1.65 | 0.184 | 2.12 | 0.79 | 0.0146 | 0.0005 | 330  |
| Lena                  | diss.        | 0.47 | 0.009 | 0.04 | 0.14 | 0.0032 | 0.0004 | 19   |
|                       | part.        | 0.49 | 0.40  | 2.50 | 0.55 | 0.0046 | 0.0040 | 590  |
|                       | total        | 0.96 | 0.41  | 2.54 | 0.69 | 0.0078 | 0.0044 | 610  |
| Whole Eurasian Arctic | diss.        | 4.04 | 0.036 | 2.18 | 1.93 | 0.0077 | 0.0015 | 64   |
|                       | part.        | 5.66 | 2.41  | 15.6 | 4.85 | 0.058  | 0.0088 | 5320 |
|                       | total        | 9.7  | 2.45  | 17.8 | 6.78 | 0.066  | 0.010  | 5384 |

*Table 9: Comparison of heavy metal fluxes to the Arctic region by air and three Russian rivers (in tonnes/year)*

| Pathway  | Hg  | Cd  | Pb    | Zn     |
|--|-----|-----|-------|--------|
| Atmospheric deposition (Akeredon et al., (1993)) | 40  | 47  | 2 400 | 1 350  |
| Riverine transport (Gordeev, 2001)               | 10  | 66  | 2 450 | 17 800 |
| Air Water  | 4.0 | 0.7 | 1.0   | 0.08   |

Recently a program on Transport and Fate of Contaminants in the Northern Seas was established in Norway with the support from the Norwegian Ministry of the Environment and the Norwegian Ministry of Foreign Affairs. Selected results were presented by Carroll (2001). This project will provide the information on:

- contaminants in the Pechora bay and adjacent areas,
- hydrological information for the Pechora and Ob-Yenisey river systems,
- contaminants in snow and ice for the Ob-Yenisey bays, and
- snow and ice distributions in the Ob-Yenisey system.

The final results from the above mentioned project will improve our knowledge on discharges of various chemicals with the Pechora, Ob, and Yenisey rivers to the Arctic region.

It is planned to use the above mentioned inventories for atmospheric emissions to estimate the atmospheric deposition of Hg and possibly other trace metals in the Arctic region and compare these inputs with the riverine flux. At present only some crude estimates can be made. Some time ago Akeredolu et al. (1993) have estimated the atmospheric deposition of selected heavy metals to the Arctic region from emissions in the Eurasian basin. This basin was defined as the region with emission contributing more than 90 % of the atmospheric deposition of heavy metals in the Arctic. These estimates can be compared with the estimates of the riverine input presented by Gordeev. The results of this comparison are given in Table 9. It can be concluded that the atmospheric transport is the main pathway for Hg entering the Arctic region. The opposite is true for Zn. For Pb and Cd the atmospheric transport and the riverine pathway are equally important. However, one should be cautioned that the above mentioned estimates need further verification.

## **5. Sources and emissions of contaminants in "polluted" Arctic**

Combustion of fossil fuels to produce electricity and heat, as well as several industrial activities, such as non-ferrous and ferrous metal production, fertilizer production, and gasoline consumption in the industrial and municipal centers in Northern Russia generate emissions of several pollutants to the atmosphere and discharges to the Northern rivers. Atmospheric emissions of sulfur dioxide and nitrogen oxides, a number of trace metals and selected POPs in the Kola Peninsula, the Norilsk area and the Northern Urals are of particular importance. The emission sources and emissions of trace metals and selected POPs have been included in discussion earlier in this document. They are included in global emission estimates for trace metals and POPs, presented in previous sections of this work.

Atmospheric emissions of acid rain causing pollutants, such as sulfur dioxide and nitrogen oxides in the above mentioned regions of Northern Russia have been discussed in the First AMAP assessment (AMAP, 1998). Rather limited information has been available in the literature about recent emission quantities of these pollutants. The Russian authorities reported to the UN ECE Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air

Pollutants in Europe that the total Russian sulfur dioxide emissions decrease each year since 1997 (the end of the First AMAP assessment) by 7-8 % (EMEP, 2000). It can be speculated that emissions from sources on the Kola Peninsula and in the Urals contribute to this decrease, but it is difficult to quantify this contribution without information from the Russian authorities.

Emissions of nitrogen oxides in Russia have not changed significantly from year to year during the last 5 years. Again, it is speculated that these emissions are quite unchanged in the Russian Arctic since the time of the First AMAP assessment. However, this would need to be documented during the Second AMAP assessment.

## **6. Emission scenarios**

At the present, there are no specific targets for the reduction of emissions of contaminants, their fluxes nor inputs to the Arctic environment within the AMAP. Therefore, no specific emission reduction scenarios have been developed within AMAP so far. However, enough information exists to start developing such scenarios using criteria accepted in other international programs/conventions aiming at the reduction of contamination load to the marine and terrestrial ecosystem through the atmosphere and river streams. These programs/conventions include: the OSPARCom Convention on the reduction of contaminant and nutrient loads to the North Sea, the HELCOM Convention on the reduction of these loads to the Baltic Sea and the UN ECE protocols on the reduction of HMs and POPs in the UN ECE area.

In general, it can be suggested that at least three types of scenarios may be developed for the change of the environment in the Arctic:

- Business-as-Usual (BAU) scenario, assuming that the contamination for the Arctic environment will continue at a current level of emission/discharge rates with a certain degree of modification of emission control installations and industrial/other anthropogenic processes,
- Policy Target (POT) scenario, assuming that the international agreements on emission/load reductions in the regions contributing to the inputs of chemicals to the Arctic environment will be fully or partly implemented, eg. agreements within the UN ECE, OSPARCom, HELCOM, and
- Deep Green (DEG) scenario, assuming that all controls available now and to be developed in the near future, will be implemented to reduce the inputs of contaminants and nutrients to the Arctic environment, even without major consideration of the cost of these controls.

The above mentioned scenarios can be approached for the Arctic environment until the year 2010, similarly to the scenarios to be developed within other international conventions/programs, including the UN ECE. Data sets needed for such scenarios are available at least for Pb and other heavy metals emitted on particles.

Scenarios for further development of Hg inputs to the Arctic can also be constructed, but they are more complicated than the scenarios for Pb and Cd due to variety of physical and chemical forms of Hg emitted from large number of sources, as well as due to re-emission of Hg from aquatic and terrestrial surfaces to the air.

Scenarios for POPs can also be approached but they would be with different degree of complexity. Scenarios for POPs which are by-products in various anthropogenic processes, can be developed in a similar manner as scenarios for Pb and Cd. The development of scenarios for POPs, such as pesticides is complicated by a fact that it is difficult to foresee their future substitutes at present.

## **7. Final remarks**

This Working Document is meant to be a report on activities aiming at the improvement of our knowledge of sources and emissions of various contaminants, carried out after the First AMAP assessment in 1997. This document is not yet the assessment report but eventually is going to be a contribution to the Second AMAP assessment after the Workshop on Sources, Emissions and Scenarios. The Working Document is a continuation of reporting of data that started at the 1999 AMAP Workshop in Bergen. The recommendations and conclusions from the later meeting are examined in the Working Document. They are enclosed in Annex 1.

Major emphasis in the Working Document is placed on POPs and trace metals for at least two major reasons. Selected POPs and trace metals, particularly Hg and Cd are found to may cause environmental and human health problems in the whole area of the Arctic, while pollutants causing increased acidification pose a problem in certain parts of the Russian Arctic, often called "polluted Arctic." As such, these problems should be treated with local policies and strategies. Another reason for prioritizing POPs and HMs in this Document is the fact that the Second AMAP assessment starts with these contaminant groups.

Concerning the period from the First AMAP assessment, major progress has been made in completion and updating the information on global emissions of selected POPs, such as PCBs, HCHs, dioxins and mercury. Emission inventories for the reference year 1995 have been estimated with emission maps providing the information on spatial distribution of emissions, needed for the modeling purposes.

Less information is available on global emissions of other important organic and inorganic contaminants, although it should be added that the regional emission inventories are available for Cd, Pb, various pesticides, HCB, and other POPs in Europe and North America. An effort can be suggested to combine these regional inventories and to produce a global survey with global emission maps.

Discharges of contaminants to rivers and other aquatic ecosystems are much less evaluated than their atmospheric emissions.

Major emphasis in estimating the emissions of contaminants to the air and water was placed on anthropogenic sources. For some contaminants, including mercury, natural sources and re-emission from terrestrial and aquatic surfaces to the air can be important in the assessment of the drivers of environmental change of the Arctic environment. Some approaches have already been undertaken in this direction, e.g. in Canada.

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**Recommendations concerning emission inventory activities  
elaborated at the AMAP Workshop on Modelling and Sources in  
Bergen, Norway,  
14–16 June 1999**

*Recommendation 20:*

**Update global usage and emissions of HCHs (1°x1°) by adding  $\gamma$ -HCH (1990) with monthly resolution.**

| <i>Timeframe:</i>    | <i>Who:</i>                            | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|----------------------|--|-------------------|-----------------------|------------------|
| Short-term (planned) | Li, Scholtz                            | –                 | GEIA, UN ECE          |                  |
|                      | High activity, completion by end 2000) |                   |                       |                  |

*Recommendation 21:*

**Project global emissions of HCHs ( $\alpha$ - and  $\gamma$ -) from 1990 to 2000 and 2010.**

| <i>Timeframe:</i> | <i>Who:</i> | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|-------------------|-------------|-------------------|-----------------------|------------------|
| Medium-term       | Li, Scholtz | Low to Medium     | GEIA, Canadian NPC    | –                |

*Recommendation 22:*

**Generate a global usage and emission DDT, DDE and toxaphene inventory (1°x1°) for 1990 and 2000.**

| <i>Timeframe:</i>    | <i>Who:</i>                            | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|----------------------|--|-------------------|-----------------------|------------------|
| Short-term (planned) | Li, Scholtz                            | –                 | GEIA, UN ECE(         |                  |
|                      | High activity, completion by end 2000) |                   |                       |                  |

*Recommendation 23:*

**Support development of [1] a soil PCB inventory and [2] first estimate of a (1°x1°) global emission inventory of PCBs with congener resolution and emphasis on coplanar species.**

| <i>Timeframe:</i>  | <i>Who:</i>                                  | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|--|--|-------------------|-----------------------|------------------|
| Medium- to long-term; preliminary work on [2] is underway, completion by end 2000? | [1] Li (AES) –<br>[2] Pacyna, Breivik (NILU) | –                 | GEIA, UN ECE, EU      | –                |

*Notes:*

Linked to the recommendation concerning 'a soil survey of OC's and PCB's within the AMAP/EMEP domains, involves [1] compiling the soil PCB inventory and [2] using exchange estimates to calculate emissions based on soil/vegetation surface exchange with the atmosphere

*Recommendation 24:*

**Update GEIA 1990-data (1°x1°) for Pb and Hg for 1995, with emission elevation resolution split between 0-100 m and >100 m.**

| <i>Timeframe:</i> | <i>Who:</i>          | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|-------------------|----------------------|-------------------|-----------------------|------------------|
| Short-term        | Pacyna, Scholtz<br>– |                   | Low to Medium         | GEIA, UN ECE     |

*Recommendation 25:*

**Support the development of a speciated global emission inventory for Hg<sup>0</sup>, Hg<sup>2+</sup> and Hg(p).**

| <i>Timeframe:</i>     | <i>Who:</i> | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|-----------------------|-------------|-------------------|-----------------------|------------------|
| Short- to Medium-term | Pacyna      | Medium            | GEIA, UN ECE          | –                |

*Notes:*

Difficult but essential for Hg modeling work – could be achieved by extrapolating ongoing work within NILU (EU project) on Europe to the global coverage.

*Recommendation 26:*

**Develop a first global 1995 (+1990=) emission inventory for Cd (1°x1°), with emission elevation resolution split between 0-100 m and >100 m.**

| <i>Timeframe:</i> | <i>Who:</i>          | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|-------------------|----------------------|-------------------|-----------------------|------------------|
| Medium-term       | Pacyna, Scholtz<br>– |                   | Medium                | GEIA, UN ECE     |

*Recommendation 27:*

**Resolve Cd and Pb emissions into particulate source fractions.**

| <i>Timeframe:</i> | <i>Who:</i> | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|-------------------|-------------|-------------------|-----------------------|------------------|
| Long-term         | ?           | Medium            | GEIA, UN ECE          | Low              |

*Recommendation 28:*

**Promote activities to estimate natural global emissions of Hg (soil fluxes, ocean fluxes, volcanoes).**

| <i>Timeframe:</i> | <i>Who:</i>          | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|-------------------|----------------------|-------------------|-----------------------|------------------|
| Long-term         | Petersen (GKSS)<br>– |                   | Medium                | GEIA, UN ECE     |

*Recommendation 29:*

**Support the development of an update of 1985 global GEIA-inventory for SO<sub>x</sub>, NO<sub>x</sub> inventory to 1990, with emission elevation resolution split between 0-100 m and >100 m.**

| <i>Timeframe:</i>                                      | <i>Who:</i>                                  | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|--|--|-------------------|-----------------------|------------------|
| Short-term (activity underway, completion by end 2000) | Scholtz (CGEIC), High Benkowitz (Brookhaven) | –                 | –                     | GEIA             |

*Recommendation 30:*

**Support the development of global GEIA (1°x1°) NH<sub>3</sub> and VOC emission inventories for 1990.**

| <i>Timeframe:</i> | <i>Who:</i> | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|-------------------|-------------|-------------------|-----------------------|------------------|
| Short-term        | –           | –                 | GEIA                  | –                |

*Notes:*

Required to support proposed Hg modelling work.

*Recommendation 31:*

**Develop approaches to estimating changes in emissions over time to account for changing levels of economic activity, changes in fuels and fuel use, developments in control technology, etc.**

| <i>Timeframe:</i> | <i>Who:</i> | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|-------------------|-------------|-------------------|-----------------------|------------------|
| –                 | –           | –                 | –                     | –                |

*Notes:*

There is a general need for up-to-date emissions inventories; current work tends to produce inventories with a time lag of 5–10 years relative to the current situation. For North America, a system has been developed to reprocess data and rapidly generate updated gridded emissions inventories. Similar systems need to be developed for inventories on a global scale.

*Recommendation 32:*

**Provide size distributions for particulate emissions by source type.**

| <i>Timeframe:</i> | <i>Who:</i> | <i>Resources:</i> | <i>Organizations:</i> | <i>Priority:</i> |
|-------------------|-------------|-------------------|-----------------------|------------------|
| –                 | –           | –                 | –                     | –                |



**Appendix B**  
**Individual Presentations**



## **Individual Presentations - Session I:**

**6.1 Individual Presentations – Session I**

**6.1.1 *Janina Fudala***

### **AMAP WORKSHOP ON SOURCES, EMISSIONS AND DISCHARGES**

**NILU, Kjeller, 23 - 24 August, 2001**

### **EMISSION INVENTORIES IN POLAND**

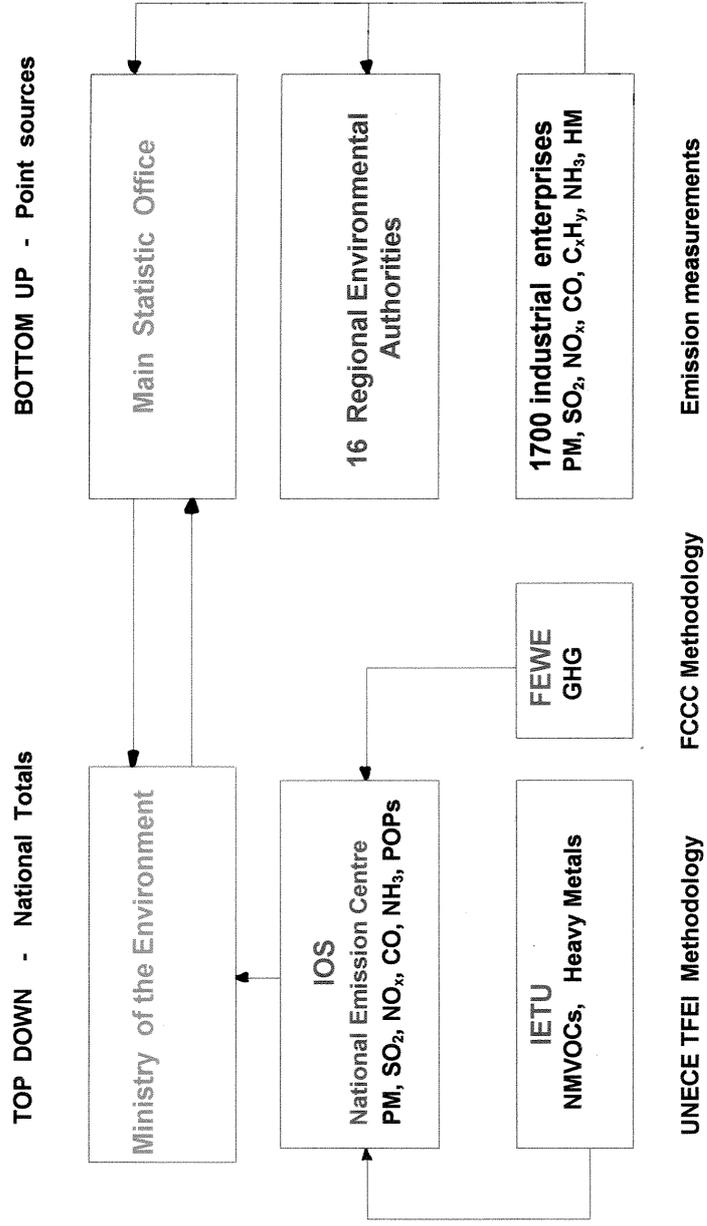
**Janina FUDALA**

**Institute for Ecology of Industrial Areas**

**Katowice, Poland**



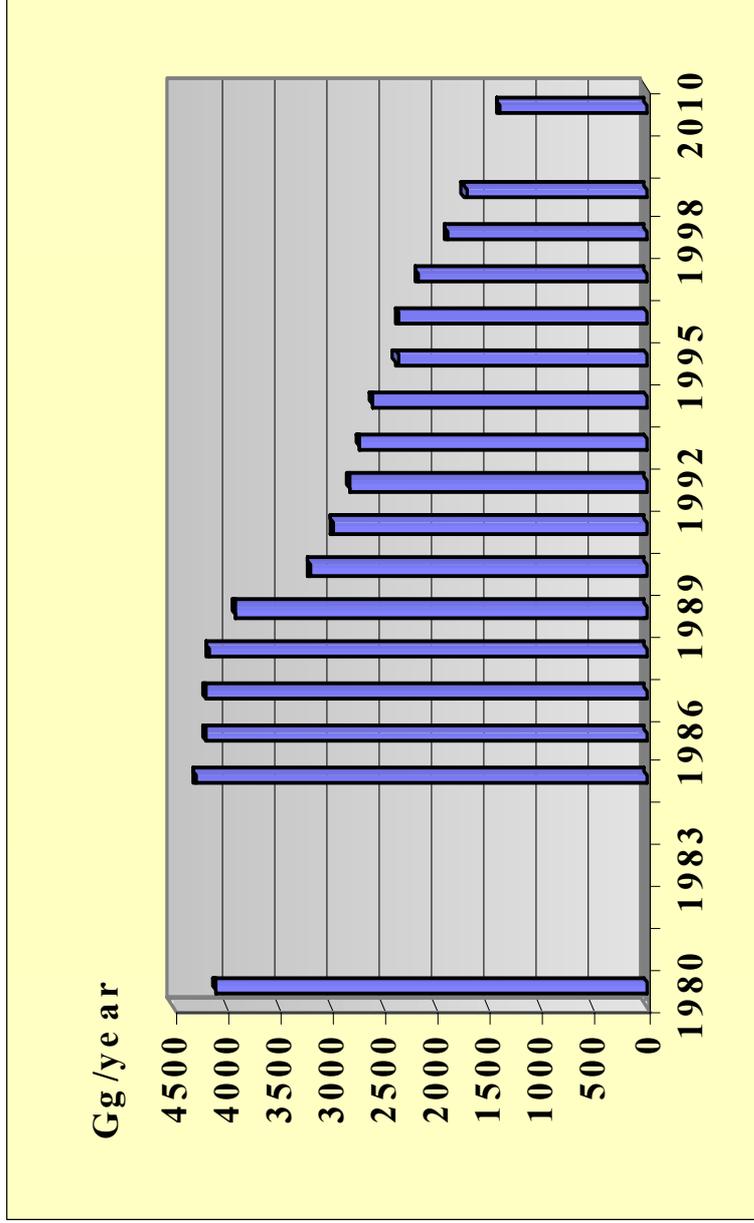
STRUCTURE OF THE EMISSION INVENTORIES IN POLAND



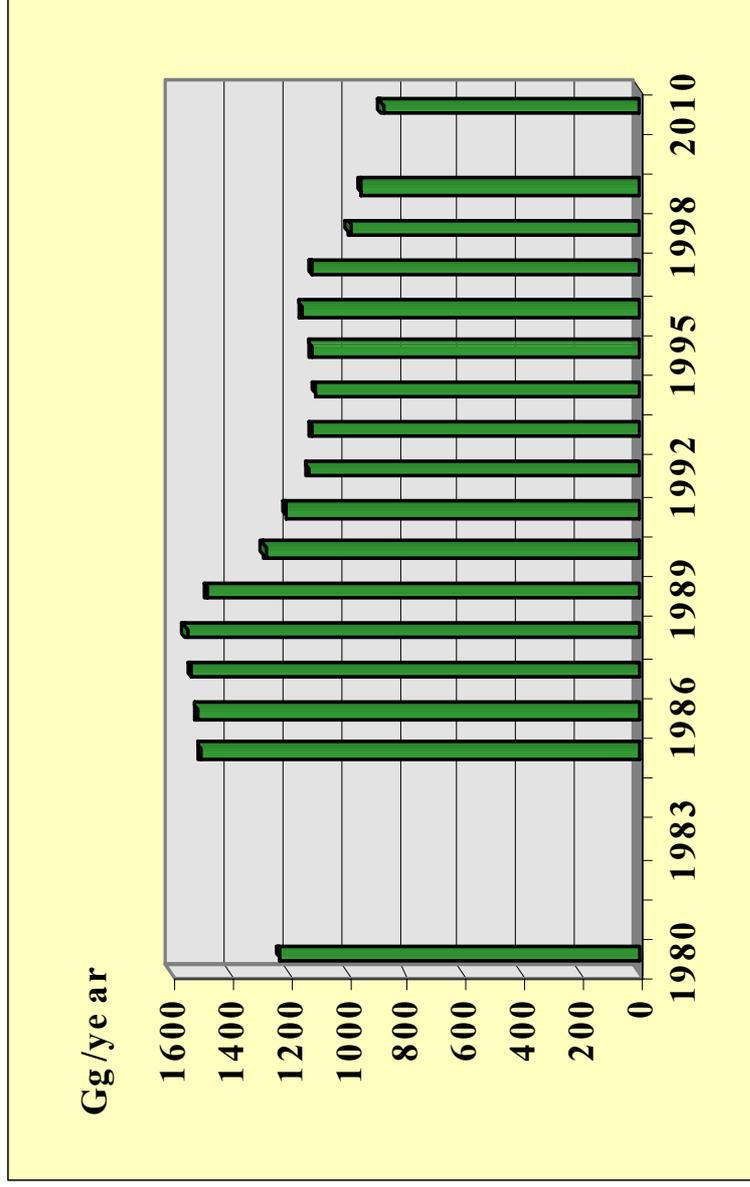
**Emissions of air pollutants in the 1999 in Poland and their contribution to the European emissions**

| <b>Pollutant</b> | <b>Unit</b> | <b>Poland</b> | <b>Europe</b> | <b>Polish contribution to European emission [%]</b> |
|------------------|-------------|---------------|---------------|---|
| PM               | [Gg]        | 871           | 12282         | 7,09  |
| SO <sub>2</sub>  | [Gg]        | 1718          | 14233         | 12,07   |
| NO <sub>2</sub>  | [Gg]        | 950           | 13549         | 7,01  |
| NMVOG            | [Gg]        | 731           | 12710         | 5,75  |
| NH <sub>3</sub>  | [Gg]        | 341           | 4066          | 8,39  |
| CO               | [Gg]        | 4363          | 49174         | 8,87  |
| CO <sub>2</sub>  | [Gg]        | 329739        | 4444000       | 7,42  |
| N <sub>2</sub> O | [Gg]        | 75            |               |   |
| CH <sub>4</sub>  | [Gg]        | 2250          | 21013         | 10,71   |
| PCB              | [kg]        | 2304          |               |   |
| BaP              | [kg]        | 53492         |               |   |
| BbF              | [kg]        | 55117         |               |   |
| BkF              | [kg]        | 14937         |               |   |
| IcdP             | [kg]        | 56960         |               |   |
| PAH (S6)         | [Mg]        | 451           |               |   |
| PCDDs/Fs         | [mgTEQ ]    | 287406        |               |   |
| Cd               | [Mg]        | 61,7          | 302,          | 20,40   |
| Pb               | [Mg]        | 745           | 10504         | 7,09  |
| Hg               | [Mg]        | 27,1          | 195           | 13,90   |

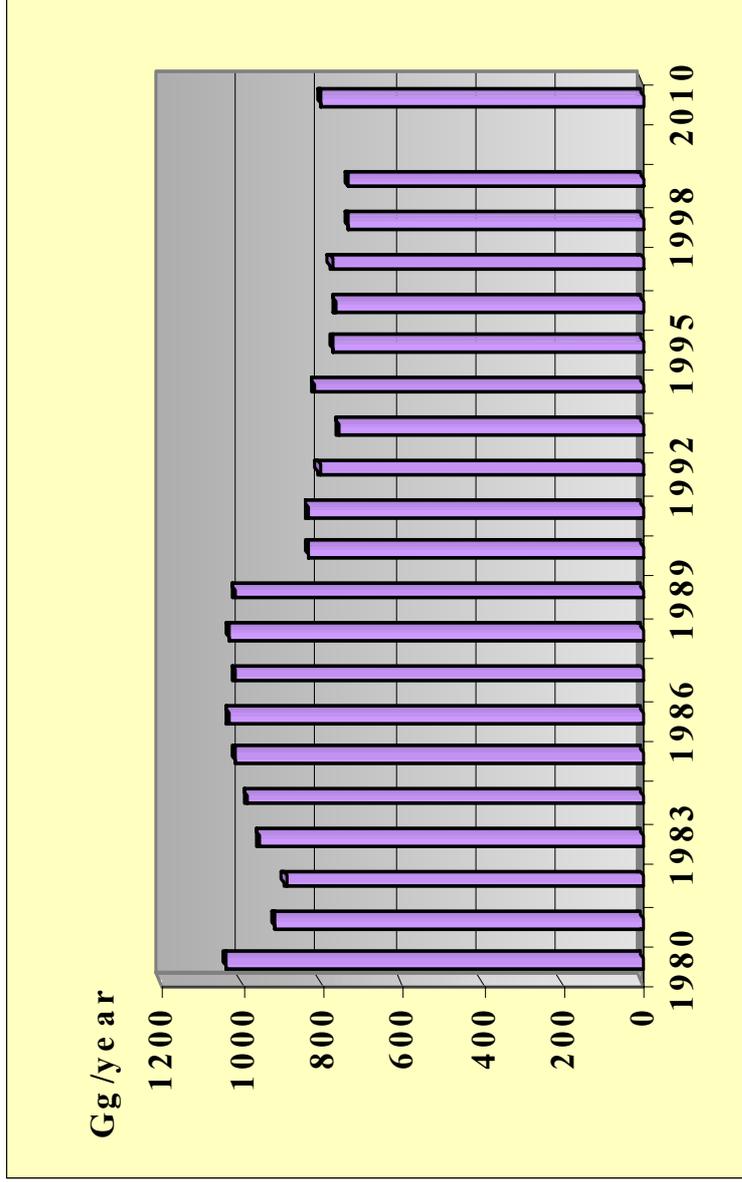
## SO<sub>2</sub> Emission Changes in Poland



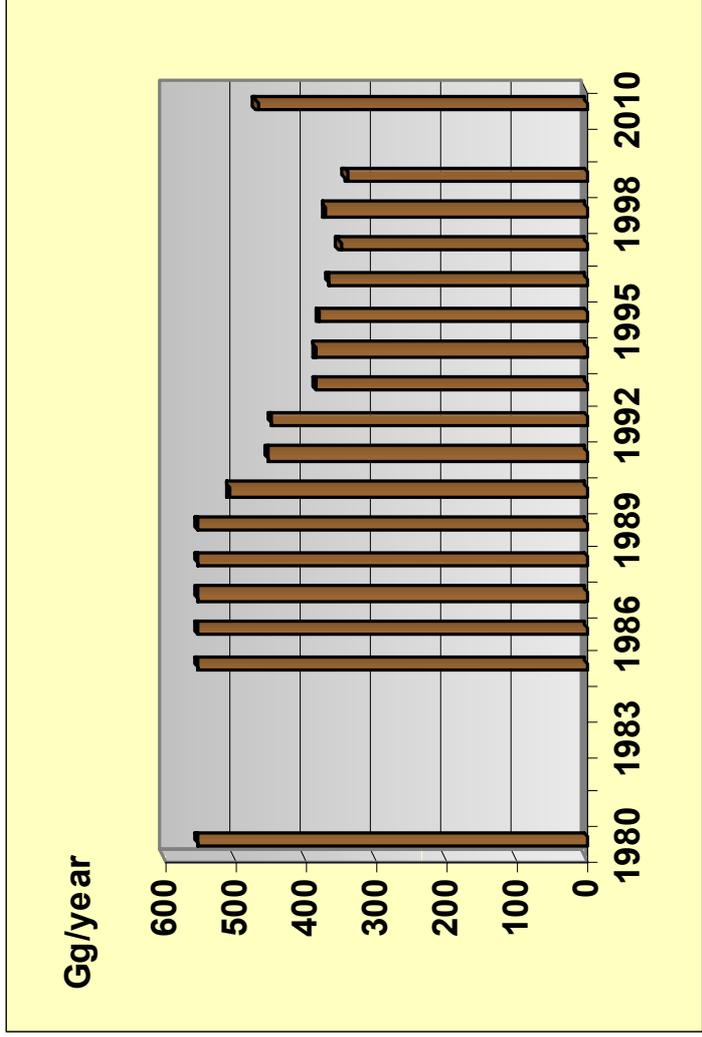
## NO<sub>2</sub> Emission Changes in Poland



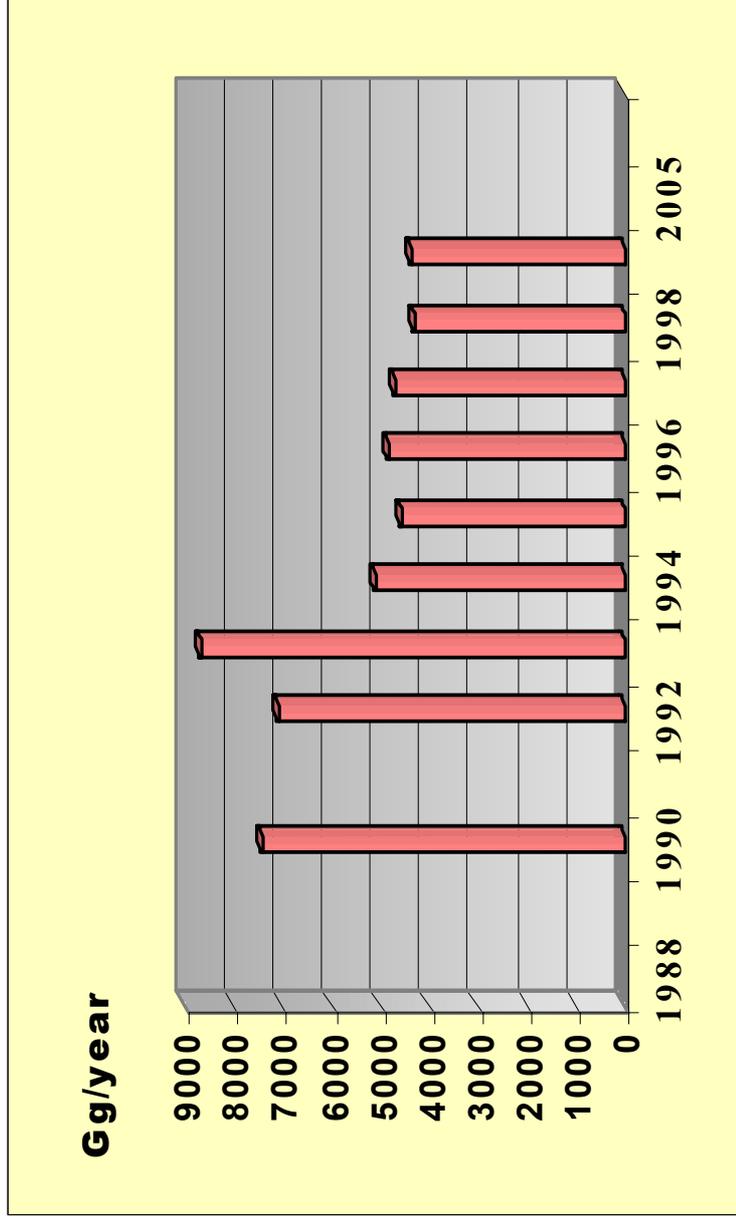
## NMVOCs Emission Changes in Poland



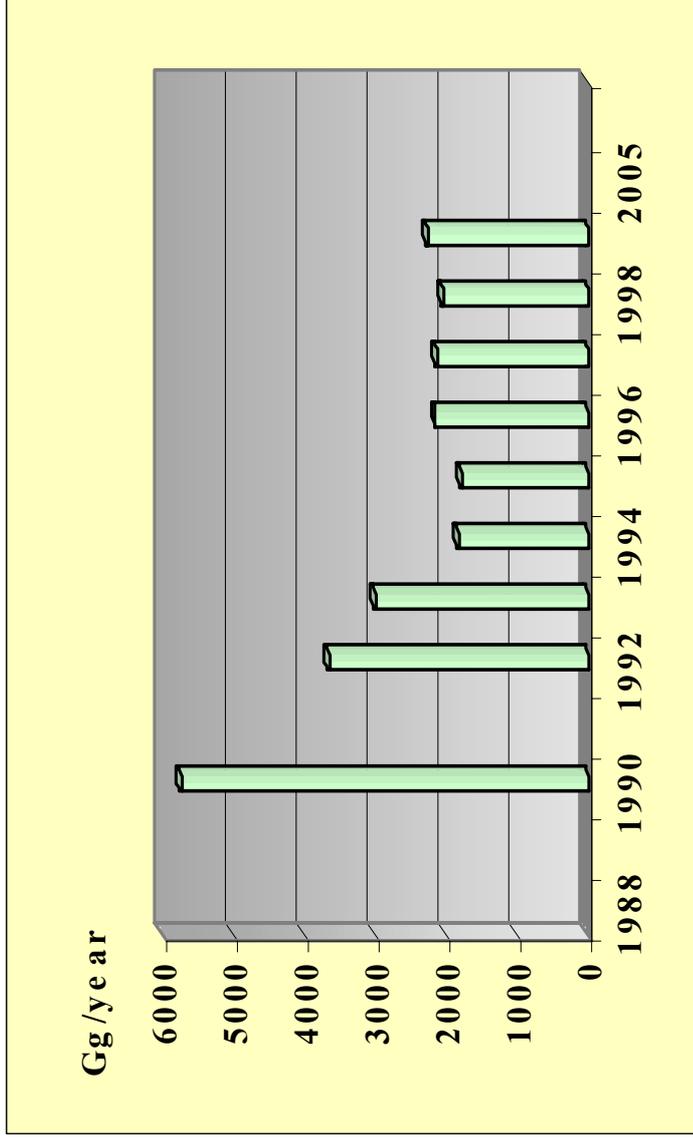
# NH<sub>3</sub> Emission Changes in Poland



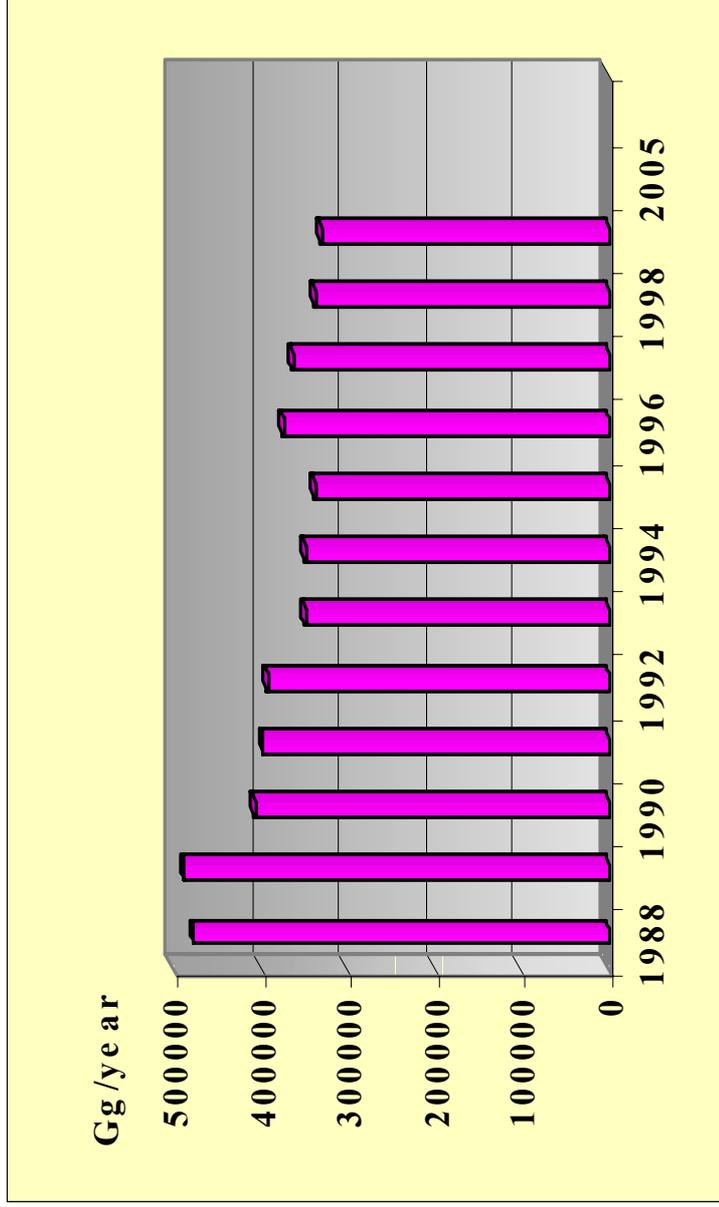
## CO Emission Changes in Poland



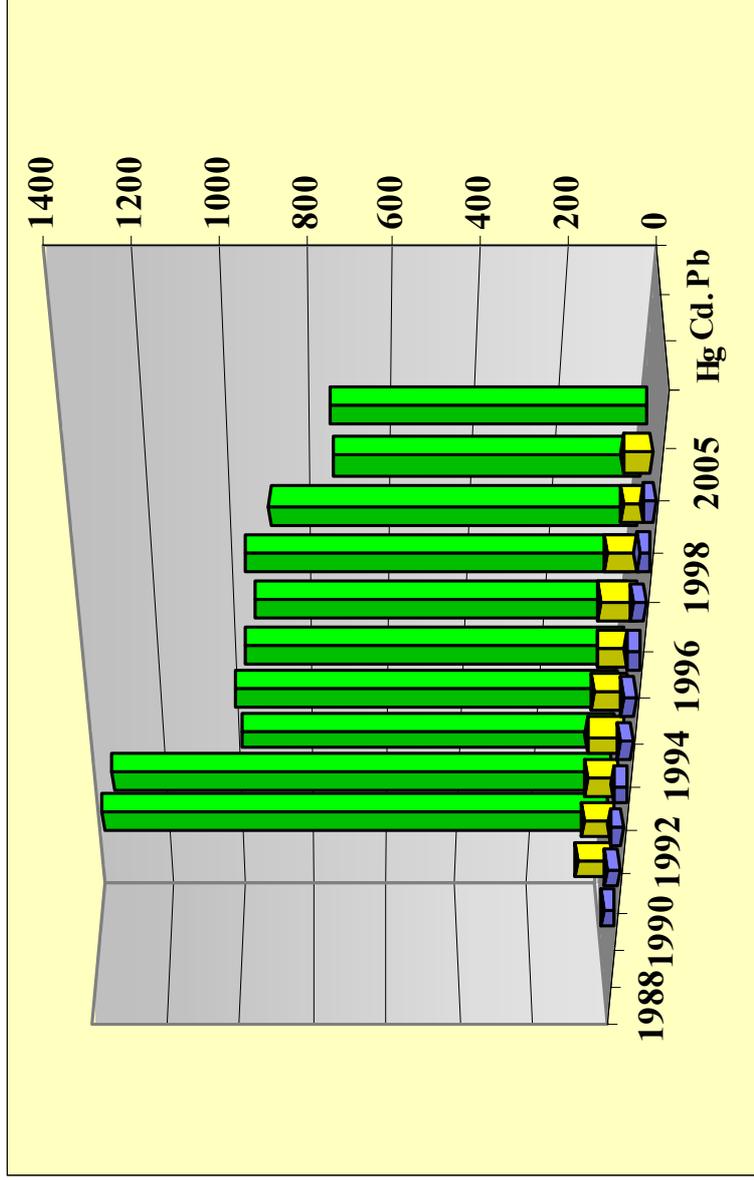
# CH<sub>4</sub> Emission Changes in Poland



# CO<sub>2</sub> Emission Changes in Poland



# Heavy Metals Emission Changes in Poland





6.1.2 *Sergey Kakareka*

**IDENTIFICATION AND ESTIMATION OF  
SOURCES OF POPS ENVIRONMENTAL  
CONTAMINATION  
IN BELARUS**

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Minsk, Belarus

## **Abstract**

The paper considers POPs emission factors estimation and their application for POPs emission evaluation on the territory of Belarus.

Emissions of the following groups of pollutants are discussed: PAHs, dioxins/furans (PCDD/PCDF) and polychlorinated biphenyls (PCB) as requested for EMEP database.

## **POPs emission factors estimation project**

Parties to the 1979 Convention on Long-Range Transboundary Air Pollution are obliged to present information on national emissions appropriate to the requirements of EMEP in completeness and structure. In particular, in the annual information all Parties submit data on national annual emissions of selected POPs; it is recommended to present the information in SNAP source-sectors split; through the certain intervals of time the Parties are recommended to submit data about large point sources emissions and emissions, distributed over 50x50 km<sup>2</sup> EMEP grid etc.

However, performance of these obligations meets significant difficulties. Actually nowadays only single countries of the Commonwealth of Independent States (CIS) estimate POP emissions on their territory. So the database for modeling of these substances is poor.

Taking into account current situation with data on national POPs emission, after consultations with the MSC-E specialists we suggested a project for improvement of POP emission inventory in the CIS countries. Its fulfilment was a contribution in-kind of the Republic of Belarus into the EMEP for the years 1999-2000 through MSC-East. This project was done under the recommendations of the EMEP Steering Body (EB.AIR/GE.1/1999/2).

Title of the project: Evaluation of Emission Factors of Selected POPs from Main Activities of the CIS Countries

## **Goal:**

Scientific and methodological provision of POP emission inventory improvement in the Former Soviet Union countries

## **Project objectives**

- Test study of POPs content in emissions
- Research and analysis of selected technologies for the estimation of emission factors of POPs
- POPs emission factors elaboration

Project was divided into 2 stages: first stage – 1999, and second stage - 2000  
1999 report included short description of legislative and regulative measures on POPs and methodologies of their testing in FSU countries, description of 8 activities - sources of POPs releases into the environment, estimates of PCB emission factors from electrical equipment, approbation of the Atmospheric

Emission Inventory Guidebook for POPs emission inventory on the territory of the Republic of Belarus.

2000 report contains the description of methodology of work and results obtained in 2000, including:

- summary on PAH, PCDD/PCDF and PCB emission factors for 22 source categories of their emissions into the environment;
- new experimental data on PAH and PCB content in particulate and gaseous emission.

## **TEST STUDY OBJECTS**

In 2000 the following POPs emission source categories were tested:

- solid fuels combustion in residential combustion units
- processes in ferrous industry
- open burning of municipal refuse
- forest fires (simulation)
- peatland fires (simulation)
- leaves fall-off and other landscape refuse burning

### **Residential combustion units**

- *boiler units of small efficiency* (autonomous boiler plants for schools, hospitals, kindergartens, small industrial and hand-feed boilers used for residential heating)
- *residential furnaces* (heating, heating-cooking, cookers, sauna furnaces, fireplace, furnaces for greenhouses cooking, etc)

### **Open burning**

- burning (fires) in natural conditions  
simulation of various types of open burning

## **METHODOLOGY OF EMISSION SOURCES TESTING**

Experimental work for the POPs emission factors evaluation included:

- aerosol and vapour POPs sampling from off-gases
- fly ash sampling from stack walls and from control equipment
- samples preparations
- analytical determination of POPs in samples
- data processing

## **METHODOLOGY OF SAMPLING**

- **off-gases sampling**

Method of external filtration (4 sampling schemes)

Method of internal filtration (2 sampling schemes)

**fly ash (soot) sampling from gas conduits and dust control equipment**

## **POPs ANALYTICAL DETERMINATION**

**PAH** - gas chromatography and mass spectrometry (Selected Ion Monitoring method, SIM)

### **PAH compounds:**

- naphthalene
- acenaphthylene
- acenaphthene
- fluorene
- phenanthrene
- anthracene
- fluoranthene
- pyrene
- benzo(a)anthracene
- chrysene
- benzo(b)fluoranthene
- benzo(k)fluoranthene
- benzo(a)pyrene
- indeno(1,2,3-c,d)pyrene
- dibenzo(a,h)anthracene
- benzo(g,h,i)perylene

**PCB** - gas chromatography and modified method EPA 8081A using chromatography mass spectrometer HP 5890/5972

### **PCB compounds:**

- dichlorobiphenyl
- trichlorobiphenyl
- tetrachlorobiphenyl
- pentachlorobiphenyl
- hexachlorobiphenyl
- heptachlorobiphenyl
- octachlorobiphenyl
- nonachlorobiphenyl
- decachlorobiphenyl

**HCB** - EPA 8270 GC/MS method

**SOME RESULTS**  
**PAH EMISSION FROM RESIDENTIAL FUEL COMBUSTION**  
**PAH emission factors for firewood combustion in residential furnaces, g/t fuel**

| Compound                | Russian Oven |      | Stove     |      | Kitchen-Range |      |
|-------------------------|--------------|------|-----------|------|---------------|------|
|                         | range        | mean | range     | mean | range         | mean |
| Benzo(b)fluoranthene    | 1.0 - 41.4   | 18.5 | 0 - 3.7   | 1.8  | 0 - 4.4       | 2.4  |
| Benzo(k)fluoranthene    | 2.2 - 9.3    | 4.9  | 0 - 1.0   | 0.5  | 0 - 0.9       | 0.6  |
| Benzo(a)pyrene          | 3.7 - 20.5   | 8.2  | 2.2 - 2.3 | 2.2  | 1.2 - 2.0     | 1.6  |
| Indeno(1,2,3-c,d)pyrene | 2.2 - 6.3    | 4.1  | 0.6 - 1.4 | 1    | 0.4 - 1.6     | 1    |

**PAH emission factors for peat combustion in residential sector, g/t fuel**

| Compound                | Emission Factors for the CIS |      | Technical Paper...,<br>1995 |
|-------------------------|------------------------------|------|-----------------------------|
|                         | range                        | mean |                             |
| Benzo(b)fluoranthene    | 0.00 - 3.0                   | 1.55 | 1.15                        |
| Benzo(k)fluoranthene    | 0.00 - 0.94                  | 0.39 | 0.525                       |
| Benzo(a)pyrene          | 0.000 - 2.14                 | 0.79 | 0.845                       |
| Indeno(1,2,3-c,d)pyrene | 0.04 - 0.99                  | 0.39 | 1.11                        |

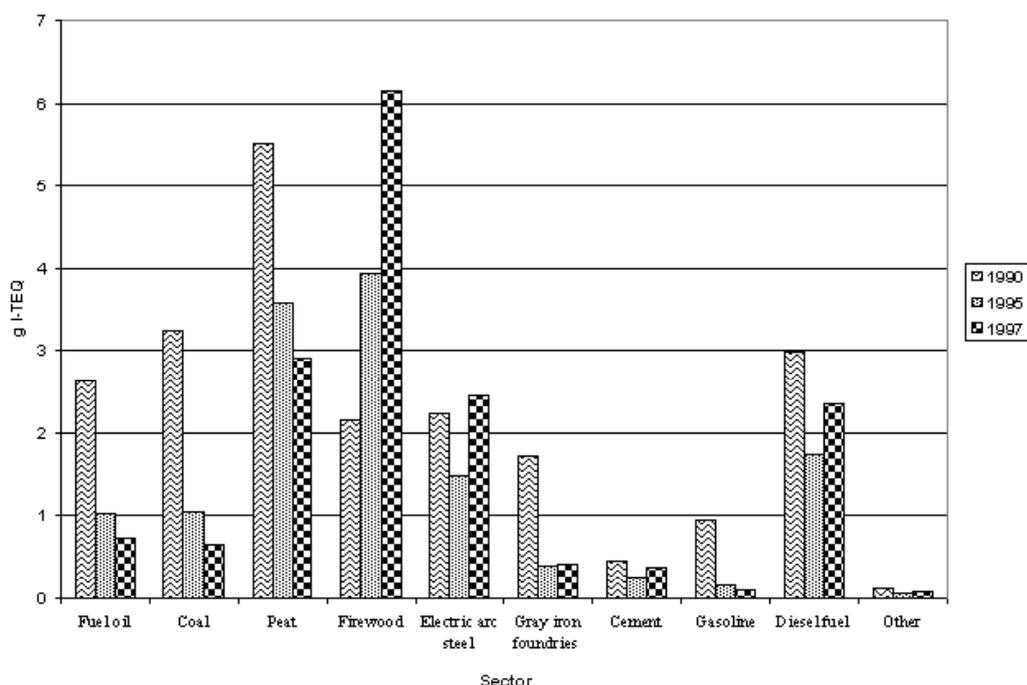
## POPs emission inventory in Belarus

Estimations on the basis of emission factors obtained have showed, that the main contribution into dioxin emissions has firewood and peat combustion. Only electric steel smelting can be considered as a large emission point source. Belarus as a whole is not a large emission source of such POPs as dioxins. Its share in European emissions is significantly below 1%.

According to estimates, firewood, peat and diesel fuel combustion have the largest contribution in PAH emissions. However, a number of important sources such as wood preservation, forest (and other) fires on this stage got only quality evaluation.

Leakage from transformers and damaged capacitors were estimated as the main source of PCBs discharge into the environment.

### Dioxin/furan emission on the territory of Belarus per source sectors in 1997, g I-TEQ



## Conclusions

The analysis of technological peculiarities of major sources of POPs emission on the territory of the FSU countries have been carried out. Solid fuel combustion in power generation and residential sector, wood preservation, mobile sources and other processes have been analysed. For these sources available information necessary for estimation of emission factors have been generalized. Main attention was paid onto PAH emission testing.

Most detailed study was conducted for various types of residential furnaces (Russian stove, heating furnace, heating boiler, kitchen range etc.) and for open burning sources (using model unit for simulation of open burning of household wastes, forest fires, peatland fires, leaves fall-off and agricultural wastes).

It has been detected that firewood combustion in residential sector is the most significant source of PAH emission: mean content of benzo(a)pyrene in fly ash and soot amounts 224 mg/kg. Similar PAH content in emission from wood waste. PAH emission from peat briquette combustion approximately 10 times lower.

Open burning processes result in significant amounts of PAHs, mainly low molecular species: naphthalene, acenaphthylene, phenanthrene, fluorene etc. Maximum levels of 16 PAH sum and 4 indicator PAH sum have been detected in fly ash and soot from household waste combustion.

Low concentrations of PCBs in soot from chimney walls of residential furnaces for solid fuels combustion have been detected. In flue gases PCBs have not been revealed. PCBs have not been detected also in emission from open burning.

Hexachlorobenzene has not been detected in emission and soot from fuel combustion and open burning.

Emission factors obtained have been applied for preparation of national emission data on POPs for Belarus. They also will be useful for FSU countries national emission experts in preparation their emission data.

**6.1.3 *Pieter van der Most***

**POLLUTANT RELEASE AND TRANSFER REGISTERS**

**Design, support, and cooperation**

**Pieter van der Most**

**(Inspectorate for Environmental Protection  
The Netherlands)**

**Paper presented at the AMAP Workshop on Sources, Emissions and Discharges**

**August 23-24, 2001 Norwegian Institute for Air Research in Kjeller.**

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### ***Introduction***

The ultimate goal of an environmental policy is improving the quality of the environment. To achieve this goal a source directed policy needs to be developed. To make such a policy effective tools are needed giving a relation between concentrations measured and their origin. A Pollutant Release and Transfer Register can in combination with transport models and measurements provide such a tool.

Many countries have started with the development of such instruments. However PRTR's covering emissions from all sources to all compartments are rare so far. Harmonisation of methods and definitions is also an important aspect. The work on the UNECE/EMEP convention on long range transport of pollutants has since long recognized that need and established a taskforce on emission inventories, also responsible for a guidebook aiming at the support of the reporting obligations.

In the Environmental conference in Rio de Janeiro in 1992 the need for PRTR systems as a tool for developing an environmental policy was recognized. The OECD coordinates those activities supported by UN organisations like UNITAR and UNEP Chemicals. This resulted in 2000 in the establishing of a taskforce on release estimation techniques aiming at information exchange between different countries. The third development is rather recent. In the Aarhus convention the right of the public to receive information about the environmental situation was recognized. The Aarhus PRTR taskforce aims at defining a legally binding instrument for giving the public access to this information. In this paper the three developments will be described with some conclusions about an optimal cooperation.

### ***UNECE/EMEP Taskforce on Emission Inventories***

The Taskforce on Emission Inventories started in 1991 as a support of the Long Range Transport of Air Pollutants (LRTAP) reporting obligations related to the European Monitoring Program (EMEP). To support the different countries in fulfilling their obligations an Atmospheric Emission Inventory Guidebook was developed. Responsible for the contents of this guidebook are four expert panels: Combustion and Industry, Transport, Agriculture and Nature, and Projections and Verification. The Guidebook is structured in chapters with each chapter presenting information in a common format. This format covers in 18 standard points the relevant information necessary for incorporating the described activity in a PRTR. A recent development consists in defining a simple method giving advised default values enabling a country to make a first estimation of the relevance of a source. If the source appears to be relevant a detailed method is described as an example of a possible approach. So far the guidebook only gives information about air pollutants for those activities incorporated in the reporting obligations. A discussion about going multimedia has been going on for some time.

### ***OECD Taskforce on Release Estimation Techniques***

In the 1992 the Rio de Janeiro Environmental Conference all countries are advised to establish a Pollutant Release and Transfer Register. The OECD is stimulating and coordinating these activities in close cooperation with organisations of the United Nations like UNEP Chemicals and UNITAR. Since then in several countries like Australia or Japan PRTR systems are operational

whereas in many other countries pilots or parts of PRTR's are operational. This resulted in a situation where the need for a forum for exchange of experiences about approaches and methods. In 1999 in a workshop in Canberra it was agreed to institute a taskforce for that purpose. This taskforce started at a Stockholm meeting in september 2000 and met again in Washington in april 2001. Interesting activities from the program are among others:

- create a compendium of release estimation techniques for point- and diffuse sources.
- establish a clearinghouse for these activities
- establish an informal communication relay

### ***UNECE Taskforce on Pollutant Release and Transfer Registers (Aarhus)***

In 1998 the Convention on access to information, public participation in decision making and access to justice in environmental matters was signed in Aarhus. To make the relevant information available to the public a PRTR is to be designed following the guidelines produced in an earlier stage by OECD. The structure defined should become a legally binding instrument to be implemented in the countries that ratify the Convention.

At the moment the Taskforce consists of a working group that is preparing a protocol to be adapted at the Environmental conference in Kiev in 2003, and a technical group that is discussing the activities and the substances to be incorporated in different steps. If the implementation becomes legally binding the importance of international agreement on methods and definitions like defined in the other taskforces becomes more important.

### ***Conclusions***

The importance of having an insight in the size and the location of the different sources of pollution is recognized worldwide. A Pollutant Release and Transfer Register is an essential tool for defining an effective policy as well as for monitoring the results of that policy. As many systems become operational the importance of communication and exchange of experiences become increasingly important. Activities like those started by the OECD taskforce on release estimation techniques can provide the instruments for that. As to the input for those systems there is still a lot of work to be done. For instance the information about emissions from metals and persistent organic pollutants is still limited. Also the methods for estimation of emissions to water have a long tradition of being hidden in gray literature often in inaccessible languages. But the instruments for improving this situation are available or in the process of being developed.

#### 6.1.4 *Slava Gordeev*

### **Heavy metals in the Russian Arctic Rivers and some of their estuaries: concentrations and fluxes**

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#### **Abstract**

The review of available information about HM concentrations in water and suspended matter of the main Russian Arctic rivers and some of their estuaries is presented.

The reliable data of the recent years show that in the lower courses of the biggest Arctic rivers - the Ob', Yenisey and Lena - HM concentrations in water and SPM do not exceed the global averages for the river discharge. Furthermore, the concentrations of such HM as Cd, Pb and As are appeared to be the lowest in comparison with other big rivers in the world.

Preliminary assessments of HM fluxes show that the outflow of HM considered (Cu, Pb, Zn, Ni, Cd, Hg, Fe) in particulate form is prevailed over their input in dissolved form - from 58% for Cu up to 98% and more for Pb and Fe.

The studies of dissolved HM behaviour in the river-sea mixing zones of the Ob, Yenisey and Lena Rivers (Martin et al., 1993; Dai, Martin, 1995; Guieu et al., 1996) permitted to assess the effective river concentrations of some HM and on its base to calculate net riverine fluxes into the ocean. These calculations show that net river input of Cu and Ni to the Eurasian Arctic Ocean is in 1.2-1.4 times higher than their gross input, for Fe net river input is 4 times less than gross river input.

#### **1. Introduction**

Heavy metals (HM) are among the most significant pollutants in the environment. Pollution by some of them, by lead, for example, has assumed a global character. Higher than natural concentrations of lead were recorded even in the glacial ice cores in Antarctic (Murozumi et al., 1969).

Shallow seas of the Russian Arctic are potentially influenced by the runoff of the biggest Siberian rivers such as the Ob', Yenisey, Lena and other. Naturally, the rivers are considered as one of the main sources of pollutant input, including HM, to the Arctic ocean.

To have an adequate notion about practical level of HM's pollution of river and sea waters and suspended particulate matter, we need in reliable data about HM concentrations in aquatic environment. The marine chemists were the first who have realized the problem of reliable HM concentration receiving. Wide scattering of data and an absence of any regularities in distribution of dissolved HM with depth in the oceans forced the specialists to fulfil the great volume of methodical investigations and numerous intercalibrations. As a result, the criteria were produced to evaluate the analytical data from a point of view of their reliability.

First of all it means good correlation of the intercalibration results and “oceanographic consistency”. The last one assumes that the detailed distributions of trace metals with depth in ocean must be smooth and oceanographically consistent with known chemical and physical processes in the ocean (Boyle et al., 1974; Bruland, 1983).

In 1979 well-known marine geochemist John Edmond declared that “the trace-metal data (in sea water) before 1973 are almost entirely invalid. It is reasonably to expect that a similar situation holds for fresh waters” (Edmond, 1981).

The first systematic measurements of HM in the rivers of the former Soviet Union, including the Arctic rivers were started by the Hydrometeorological Survey of the USSR at the beginning of 50-th (Konovalov, 1959; Konovalov et al., 1968). The investigations of HM and their forms in river water were carried out in the Geological Institute of the USSR Academy of Sciences and in other institutes (Nesterova, 1960; Kontorovich, 1968). Big volume of data on HM in the Arctic rivers, parallel with other pollutants, was obtained in framework of the State System for Observation and Control of Environmental Pollution of the USSR (and Russian Federation later) State Committee for Hydrometeorology (Melnikov, 1991; Skakalski, 1997). But at those time a proper attention was not payed to the problem of water samples contamination by the elements determined during sampling, storage and analysis.

As a result, we know now, were measured the concentrations of dissolved Pb, Cd, Zn, Cu, Hg and other metals in river waters that exceeded the real natural level in tens, hundreds, and sometime thousands times. The results for suspended particulate form of the same HM were more close to reality but quite often their levels were overestimated also. So, many works of 50-th - 60-th of the last centry have today mainly historic value.

The aim of this work is to present a review of the most reliable recent data on HM concentrations in water of the Russian Arctic rivers and of the estuaries of the largest Arctic rivers and to assess the riverine gross and net fluxes of HM to the Arctic coastal seas.

## 2. Methodical aspect

In the light of spoken above about methodical problems in receiving of reliable data on the HM concentrations in river waters it is necessary to consider the methods of sampling and analyses which were used in the publications referenced.

In framework of the USSR State Committee for Hydrometeorology in period of 50-th - 60-th of last century water samples were sometime collected by metallic samplers, water was not filtered, or was filtered through paper filters “Blue line” with pore size of few  $\mu\text{m}$ . The analyses were usually carried out by low sensitive spectral emission method with chemical preconcentration of metals. In 80-th - 90-th the situation began to change to better side. So, in the Hydrochemical Institute of Roshydromet started to use nonmetallic samplers, glass, polyethilen or Teflon-coated sample containers. Samples were filtered using nuclepore filters. Dissolved metals were measured by flame and flameless AAS and other modern methods of analysis (Nikanorov and Zhulidov, 1991; Zhulidov and Emets, 1998). As a result the measured concentrations of many HM (Pb, Cd, Zn and other) appeared to be much lower. For example, dissolved Cd, Pb and Zn were measured in water of the pristine Usman river, Don basin, in the Voronezh Biosphere Reserve from 1975 to 1993. The results were in 1975 and 1993 accordinally: Zn - 36 and  $<0.1$ , Cd - 0.29

and 0.04, Pb - 0.40 and 0.04  $\mu\text{g/l}$  (Zhulidov and Emetz, 1998). The authors note that the decrease in HM concentrations in the Usman river was an artefact due to contamination problem during sampling, filtration, storage and analysis processes. In 80-th - 90-th the samples of the Ob', Yenisey, Lena and another large Arctic rivers were analysed. Some of these results are presented in paper (Alexeeva et al., this volume).

Skakalsky (1997) summarised the data of stationary hydrochemical observations for period 1980-1995 for the Ob, Yenisey and Lena rivers including analyses of dissolved Cu, Zn and Fe. No localities of stations and number of samples were given.

Melnikov (1991) has shown the mean values of HM concentrations in water of some Siberian rivers in 1988. Again no mentions about methodical approach and localities and number of samples.

In Table 1 the data of both works that are typical for the Roshydromet System are shown and for comparison are included the recent results for the same rivers (Martin et al., 1993; Dai and Martin, 1995).

It is quite obvious very big difference between data of Skakalski and Melnikov from one side and data of Martin with co-authors from other side, particularly for Pb and Zn.

Recently was published with AMAP financial support the Atlas of environment pollutions of the aquatoria and coastal zone of the Russian Arctic Seas. The data on dissolved and particulate HM are belong to the regional centre "Monitoring of the Arctic" (Dr. S.A. Melnikov is a head of this centre). In this original handbook on chemical oceanography and ecology and monitoring of antropogenic impacts in the arctic ecosystems a significant place takes the data on HM content in many objects of the arctic environment including river water. There are the tables with a range of HM concentrations (Zn, Sn, Mn, Ni, Cu, Cd, Pb, Co, Cr, Fe) in surface waters of the lower courses of the Arctic rivers (in  $\mu\text{g/l}$  of unfiltered water), and in river suspended matter (in  $\mu\text{g/mg}$  of dry weight). Number of stations and water samples and the time of sampling are not pointed. Average concentrations of HM in unfiltered water and SPM were taken from the figures.

In November 1999 and 2000 in Siberia (Tomsk city) the I and II Interregional Conferences on Ecology of Poymas of Siberian Rivers and Arctic were held. The reports with the results of HM determinations in the middle reach of the Ob' river and some its tributaries were presented (Shvartsev et al., 1999; Sorokovikova et al., 1999; Leonova et al., 2000).

Shvartsev et al. (1999) collected water samples of the Ob' river in its middle reach (from Tom' river to Vakh river, 20 samples). Water was filtered through paper filter "Blue line". Cu, Zn, Pb and Cd were determined by inverse volt-ampereometric method, Ti, Co, Ni, Mn by quantitative emission spectral method, and other elements by NAA.

Sorokovikova with co-authors (1999) collected 53 samples of water on the Tom' river (Seversk city) and on the Ob' river (down to Surgut city). Samples were filtered through the nylon filters with 0.2  $\mu\text{m}$  pore size. Analyses of HM in filtered water were carried out by ICP-MS method.

In September, 2000 Leonova et al. (2000) sampled the Ob' river from Novosibirsk city to the mouth of the Tom' river and this river from mouth to Tomsk city. Water was collected from depth 0.6 h, was filtered through nuclepore filters (0.45  $\mu\text{m}$ ), HM were determined by AAS.

Sampling was performed during the expedition to the Lena River from Yakutsk city to the Lena delta in July and August 1994, 23 Stations (Rachold et al., 1996) and to the Lena, Yana and Khatanga Rivers in 1995-1996 (Rachold, 1999).

Integrated water samples were obtained by a special river water sampler. The water was filtered on board of the ship by vacuum filtration through Millipore filters (0.45  $\mu\text{m}$ ) in 1994 and Nuclepore filters (0.4  $\mu\text{m}$ ) in 1995-1996.

HM content in SPM samples were analysed after total acid digestion by ICP-MS.

At last I describe the methods of sampling and analyses of water and SPM that were using in framework of the Scientific Program on Arctic and Siberian Aquatorium (SPASIBA; 1989-1995, J.-M. Martin and V.V. Gordeev - co-chairs of the program).

I attract attention to this issue because namely the results of these works (Martin et al., 1993; Kravtsov et al., 1994; Dai and Martin, 1995; Coquery et al., 1995; Guieu et al., 1996; Nolting et al., 1996; Gordeev et al., in press) were used to evaluate the gross and net input of HM by the Arctic rivers to the Arctic Ocean.

There were three expeditions: in 1989 and 1991 to the Laptev Sea and the Lena delta, in 1993 to the Kara Sea and the Ob' and Yenisey estuaries. The main works in all three expeditions were carried out in open sea and in the mixing zones of sea and river waters. To have the data on the river end member the river water samples were collected in the lower courses of the rivers (Lena river - 2 stations (1989) and 6 stations (1991), Ob' river - 2 stations and Yenisey river - 3 stations (1993)). In all cases, a flat vertical CTD profile was found, showing the absence of water stratification. Surface samples were taken from a small plastic boat using acid-clean 10-liter polypropylene bottles. Sub-surface samples were collected with a Teflon pump powered by high purity nitrogen gas flux. The pumped water was directly carried to the filtration system in the class-100 laminar flow bench by PTEE tubing. Samples deeper than 10 m were sampled by Teflon coated Go-Flo bottles fixed on a clean plastic hydrowire. The samples were filtered immediately through Nuclepore filters (0.4  $\mu\text{m}$ ,  $d = 47$  mm) under N pressure.

HM concentrations were measured by graphite-furnace AAS after extraction in Class-100 clean room using a method modified from Danielsson et al., 1982.

The accuracy and the precision of the analyses have been tested using NASS-1, NASS-3 and SLRS-1 dissolved trace metal standards (Canada). In Table 2 are shown the results of the reference material after pre-concentration. Only in the case of Pb, one of 3 analyses was obviously contaminated leading to a bad recovery (201%) and a high standard deviation (84%).

In work (Nolting et al., 1996) SPM was collected by pumping water over a Nuclepore filter (0.45  $\mu\text{m}$ ) and analyses were carried out by flame and graphite-furnace AAS.

Kravtsov with co-authors, 1994 - the determinations of Cd, Pb, Cu and Zn were carried out in filtered water immediately after sampling by anodic stripping voltamperometry method directly on board of the research ship. It worth to note that this method recovers the labile ionic forms of HM.

Coquery and Cossa (Coquery et al., 1995) analyzed water samples for total dissolved and total particulate mercury. For the separation of dissolved and particulate Hg species, water samples were filtered on board ship under laminar flow hood through quartz fiber filters (Whatman QM-A, 0.8  $\mu\text{m}$ ). Filtered water was transferred into Teflon bottles and acidified with  $\text{HCl}_{\text{conc}}$ . All Hg species were

detected by cold vapour atomic fluorescence spectrometry after transformation to  $\text{Hg}^0$ , using a Merlin instrument (PSAnalytical).

Method accuracy was checked using available reference material (Hg in water 1641 d from the U.S. National Bureau of Standards; marine sediments BEST-1 from the National Research Council of Canada). Precision was 8% for  $(\text{Hg}_T)_D$  and 5% for  $(\text{Hg}_T)_P$  determinations.

### 3. HM concentrations in river water and suspended matter

#### 3.1. The White Sea basin

The biggest rivers of the White Sea basin are the North Dvina (water discharge  $110 \text{ km}^3 \cdot \text{a}^{-1}$ , sediment discharge  $3.8 \cdot 10^6 \text{ t} \cdot \text{a}^{-1}$ ), the Mezen ( $27.2 \text{ km}^3 \cdot \text{a}^{-1}$ ,  $0.9 \cdot 10^6 \text{ t} \cdot \text{a}^{-1}$ ) and the Onega ( $15.9 \text{ km}^3 \cdot \text{a}^{-1}$ ,  $0.3 \cdot 10^6 \text{ t} \cdot \text{a}^{-1}$ ).

Total discharge of all rivers in the basin is  $463 \text{ km}^3 \cdot \text{a}^{-1}$  and  $22 \cdot 10^6 \text{ t} \cdot \text{a}^{-1}$  (Gordeev et al., 1996).

In the basin, and on the Kola peninsula especially, are concentrated many mineral deposits and geochemical provinces with industrial concentrations of many rare elements. The large-scale mining and metallurgic centers are situated here: industrial company “Apatit” with mine and the plants for ore concentration; “Severonikel” with smelting complex produced Cu, Ni, Co and accompanying Au, Ag, Pt and Se; Olenegorsk ore-concentrated complex which obtains and concentrates iron-ores; large metallurgical plant “Pechenganikel”.

The result of these mining and metallurgic works activity is significant enrichment by many metals, especially by Ni and Cu, of aerosols over the aquatories of the White Sea and other Russian Arctic Seas (Shevchenko et al., 1999; Melnikov, 1991).

The reliable data on dissolved HM concentrations in the White Sea basin rivers do not exist.

HM content in suspended particulate matter (SPM) of the North Dvina and Mezen rivers, collected by filtration, were measured by Morozov et al., (1974). No enrichment by Ni, V, Cr, Co, Cd was observed.

In recent work (Howland and Pantiulin, 1994) the analyses of Zn and Pb were carried out in SPM of some small rivers of the White Sea basin: Kol’vitsa - Zn =  $35 \pm 12$ , Pb =  $52 \pm 40 \text{ } \mu\text{g/g}$ , Voronya -  $32 \pm 29$  and  $35 \pm 11$  accordingly, Knyaja -  $56 \pm 55$  and  $66 \pm 43$ , Chupa -  $102 \pm 35$  and  $28 \pm 23$ .

These Zn and Pb contents do not record any enrichment of SPM in comparison with their global river average.

#### 3.2. The Barents Sea basin

One of the most important anthropogenic HM source to water and sediments of the Barents Sea is mining-metallurgical plants in its basin, at the Kola peninsula especially. Activity of these plants is accompanying with emission to the atmosphere of sulfur compounds and dust particles enriched by HM. Particularly there are high fluxes of Ni and Cu. Annual deposits of these metals on land surface at distance 10 and 60 km from the plants are estimated to be  $50 \text{ mg} \cdot \text{m}^{-2}$ , 28 and 5 and 6  $\text{mg} \cdot \text{m}^{-2}$  accordingly at background level 0.7 and  $3.2 \text{ mg} \cdot \text{m}^{-2}$  (Lukin et al., 1998).

Through atmosphere transported fluxes of anthropogenic substances reflect the ecological conditions of small lakes in the best way. It was shown by Moiseenko et al., 1997 that the areas of the highest Ni and Cu concentrations in lake water are restricted by zone of 30-40 km around the melting shops of the plants. Concentrations of Ni, Cu, Co, Hg, Cd, exceeded the background level in 10-380 times in bottom sediments of the lakes in radius of 40 km, have positive correlation between each other indicating the common source of their distribution.

The biggest river of the Barents Sea basin the Pechora River transports annually to the sea  $131 \text{ km}^3$  of water and  $13.5 \times 10^6 \text{ t}$  of suspended matter. Pollution of the Pechora river and its tributaries has began in 60-th when were dropped to tributary of the Pechora Izhma and its tributary Ukhta more than  $20\,000 \text{ m}^3 \cdot \text{day}^{-1}$  of sewage from oil-refining plant. At mid of 60-th to the Vorkuta River were edjected more than  $130\,000 \text{ m}^3 \cdot \text{day}^{-1}$  of untreated industrial and farm-domestic sewage. In the following years the situation was aggravated due to damages of the oil-pipe-lines. So, in 1994 the great accident has happened on the Kharayaka River in the result of which 100 000 - 200 000 t of crude oil were inflowed to the Pechora Sea through the Kolva River-Usa River system (Lukin and Dauvalter, 1997).

It is quite well-known (Nriagu and Pacyna, 1988; Pacyna, 1997) that oil pollution and combustion of oil is the most important source of V and Ni Emission of the mining-metallurgical plants in the Barents Sea basin are expected to be a source of atmospheric As, Cd, Cu, In, Sb, Zn, Pb and Se (Pacyna, 1997).

The available recent data on HM in water and SPM of the Pechora River are given in Atlas of contamination..., 1999 only (Table 3).

Unfortunately the interpretation of the data for unfiltered water of the Pechora river is difficult because we don't know what part of suspended matter was involved in these analyses. Content of Pb and Cd in SPM are significantly higher than global average content.

Lukin and Dauvalter, 1997 stated that the objects to be investigated were water, sediments and whitefishes. Unfortunately HM concentrations in water and SPM of the Pechora river are absent in this publication. Bottom sediments (Table 3) were collected in July, 1995 at three stations and in October, 1995 from one station. The authors conclude that during industrial development of the region, the contents of HM in sediments has increased by 0.7-2.5 times in surface layer (Ni, Cu, Co, Zn, Cd, Pb) in comparison with the background. The lead had the largest values that probably was connected with lead coming in composition of mining water of coal deposits.

It is difficult to arrive to any definite conclusion from these results about real impact of the accident on the level of HM pollution in the Pechora river.

Geoecological investigations in the Pechora River mouth and in adjacent Pechora Sea aquatorium started in 1991 have shown an absence of any significant pollution in the region (Ivanov et al., 1999).

It is obvious that we need in more comprehensive new researches in the Pechora river and in the Barents Sea basin as a whole.

### 3.3. Kara Sea basin

The principal source of dissolved and particulate sedimentary material influx to the Kara Sea is the discharges of the largest Arctic Rivers Ob' and Yenisey which transport annually to the Sea  $1049 \text{ km}^3$  of water and  $22.4 \cdot 10^6 \text{ t}$  of SPM, or 71%

and 67% accordingly from total water and sediment discharges (Mikhailov, 1997; Gordeev et al., 1996).

Another important source is fallout of aerosols on the Ob' and Yenisey estuaries and sea aquatorium. High coefficients of enrichment of aerosols by Cr, Cu, Sb, Pb and Ni ( $>10$  - ratio of Al - normalized content of metals in aerosols to Al - normalized content of metals in the rocks of the Earth crust) were recorded for aerosols sampled in marine aquatorium the nearest to the Kola peninsula and the Norilsk city (Schevchenko et al., 1999). It is obvious the mining-metallurgical plants on the Kola peninsula and the Norilsk smelter are the main sources of enriched by HM aerosols, parallel with aerosol material from the West Europe. Norilsk smelter pollutes not only the air, but nearest rivers and lakes also. The Schuchia River in the Lake Piasino basin is probably one of the most heavily contaminated rivers on the territory of Russia for Cu and Ni. The mean annual Cu concentration in the river close to the city Norilsk in 1981 was  $3310 \mu\text{g}\cdot\text{l}^{-1}$  (Zhulidov, Emetz, 1998). However, as shown below, in the lower course of the Yenisey River concentrations of dissolved Cu and Ni, and other HM also, practically do not exceed the background level.

Reliable data on HM concentrations in the rivers, estuaries and the aquatorium of the Kara and Laptev Seas were obtained in framework of the Program SPASIBA (see paragraph "Methodical aspect" above).

Recent results of HM determinations in water and SPM of the Ob and Yenisey Rivers are presented in Table 4. Comparison of the results of different authors shows that there are significant disagreements between them. If the concentrations of dissolved Cu and Ni after Dai and Martin, 1995, Atlas..., 1999, Shvartsev et al., 1999 are comparable (data by Kravtsov et al., 1994 on Cu are lower because these analyses determined labile dissolved forms of HM only), then Pb and Cd concentrations in Dai and Martin, 1995 are lower more than one order of value. Attract attention very large range and very high maximum concentrations of Pb, Cd and Zn concentrations in Atlas..., 1999. Higher, than in Dai and Martin, 1995, Pb and Cd concentrations in Kravtsov et al., 1994. Taking into account the analytical problem of HM determination it should be probably accepted the data by Dai and Martin, 1995 as the nearest to naturally existed in rivers concentrations.

Comparison of reliable data with global average HM concentration shows (Table 4) that there is no reason to say about heightened dissolved HM concentrations in the Ob' and Yenisey Rivers. Futhermore, the concentration of dissolved Pb, Cd and Hg in these rivers are much lower than the global average values. HM content in SPM of the Ob and Yenisey Rivers is also very similar to the mean content in the World Rivers (if do not take into account some high concentrations of Zn, Pb, Cd, Cu in Atlas..., 1999).

Two tables with "raw" data for middle course of the Ob' river (Table 5 and 6) are presented here to give an idea about distribution of HM along the river.

Interpretation of the results in both works (Shvartsev et al., 1999; Sorokovikova et al., 1999) is very scarce. Shvartsev et al., 1999 note increasing of Zn and Mn from south to north along the river. They explain it by association of metals with organic acids the concentration of which increases from south to north due to input of swamp waters to the main stream of the river.

Sorokovikova et al., 1999 mention about significant range of concentrations along the river and absence of definite tendency in this variation. Unfortunately the authors do not give any explanation of the anomaly in metal concentrations

near village Narym (extremely low concentrations of Fe, Zn and Mo and high concentration of Be).

Leonova et al., 2000 present the data for HM in the Ob' river and its tributary the Tom' river. Comparative analyses of the data has shown that the Tom' river was subject to stronger technogenic impact than the Ob' river was. Especially high concentrations of Hg were detected in the lower course of the Tom' river ( $Hg_{diss.} - 0.045-1.5 \mu\text{g/l}$ , av. 0.40,  $Hg_{part.} = 8-32 \mu\text{g/g}$  ( $n=5$ ), in comparison with the Ob' river:  $Hg_{diss.} - <0.02 \mu\text{g/l}$ ,  $Hg_{part.} - 0.4-1.7 \mu\text{g/g}$  ( $n=4$ )).

In general available information indicates an absence of more or less visible HM contamination in the lower courses of the Ob and Yenisey Rivers.

### 3.4. Laptev Sea and East - Siberian Sea basins

The largest rivers of the Laptev Sea basin - the Lena River, the Khatanga River and the Yana River deliver all together  $645 \text{ km}^3$  of fresh water,  $22.8 \cdot 10^6 \text{ t}$  of suspended matter and  $69 \cdot 10^6 \text{ t}$  of dissolved solids annually that is 86.5, 90.8 and 90.5% from total river discharge to the sea (Mikhailov, 1997; Gordeev et al., 1996). Important role play the atmospheric fallouts on the sea aquatorium (Shevchenko et al., 1999) and sea ice transported sedimentary material to the sea (Holemann et al., 1999).

The main rivers of the East-Siberian Sea basin are the Indigirka River and the Kolyma River. Water discharge and sediment discharge of two rivers are  $193 \text{ km}^3$  and  $29 \cdot 10^6 \text{ t}$  annually (Gordeev et al., 1996).

Available information about HM concentration in water and suspended sediments of the rivers mentioned is shown in Table 7.

Comparing HM concentrations in the Lena river water with the global average we see that dissolved Ni, Zn, Pb and Fe are very similar, and Cu and Cd in the Lena river are even below the global average concentrations. Concentration range in other rivers (Khatanga, Yana, Indigirka and Kolyma) is quite significant to make definite conclusion after comparison with the global average.

HM contents in the Lena river SPM do not exceed the mean contents in the World rivers. Elevated contents of As, Bi and Sb in the Yana river suspended sediments are related to granitic intrusions forming the source of Au and Sn ore-deposits in the Yana basin. High Co, Cu, Fe, Ni and V content in SPM of the Khatanga river is a result of basaltic rocks of the Siberian Trap in its basin (Rachold, 1999).

Thus the recent reliable data on HM concentrations in water and SPM of the lower streams of the largest Russian Arctic rivers don't indicate the existence of any significant anthropogenic impact in the Russian Arctic region.

## 4. Riverine fluxes of HM

The first assessments of gross river fluxes of dissolved and particulate HM by the Russian Arctic rivers were carried out by Kononov, 1959. Similar assessments were presented by Telang et al., 1991.

The first assessment of HM gross and net fluxes by the Lena river and all Arctic rivers to the Arctic Ocean based on reliable HM data were published by Guieu et al., 1996. Gordeev and Tsirkunov, 1998 presented the assessments of the input of Cu, Pb, Zn, Ni, Cd, Hg and Fe in dissolved and particulate forms by the largest

Siberian Rivers Ob, Yenisey and Lena. It is worth to note that the HM input assessments in these works, as well as in the present one must be considered as preliminary due to limited amount of reliable determinations and an absence of data on seasonal changes in concentrations.

In this work an assessment of gross river flux of HM is fulfilled on a base of analytical data of the SPASIBA programme in the following way. For dissolved HM were carried out the calculations of the average weighted on water discharge of the Ob, Yenisey and Lena Rivers concentrations. Total runoff of these three rivers is  $1574 \text{ km}^3 \cdot \text{a}^{-1}$ . It was assumed that discharge - weighted average concentrations (Cu - 1.36, Pb - 0.012, Zn - 0.74, Ni - 0.55, Cd - 0.0026, Hg - 0.0005, Fe -  $21.6 \mu\text{g} \cdot \text{l}^{-1}$ ) were representative for all other rivers of the Russian Arctic basin with total runoff  $1386 \text{ km}^3 \cdot \text{a}^{-1}$ .

For particulate form of HM similar procedure was proceeded but the sediment discharge - weighted average contents were evaluated on a base of data for five rivers already (Ob, Yenisey, Lena, Khatanga and Yana - for two last rivers the data by Rachold, 1999 were used). These contents (in ppm: C - 49, Pb - 21, Zn - 135, Ni - 42, Cd - 0.5, Hg - 0.08, Fe - 46200) were also considered as representative for all other rivers with total SPM discharge  $70 \cdot 10^6 \text{ t} \cdot \text{a}^{-1}$ .

The assessments of gross river fluxes of dissolved and suspended HM are given in Table 8.

It is clear from the table that in gross river flux the input in suspended particulate form is prevailed for all elements considered - from 58% for Cu to more than 98% for Pb and Fe.

It is interesting to compare this assessment of HM gross river flux with figures of Telang et al., 1991 for dissolved HM (in  $10^3 \text{ t/y}$ ).

|         |    | Telang et al., 1991 | This work |
|---------|----|---------------------|-----------|
| Ob'     | Cu | 2.19                | 0.85      |
|         | Zn | 8.3                 | 0.16      |
|         | Ni | 1.32                | 0.53      |
| Yenisey | Cu | 2.22                | 1.0       |
|         | Zn | 7.2                 | 0.82      |
|         | Ni | 1.67                | 0.34      |
| Lena    | Cu | 1.01                | 0.47      |
|         | Zn | 6.6                 | 0.04      |
|         | Ni | 1.01                | 0.14      |

It is obvious very big difference between two assessments of HM gross river fluxes, especially for Zn and Ni.

Some more difficult task is to assess net HM river fluxes. Net discharges are generally calculated by either a box-modelling approach or by calculations of effective river concentrations from metal - salinity relationships (GESAMP, 1987).

Behaviour of dissolved HM in mixing zone between the Lena river water and the Laptev Sea water was studied in Martin et al., 1993 and Guieu et al., 1996, in mixing zone between the Ob and Yenisey River waters and the Kara Sea water in Dai and Martin, 1995. Here were used the results of these works to evaluate the effective river concentrations and, again, assuming the runoff weighted average

effective concentrations to be representative for all other rivers of the Eurasian basin.

The calculations of net river discharge of HM to the Arctic Ocean were made (Table 9). The calculations show that net river flux exceeds gross river flux in 1.2 and 1.4 times for Cu and Ni, they are equal for Pb, and net river flux of dissolved Fe is 1/4 only from its gross river flux.

At present are absent the assessments of removal of sediment input in estuarine zones of the Arctic Rivers. Normally the losses of SPM in mixing zone reach 90-95% from gross SPM discharge (Gordeev, 1983; Martin, Windom, 1991). Taking into account that input of particulate metals exceeds significantly input of dissolved HM in total gross river discharge, then it is most probable the total net discharge (sum of net dissolved discharge and net particulate discharge) of Cu, Ni, Pb and Fe would be much lower than their total gross discharge due to removal on sedimentation and biological and physico-chemical processes in mixing zones.

## Conclusion

The review of all available information about HM concentrations in dissolved and particulate forms in the main rivers of the Russian Arctic shows that despite of a large volume of HM data beginning from 50-th, the reliable results have appeared during the last 10-15 years only and their quantities are quite limited.

All the available and reliable data on HM in water and SPM in the main Russian Arctic Rivers (Ob, Yenisey, Lena, Pechora, Khatanga et al.) indicate that in the lower courses of coastal seas the elevated HM concentration as a result of anthropogenic impact were not recorded.

On a base of these data the preliminary assessments of HM gross river fluxes and net river fluxes to the Eurasian Arctic Ocean were calculated.

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Table 1. Mean concentrations of some heavy metals in waters of the biggest Arctic Russian Rivers (in µg/l)

| River   | Cu                 | Zn                    | Ni     | Pb     | Fe                        | Reference  |
|---------|--------------------|-----------------------|--------|--------|---------------------------|--|
| Ob'     | 0-27<br>av. 3<br>6 | 0-129<br>av. 16<br>2  | -<br>1 | -<br>2 | 0-3120<br>av. 560<br>650  | Skakalski,<br>1997<br>Meinikov,<br>1991<br>Dai and<br>Martin, 1995 |
|         | 2.3                | -                     | 1.32   | 0.014  | 25.8                      |  |
| Yenisey | 0-66<br>av. 8<br>6 | 0-292<br>av. 36<br>35 | -<br>- | -<br>2 | 10-1520<br>av. 170<br>650 | Skakalski,<br>1997<br>Meinikov,<br>1991<br>Dai and<br>Martin, 1995 |
|         | 1.63               | -                     | 0.53   | 0.0055 | 14.3                      |  |
| Lena    | 0-11<br>av. 2<br>4 | 0-159<br>av. 12<br>12 | -<br>1 | -<br>1 | 0-820<br>av. 120<br>210   | Skakalski,<br>1997<br>Meinikov,<br>1991<br>Martin<br>et al., 1993  |
|         | 0.60               | 0.35                  | 0.30   | 0.016  | 22.9                      |  |

Table 2. Standard recovery (Guieu et al., 1996)

|                                   | Cu, nM    | Cd, nM    | Pb, nM    | Ni, nM    | Fe, nM    | Zn, nM    |
|-----------------------------------|-----------|-----------|-----------|-----------|-----------|-----------|
| NASS-3<br>(recommended<br>values) | 3.59±0.17 | 0.14±0.03 | 0.06±0.02 | 3.88±0.15 | 1.88±0.29 | 1.76±0.27 |
| This study<br>S1                  | 3.36      | 0.13      | 0.06      | 3.33      | 1.82      | 1.79      |
| S2                                | 3.78      | 0.14      | 0.06      | 3.35      | 1.81      | 1.91      |
| S3                                | 3.60      | 0.13      | 0.24      | 3.31      | 1.93      | 1.89      |
| Mean                              | 3.58      | 0.13      | 0.12      | 3.33      | 1.85      | 1.86      |
| Standard<br>deviation             | 0.21      | 0.01      | 0.10      | 0.02      | 0.07      | 0.06      |
| Standard<br>deviation %           | 6         | 4         | 84        | 1         | 4         | 3         |
| Recovery %                        | 100       | 94        | 201       | 86        | 99        | 106       |

Table 3. Concentration of HM's in water, suspended matter and bottom sediments of the Pechora River

| Object                        | Cu        | Zn       | Pb       | Cd       | Co       | Cr      | Ni       | V   | Reference                                      |
|-------------------------------|-----------|----------|----------|----------|----------|---------|----------|-----|--|
| Water, µg/l                   | 0.05-0.81 | 0.1-30.1 | 0.05-1.7 | 0.02-0.4 | 0.02-0.2 | 0.1-1.7 | 0.05-3.1 | -   | Atlas of contamination...; 1999 <sup>(1)</sup> |
|                               | 0.7       | 7.5      | 0.45     | 0.06     | 0.06     | 0.9     | 0.55     |     |  |
| Suspended matter, µg/g        | 60-300    | 200-500  | 100-170  | 1-100    | 10-60    | 100-300 | 10-200   | -   | -- --  |
| Bottom sediments, µg/g        | 180       | 180      | 120      | 40       | 30       | 150     | 60       |     |  |
|                               | 4.7       | 58.8     | 17.3     | 1.5      | 12.8     | -       | 30.8     | -   | Lukin and Dauval-ter, 1997                     |
| World Rivers: dissolved, µg/l | 1.5       | 0.6      | 0.03     | 0.01     | 0.1      | 1.0     | 0.5      | 1.0 | Gordeev, 1983;                                 |
| suspended, µg/g               | 80        | 250      | 35       | 0.7      | 20       | 130     | 84       | 170 | Martin and Windom, 1991                        |

<sup>(1)</sup> unfiltered water samples; average concentrations of metals in water and SPM were taken from the figures in Atlas..., 1999.

Table 4. Concentration of heavy metals in water and suspended matter of the Ob and Yenisey Rivers

| River                  | Object      | n <sup>(1)</sup> | Cu              | Zn               | Pb                    | Cd                       | Co      | Cr               | Ni                | Sn    | Fe          | Mn         | Hg <sup>(5)</sup>                     | Reference                             |
|------------------------|-------------|------------------|-----------------|------------------|-----------------------|--------------------------|---------|------------------|-------------------|-------|-------------|------------|---------------------------------------|---------------------------------------|
| Ob                     | Water, µg/l | 2                | 1.85-2.43       | -                | 0.011-0.017<br>0.014  | 0.00056-0.0009<br>0.0007 | -       | -                | 1.24-1.41<br>1.32 | -     | 25.8        | -          | -                                     | Dai and Martin, 1995                  |
|                        |             |                  | 0.01-0.34       | <0.1-0.43        | 0.038-0.106           | <0.04-0.053              | -       | -                | -                 | -     | -           | -          | -                                     | Kravtsov et al., 1994                 |
|                        | 20          | 1.0              | 7.5             | 0.50             | 0.08                  | -                        | 8.0     | -                | -                 | -     | -           | 11.1       | 0.00048-<br>0.00064<br>0.00056<br>n=2 | Shvartsev et al., 1999 <sup>(3)</sup> |
|                        | 4           | 2.7-4.6<br>3.4   | <1-4.3<br>2.5   | <0.5             | <0.05                 | <1                       | -       | -                | -                 | -     | -           | 2-8<br>4.3 | -                                     | Leonova et al., 2000                  |
| Suspended matter, µg/g |             | 0.02-5.9<br>1.3  | 0.1-46.5<br>3.5 | 0.08-9.6<br>0.65 | 0.01-0.5<br>0.08      | 0.02-0.22<br>0.04        | 0.8-4.3 | 0.03-4.5<br>0.8  | 0.02-1.2          | -     | 8.6-830     | 0.2-50     | -                                     | Atlas..., 1999                        |
|                        | 6           | 50               | 104             | 16               | 0.53                  | 19                       | 99      | 38               | -                 | -     | 60000       | 2900       | 0.044-                                | Gordeev et al., (in press)            |
| Yenisey                | Water, µg/l | -                | 100-300         | 100-660          | 50-350                | 5-80                     | 10-50   | 80-310           | 20-430            | 50-70 | 15400-65300 | 600-2700   | 0.056<br>0.05<br>n=2                  | Atlas..., 1999                        |
|                        |             | 3                | 1.41-1.85       | -                | 0.005-0.006<br>0.0055 | 0.0012-0.0018<br>0.0015  | -       | -                | 0.52-0.55<br>0.54 | -     | 14.3        | -          | -                                     | Dai and Martin, 1995                  |
|                        |             | 1.62             | 0.01-1.54       | 0.018-0.35       | <0.04-0.16            | -                        | -       | -                | -                 | -     | -           | -          | 0.00016-<br>0.00042                   | Kravtsov et al., 1994                 |
|                        | -           | 0.1-1.9<br>0.5   | 0.8-10<br>3.7   | 0.01-2.2<br>0.45 | 0.01-0.25<br>0.05     | 0.02-0.17<br>0.03        | 0.2-4.0 | 0.09-1.05<br>0.4 | 0.02-0.8          | -     | 8.2-250     | 0.2-75     | 0.0003<br>n=3                         | Atlas..., 1999 <sup>(4)</sup>         |
| Suspended matter, µg/g | 6           | 144              | 220             | 30               | 2.2                   | 23                       | 130     | 77               | -                 | -     | 58000       | 1900       | 0.040-                                | Gordeev et al. (in press)             |
|                        | -           | 50-750           | 100-550         | 200-530          | 10-30                 | 10-80                    | 80-280  | 10-540           | 50-200            | -     | 22300-55500 | 1900-18100 | 0.060<br>0.05<br>n=3                  | Atlas..., 1999                        |
|                        |             | 350              | 320             | 350              | 20                    | 50                       | -       | 240              | -                 | -     | -           | -          | -                                     |                                       |

(<sup>1</sup>) - number of samples, "-" - number is not known; (<sup>2</sup>) - range of concentrations and average; (<sup>3</sup>) - middle reach of the Ob river from its tributary the Tom' river to city Nijnevartovsk; (<sup>4</sup>) - unfiltered samples; (<sup>5</sup>) - Coquery et al., 1995.

Table 5. concentration of HM in the Ob' river water in 1999 ( $\mu\text{g/l}$ ) (Shvartsev et al., 1999)

| Station <sup>(1)</sup> | Hg    | Cd   | Pb   | Cu   | Zn   | Mn   | Cr   | Mo   | Ag   |
|------------------------|-------|------|------|------|------|------|------|------|------|
| 1                      | 0.10  | <0.1 | 0.3  | 1.0  | 1.5  | 5.4  | 4.4  | 0.4  | 0.1  |
| 2                      | 0.10  | <0.1 | 0.3  | 1.0  | 1.5  | 6.4  | 5.9  | 0.3  | <0.1 |
| 3                      | 0.13  | <0.1 | <0.1 | 0.5  | 0.4  | 10.3 | 7.0  | 0.4  | <0.1 |
| 4                      | 0.05  | 0.16 | 0.3  | 0.8  | 2.0  | 11.0 | 7.7  | 0.6  | <0.1 |
| 5                      | 0.06  | <0.1 | 0.2  | 0.7  | 1.0  | 9.0  | 9.5  | 0.4  | <0.1 |
| 6                      | 0.07  | 0.13 | 0.2  | 0.5  | 2.6  | 9.4  | 10.1 | 0.4  | <0.1 |
| 7                      | 0.07  | <0.1 | 2.3  | 2.0  | 1.7  | 11.3 | 10.0 | 0.4  | <0.1 |
| 8                      | 0.22  | <0.1 | <0.1 | 1.0  | 1.0  | 15.0 | <0.1 | <0.1 | 0.3  |
| 9                      | 0.03  | <0.1 | <0.1 | <0.1 | 1.0  | 12.9 | 9.6  | 0.6  | <0.1 |
| 10                     | 0.10  | <0.1 | 0.3  | 0.3  | 3.0  | 11.8 | 8.5  | 0.9  | <0.1 |
| 11                     | 0.16  | 0.20 | 0.7  | 1.0  | 5.0  | 14.1 | 10.8 | 0.6  | <0.1 |
| 12                     | 0.03  | <0.1 | 0.3  | 0.8  | 3.5  | 9.0  | 9.6  | 0.4  | <0.1 |
| 13                     | 0.02  | <0.1 | 1.0  | 1.2  | 2.0  | 10.7 | 7.4  | <0.1 | <0.1 |
| 14                     | 0.04  | <0.1 | <0.1 | 0.5  | 0.5  | 10.5 | 7.2  | 0.3  | <0.1 |
| 15                     | 0.16  | <0.1 | 0.3  | 0.6  | 42.1 | 14.2 | 16.6 | 0.9  | 0.1  |
| 16                     | 0.09  | 0.1  | 1.6  | 1.5  | 24.7 | 13.3 | 10.0 | 1.0  | 0.2  |
| 17                     | 0.21  | 0.20 | 0.5  | 1.7  | 24.2 | 18.6 | 15.3 | 0.6  | <0.1 |
| 18                     | 0.16  | <0.1 | 0.3  | 1.2  | 1.5  | 14.2 | 8.9  | <0.1 | <0.1 |
| 19                     | 0.06  | 0.10 | 0.7  | 2.0  | 28.8 | 7.6  | 3.2  | <0.1 | <0.1 |
| 20                     | <0.01 | 0.14 | 0.4  | 2.0  | 2.0  | 7.1  | <0.1 | <0.1 | 0.1  |
| Af <sup>(2)</sup>      | 0.12  | 0.20 | 0.8  | 2.1  | 54.5 | 18.9 | 2.9  | -    | 0.1  |

<sup>(1)</sup> - stations are distributed from south to north;

<sup>(2)</sup> - average for the middle Ob' river (Shvartsev et al., 1996)

Table 6. HM variations in the Ob' river, July 1999 (Sorokovikova et al., 1999)

| Element,<br>µg/l | Below<br>r. Tom' | Below<br>c.Kolpashevo | v.Narym | v.Pyrgino | Below<br>c.Nijnevarovsk | c. Surgut |
|------------------|------------------|-----------------------|---------|-----------|-------------------------|-----------|
| Be               | 0.029            | 0.032                 | 27.5    | 0.040     | 0.034                   | 0.029     |
| Cr               | 23.3             | 23.1                  | 1.7     | 26.6      | 19.7                    | 18.6      |
| Fe               | 86.5             | 91.6                  | 0.092   | 549       | 97.1                    | 91.5      |
| Al               | 13.1             | 12.2                  | 14.1    | 88.1      | 12.8                    | 10.6      |
| Ni               | 1.2              | 1.4                   | 1.4     | 2.3       | 1.2                     | 1.1       |
| Zn               | 3.9              | 1.6                   | 0.017   | 15.2      | 4.8                     | 4.1       |
| Mo               | 2.0              | 0.75                  | 0.084   | 3.1       | 1.2                     | 1.0       |
| Cd               | 0.23             | 0.19                  | 1.00    | 0.25      | 0.16                    | 0.11      |
| Pb               | 0.28             | 0.30                  | 0.043   | 1.04      | 0.50                    | 0.41      |

Table 7. Concentration of heavy metals in water and suspended matter of the rivers in the Laptev Sea and East - Siberian Sea basins

| River          | Object                       | n       | Cu                | Zn                | Pb                   | Cd                      | Co        | Ni                | Sn          | As   | Bi   | Sb   | V   | Mo   | Fe                | Mn                | Reference                      |   |
|----------------|------------------------------|---------|-------------------|-------------------|----------------------|-------------------------|-----------|-------------------|-------------|------|------|------|-----|------|-------------------|-------------------|--------------------------------|---|
| Lena           | Water,<br>µg/l               | 2       | 0.47-0.74<br>0.60 | 0.33-0.37<br>0.35 | 0.015-0.019<br>0.017 | 0.0022-0.0090<br>0.0056 | -         | 0.23-0.38<br>0.30 | -           | 0.15 | -    | -    | -   | -    | 21.6-24.3<br>22.9 | -                 | Martin<br>et al., 1993         |   |
|                |                              | 6       | 0.78-1.0<br>0.88  | 0.08-1.37<br>0.45 | 0.031-0.083<br>0.041 | 0.0022-0.012<br>0.006   | -         | 0.25-0.35<br>0.30 | -           | -    | -    | -    | -   | -    | -                 | 22.4-48.5<br>35.8 | -                              | Guiou<br>et al., 1996                   |
|                | -                            | 0.1-1.5 | 0.1-10            | 0.10-0.56         | 0.02-0.5             | 0.02-0.05               | 0.02-1.1  | 0.02-0.1          | -           | -    | -    | -    | -   | -    | 11.3-200          | 0.1-8.2           | Atlas...,<br>1999 <sup>†</sup> |   |
|                | Suspended<br>matter,<br>µg/g | 2       | 24-31<br>28       | 106-181<br>143    | 20-26<br>23          | -                       | -         | -                 | 27-35<br>31 | -    | -    | -    | -   | -    | -                 | -                 | -                              | Martin<br>et al., 1993                  |
|                |                              | 6       | 28                | 160               | 36                   | 0.25                    | 13        | 34                | -           | -    | -    | -    | -   | -    | -                 | 33200             | 1250                           | Gordeev<br>and<br>Shevchen-<br>ko, 1995 |
| Khatan-<br>ga  | Water,<br>µg/l               | 31      | 35                | 141               | 24                   | 0.65                    | 18        | 53                | 1.9         | 9.1  | 0.24 | 0.57 | 97  | 1.23 | 38000             | 1600              | Rachold,<br>1999               |   |
|                |                              | 5       | 26-56<br>42       | 156-217<br>185    | 31-59<br>42          | 0.14-1.42<br>0.96       | -         | 24-80<br>42       | -           | -    | -    | -    | -   | -    | 48400             | 526-1295<br>972   | Nolting<br>et al., 1996        |   |
|                | -                            | 100-600 | 100-430           | 110-200           | 2-3                  | 10-70                   | 20-410    | <50               | -           | -    | -    | -    | -   | -    | 25000-74300       | 700-15300         | Atlas...,<br>1999              |   |
|                | Susp.mat<br>µg/g             | -       | 0.2-15            | 1.1-18            | 0.08-0.3             | 0.03-0.2                | 0.02-0.5  | 0.06-0.9          | 0.03-0.2    | -    | -    | -    | -   | -    | -                 | 4.8-49            | 0.4-2.6                        | Atlas...,<br>1999 <sup>†</sup>          |
|                |                              | 12      | 82                | 104               | 12                   | 0.22                    | 35        | 84                | 1.6         | 9.3  | 0.13 | 0.43 | 349 | 0.88 | 61250             | 1200              | Rachold,<br>1999               |   |
| Yana           | Susp.mat<br>µg/g             | 7       | 30                | 130               | 23                   | 0.32                    | 17        | 39                | 1.5         | 26.7 | 0.23 | 2.1  | 110 | 0.96 | 38000             | 750               | Rachold,<br>1999               |   |
| Indigir-<br>ka | Water,<br>µg/l               | -       | 0.7-2.6           | 0.6-8.2           | 0.16-0.20            | 0.06-0.14               | 0.02-0.05 | 0.5-0.9           | 0.02-0.5    | -    | -    | -    | -   | -    | 5.9-22            | 0.05-1.2          | Atlas...,<br>1999 <sup>†</sup> |   |
|                |                              | -       | 0.05-2.1          | 0.1-10.4          | 0.02-0.35            | 0.01-0.4                | 0.01-0.24 | 0.03-1.8          | 0.02-0.8    | -    | -    | -    | -   | -    | -                 | 0.1-47            | 0.05-5.72                      | Atlas...,<br>1999 <sup>†</sup>          |
| Koly-<br>ma    | Susp.mat<br>µg/g             | -       | 100-1000          | 100-250           | 80-260               | 1-20                    | 10-80     | 30-350            | <50         | -    | -    | -    | -   | -    | 21800-67700       | 2200-24000        | Atlas...,<br>1999              |   |
|                |                              | -       | 1.5               | 0.6               | 0.03                 | 0.01                    | 0.1       | 0.5               | -           | 1.7  | -    | 1.0  | 1.0 | 0.5  | 40                | 10                | Gordeev,<br>1983;              |   |
| Global         | Water,<br>µg/l               | -       | 1.5               | 0.6               | 0.03                 | 0.01                    | 0.1       | 0.5               | -           | 1.7  | -    | 1.0  | 1.0 | 0.5  | 40                | 10                | Gordeev,<br>1983;              |   |

| River average | Object   | n | Cu | Zn  | Pb | Cd  | Co | Ni | Sn | As | Bi | Sb  | V   | Mo  | Fe    | Mn   | Reference                |
|---------------|----------|---|----|-----|----|-----|----|----|----|----|----|-----|-----|-----|-------|------|--------------------------|
|               | Susp.mat | - | 80 | 250 | 35 | 0.7 | 20 | 84 | -  | 5  | -  | 2.0 | 130 | 3.0 | 51000 | 1100 | Martin and Gordeev, 1986 |
|               | µg/g     |   |    |     |    |     |    |    |    |    |    |     |     |     |       |      |                          |

(1) - unfiltered water samples

Table 8. Gross river flux of dissolved and particulate heavy metals and iron to the Eurasian Arctic ( $10^3 \text{t} \cdot \text{a}^{-1}$ )

| River basin           | type of flux | Cu   | Pb    | Zn   | Ni   | Cd     | Hg     | Fe   |
|-----------------------|--------------|------|-------|------|------|--------|--------|------|
| Ob                    | diss.        | 0.85 | 0.006 | 0.16 | 0.53 | 0.0003 | 0.0002 | 12   |
|                       | part.        | 0.84 | 0.26  | 1.7  | 0.63 | 0.0033 | 0.0008 | 940  |
|                       | total        | 1.6  | 0.266 | 1.86 | 1.16 | 0.0036 | 0.001  | 952  |
| Yenisey               | diss.        | 1.0  | 0.004 | 0.82 | 0.34 | 0.001  | 0.0002 | 10   |
|                       | part.        | 0.65 | 0.18  | 1.3  | 0.45 | 0.0136 | 0.0003 | 320  |
|                       | total        | 1.65 | 0.184 | 2.12 | 0.79 | 0.0146 | 0.0005 | 330  |
| Lena                  | diss.        | 0.47 | 0.009 | 0.04 | 0.14 | 0.0032 | 0.0004 | 19   |
|                       | part.        | 0.49 | 0.40  | 2.50 | 0.55 | 0.0046 | 0.0040 | 590  |
|                       | total        | 0.96 | 0.41  | 2.54 | 0.69 | 0.0078 | 0.0044 | 610  |
| Whole Eurasian Arctic | diss.        | 4.04 | 0.036 | 2.18 | 1.93 | 0.0077 | 0.0015 | 64   |
|                       | part.        | 5.66 | 2.41  | 15.6 | 4.85 | 0.058  | 0.0088 | 5320 |
|                       | total        | 9.7  | 2.45  | 17.8 | 6.78 | 0.066  | 0.010  | 5384 |

Table 9. Net river flux of dissolved heavy metals and iron to the Eurasian Arctic ( $10^3\text{t}\cdot\text{a}^{-1}$ )

| River basin           | Cu   | Pb     | Ni   | Fe   |
|-----------------------|------|--------|------|------|
| Ob                    | 0.81 | 0.0038 | 0.50 | 2.3  |
| Yenisey               | 1.05 | 0.0031 | 0.58 | 2.8  |
| Lena                  | 0.67 | 0.009  | 0.37 | 3.6  |
| Whole Eurasian Arctic | 4.77 | 0.030  | 2.75 | 16.3 |

6.1.5 *Lars Asplin*

# **Modelling of pollutant transport in the marine environment**

Lars Asplin,  
Institute of Marine Research, Bergen

**Assumptions:**

- **Transport of water masses (not pollutants)**
- **The Barents Sea**

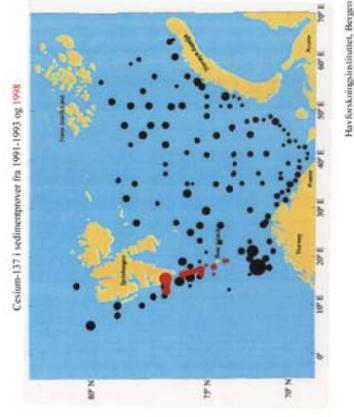
**Outline:**

- **IMR activities in the Barents Sea**
- **Validation of marine models**
- **Atmospheric forcing of marine models**
- **(Examples)**

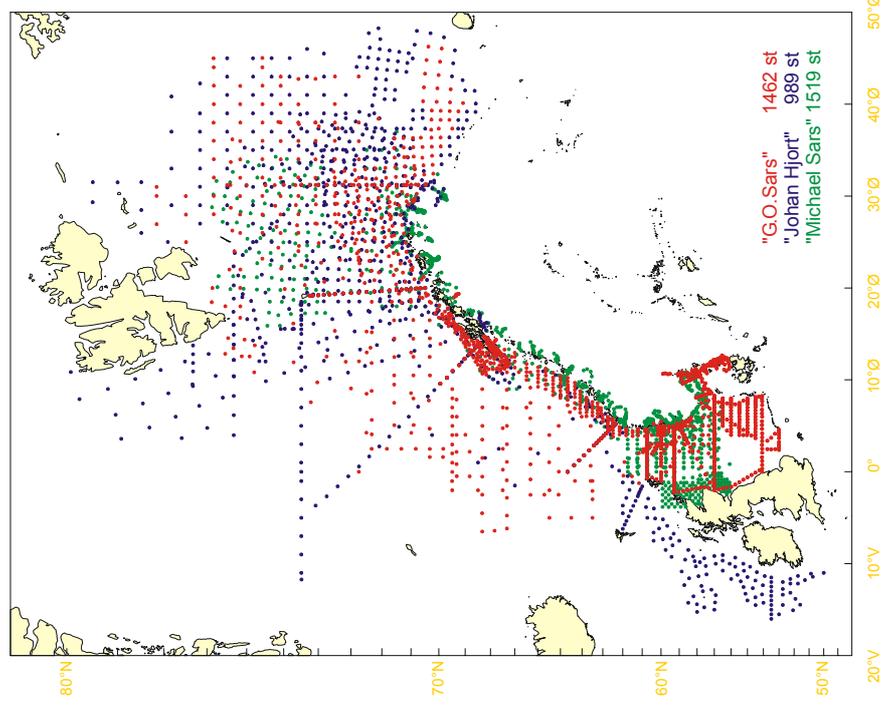
# IMR and the Barents Sea

- **Considerable field activities**  
(currents, hydrography, water quality, plankton, fish!)
- **Numerical models**  
(20km - 4km - 800m and 200 m resolution. General circulation.)

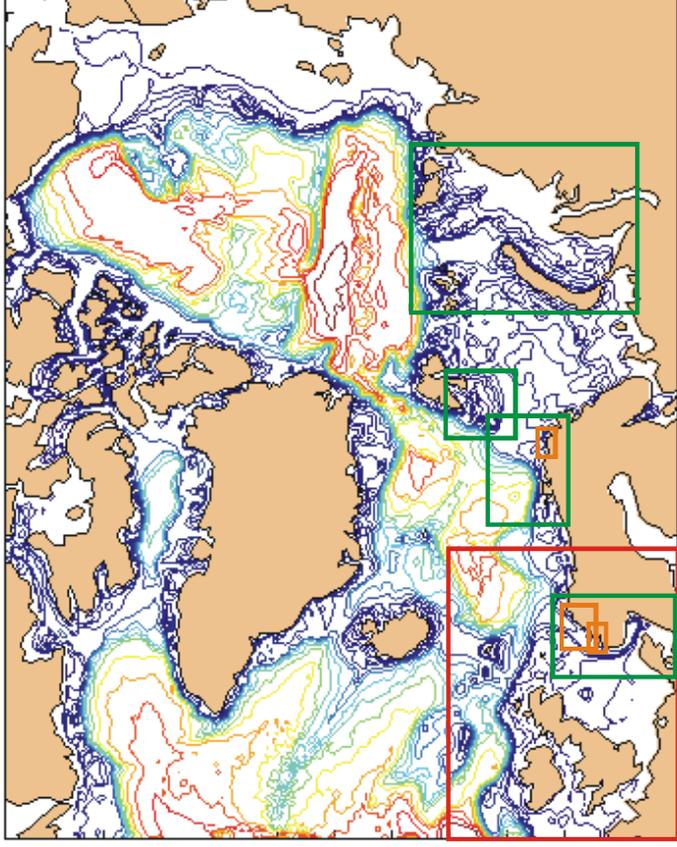
**IMR performs  
70-80% of all  
field measurements  
in the Norwegian  
waters each year**



CTD stasjoner 2000  
3970 stasjoner



## Numerical model implementations in Norwegian waters (resolution 20km - 200m)



## **Validation of marine models**

### **Dick P. Dee (1995):**

”Validation of a computational model is the process of formulating and substantiating explicit claims about the applicability and accuracy of computational results, with reference to the intended purpose of the model as well as to the natural system it represents.”

## **Validation of marine models**

**Dick P. Dee (1995):**

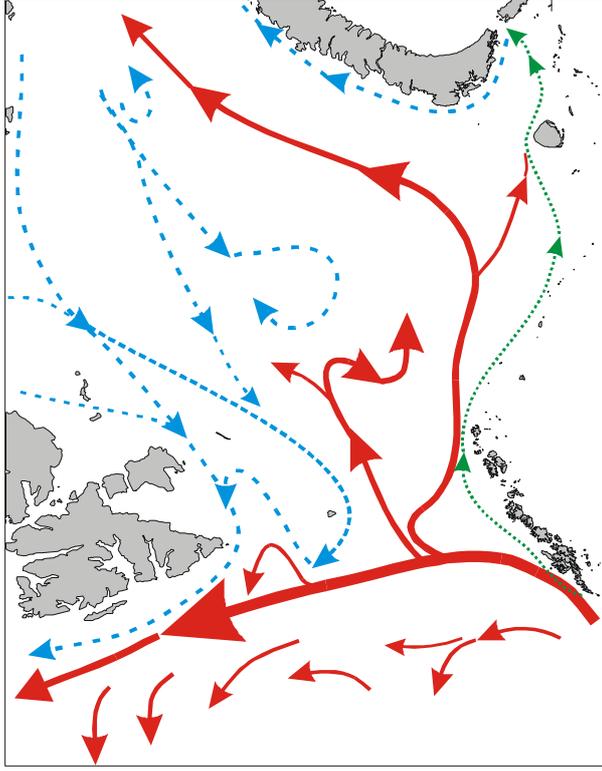
”Validation of a computational model is the process of formulating and substantiating explicit claims about the applicability and accuracy of computational results, with reference to the intended purpose of the model as well as to the natural system it represents.”

- **Validation of a numerical model is a never ending process**
- **A validated numerical model does not necessarily mean that the results are correct, but there exist a measure of the quality of the results**

**To find objective methods for numerical model validation is non-trivial**

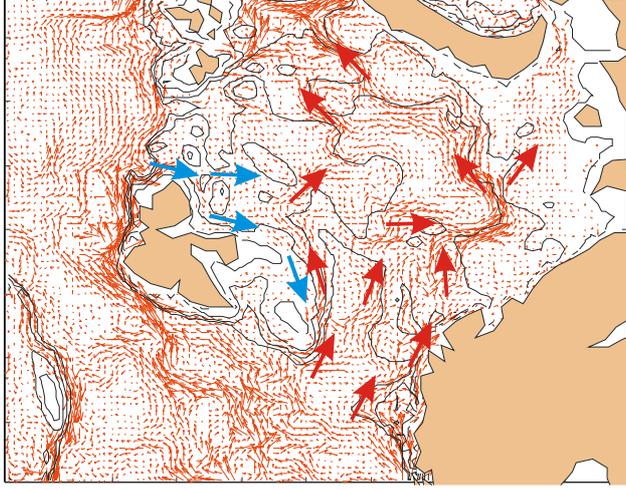
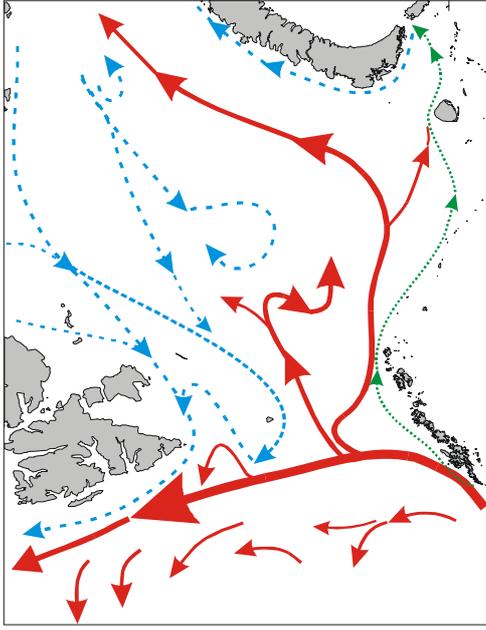
# IMR-VEINS: Transport of water through the Barents Sea. Validation 1.

Assumed current system in the Barents Sea



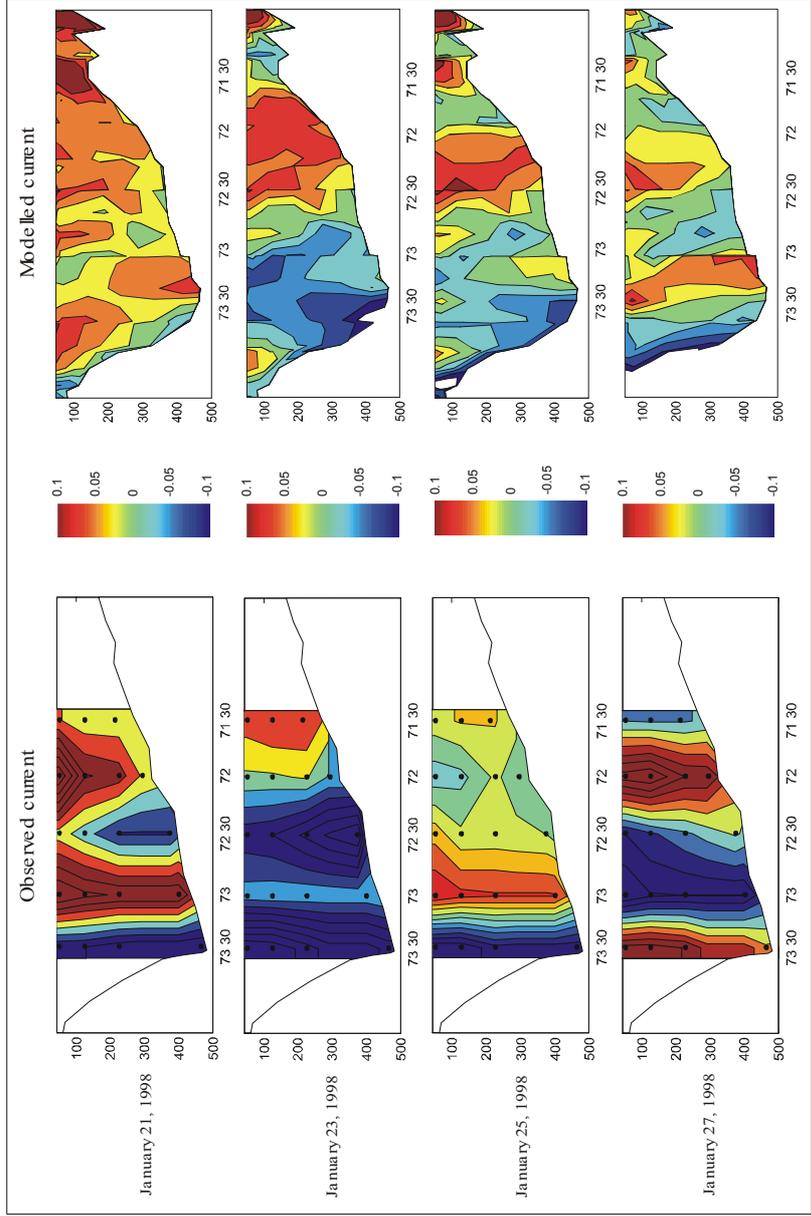
**IMR-VEINS:**  
Transport of water through the Barents Sea. Validation 1.

Assumed current system in the Barents Sea

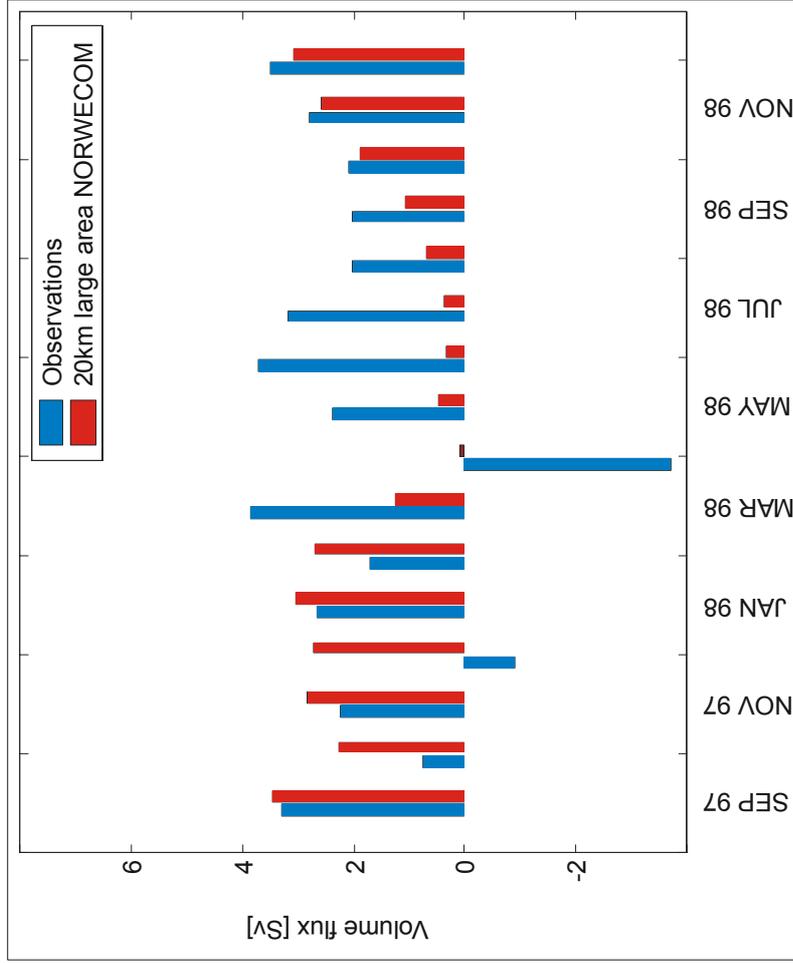


Simulated winter mean currents

# IMR-VEINS: Transport of water through the Barents Sea. Validation 2.



### IMR-VEINS: Transport of water through the Barents Sea. Validation 3.



Mean volume fluxes: obs = 1.68 Sv, sim. = 1.65 Sv

## **IMR-VEINS:**

**Transport of water through the Barents Sea.**

**Conclusions of model validation:**

- **The model results capture in general the features shown by the available observations and existing knowledge**
- **The mean values are reasonably close, and also the variability (...)**

**However:**

**In periods (weeks), there can be large discrepancies between the observed flow fields and the simulated flow...**

**Hence:**

**Although it is possible to achieve much information from marine models of today, we still need better modelling tools (resolution, forcing) and yet more and better observations!**

## **Atmospheric forcing of marine models**

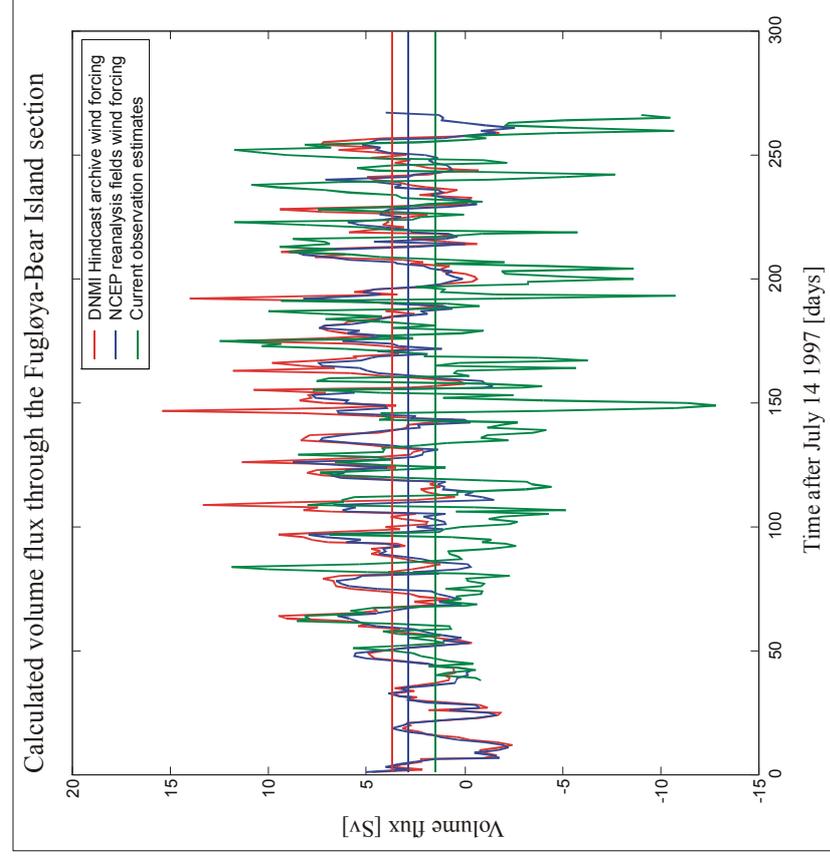
**At present marine models typically use coarse scale reanalysed atmospheric fields as forcing (DNMI Hindcast, NCEP, ECMWF):**

**Resolution > 75 km, 6 hours**

- **The wind fields can differ both quantitatively and qualitatively**
- **The wind fields might lack important unresolved features**

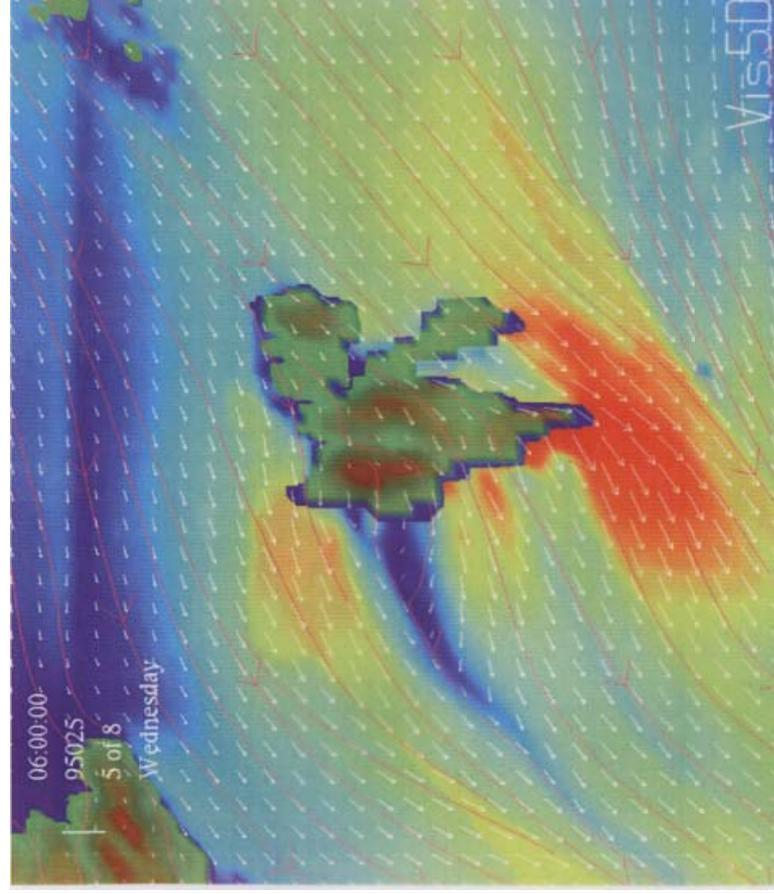
## Atmospheric forcing of marine models

### Differences in volume flux estimates from DNMI and NCEP forcing



## Atmospheric forcing of marine models

**Importance of meso scale dynamics: Easterly winds across Spitsbergen simulated with NORLAM (10 km horizontal resolution, 30 vertical layers)**



From: Paul Skeie & Sigbjørn Grønås (Geophysical Institute, UiB) Tellus (2000)

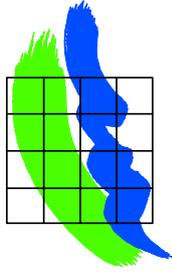
## Examples of marine model results:

- Spreading of water masses from the Kursk position
- Transport of Atlantic water through the Barents Sea
- Spreading of particles from Sellafield (or nearby...)
- Spreading of water in Sognefjorden (high resolution fjord model)

## Individual Presentations - Session II:

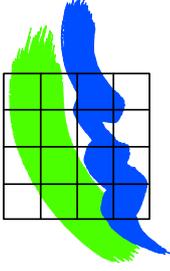
### 6.2 Individual Presentations – Session II

#### 6.2.1 *Jesper H. Christensen*



# Atmospheric modelling activities inside the Danish AMAP program

Jesper H. Christensen  
NERI-ATMI, Frederiksborgvej 399  
4000 Roskilde

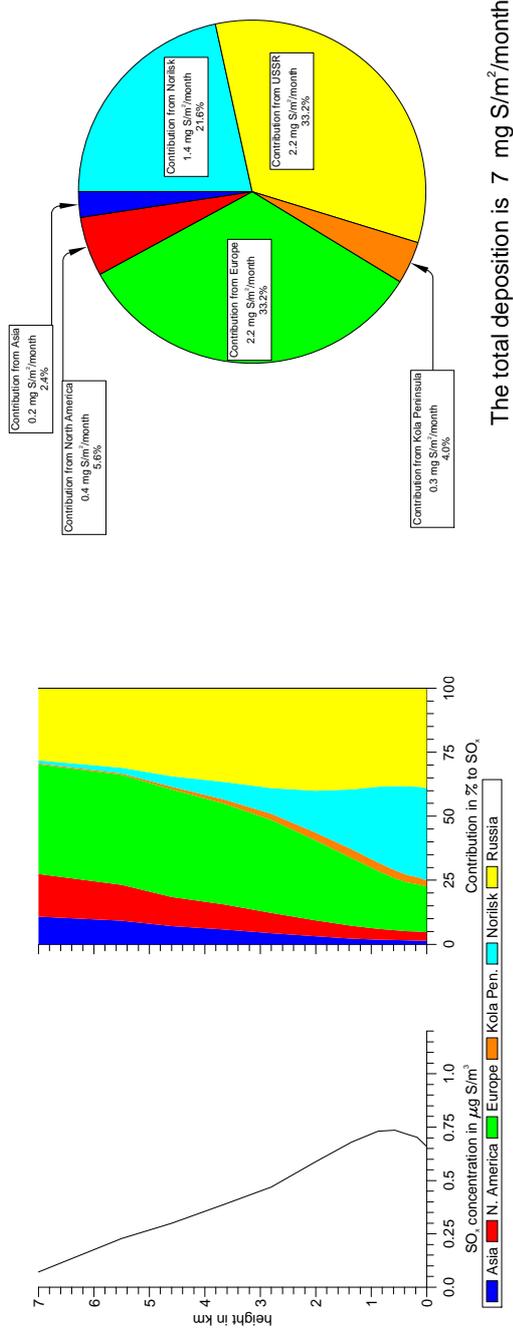


## The Danish Eulerian Hemispheric Model (DEHM) System

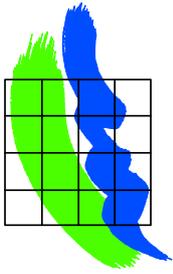
- The model work is financially supported by the Danish Environmental Protection Agency with means from the MIKA/DANCEA funds for Environmental Support to the Arctic Region
- It is a part of the Danish contribution to the international AMAP programme
- Purpose: Study the long-range transport in the troposphere of pollutants into the Arctic
- Developed since 1990. In the beginning only for Sulphur, later Lead and now also with a full photochemical scheme and Mercury.

# The Danish Eulerian Hemispheric Model in 1. Phase of AMAP

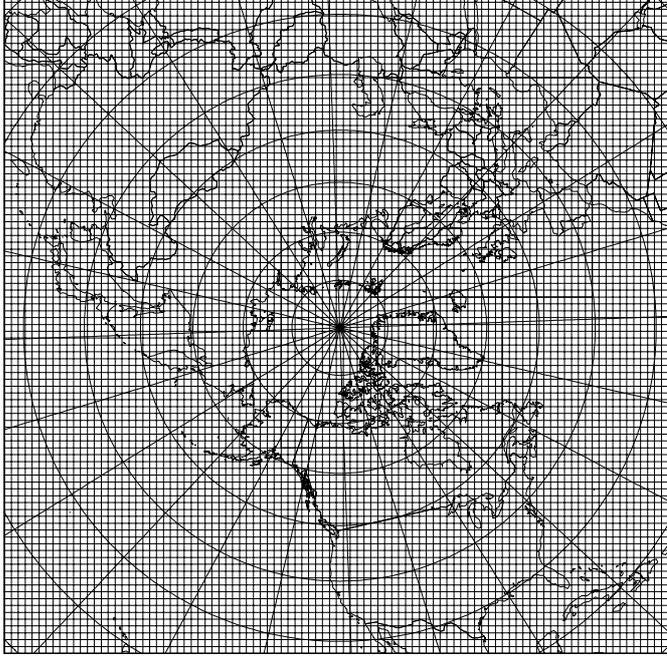
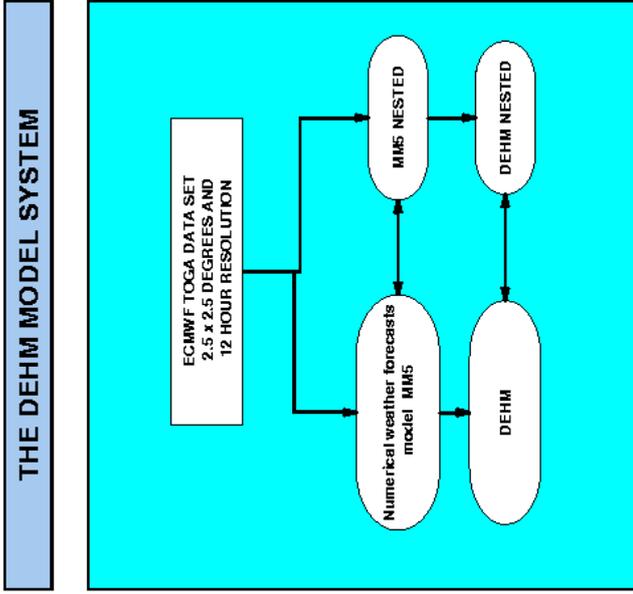
- Direct coupling to ECMWF data, no MM5 meteorological preprocessor
- Simplified linear sulphur chemistry

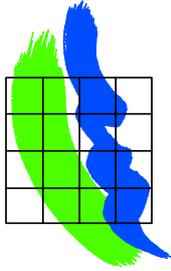


The total deposition is 7 mg S/m<sup>2</sup>/month



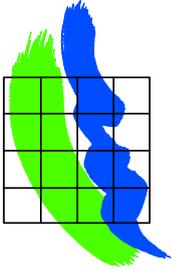
# The Danish Eulerian Hemispheric Model (DEHM) System



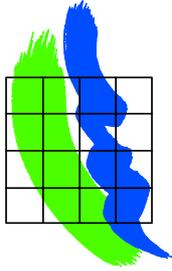


## MM5 model

- Hydrostatisk version (version 2)
- 150 km resolution at 60° N, 50 km for nested domain
- 97x97 horizontal grid-points (for mother domain and 100x100 horizontal gridpoints for nested domain) and 20 vertical layers
- Mixed Phase (Reisner) explicit moisture
- Betts-Miller cumulus parametrization
- MRF boundary layer parametrization with 5 layer soil model
- Cloud-radiation scheme



- Input data:
  - Met data from ECMWF,  $2.5^\circ \times 2.5^\circ$  lat-lon, 12 hour resolution, 21 years data from 1979 to 2000
- Output every 3 hours
- Only run for 1990 to 2000 for hemispheric domain and for 1995 and 1998 to 2000 for Europe (50 km) 1 month for Greenland (50 km) as demonstration

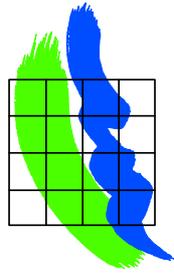


# The Danish Eulerian Hemispheric Model

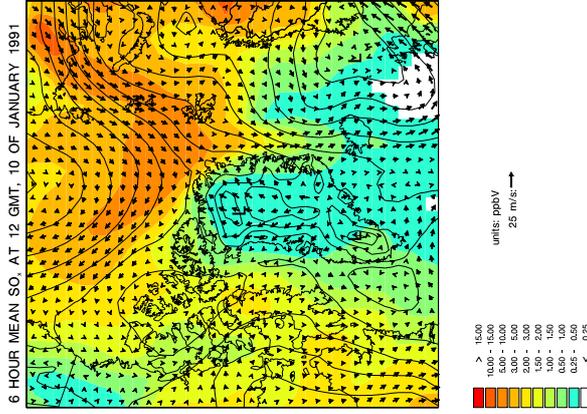
- Full three dimensional advection-diffusion equations
- 150 km grid resolution (Mother domain)
- 20 vertical levels up to 16 km
- Dry deposition based on the resistance method with 8 different surfaces
- Wet deposition based on scavenging coefficients

## Numerical methods:

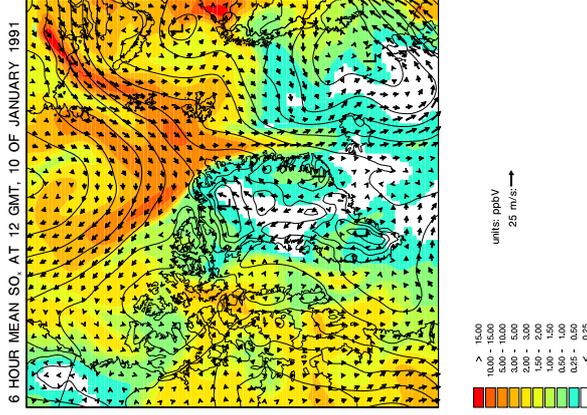
- Horizontal advection: *Accurate Space Derivatives with non-periodic boundary conditions and 2-way nesting capabilities*
- Vertical advection: *Finite Elements*
- Diffusion: *Finite Elements*



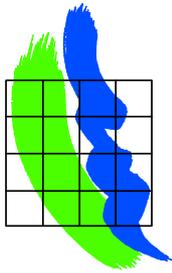
# Nested version of DEHM a demonstration with the simplified sulphur version



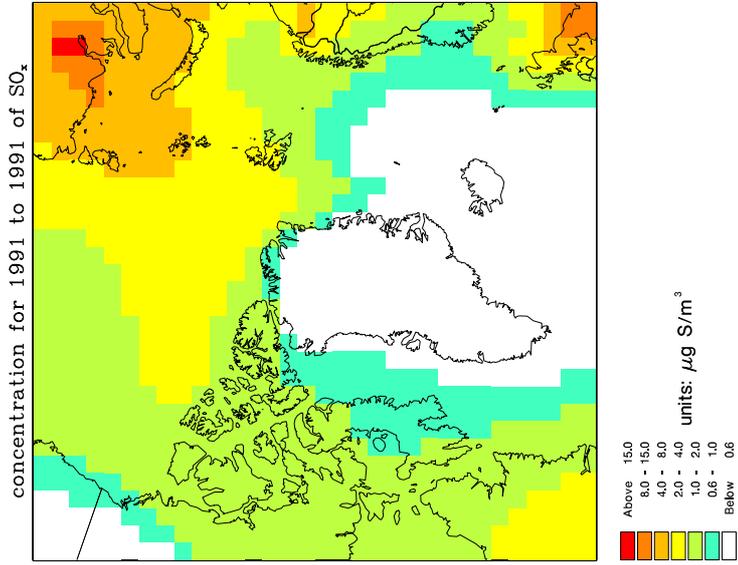
150 km resolution



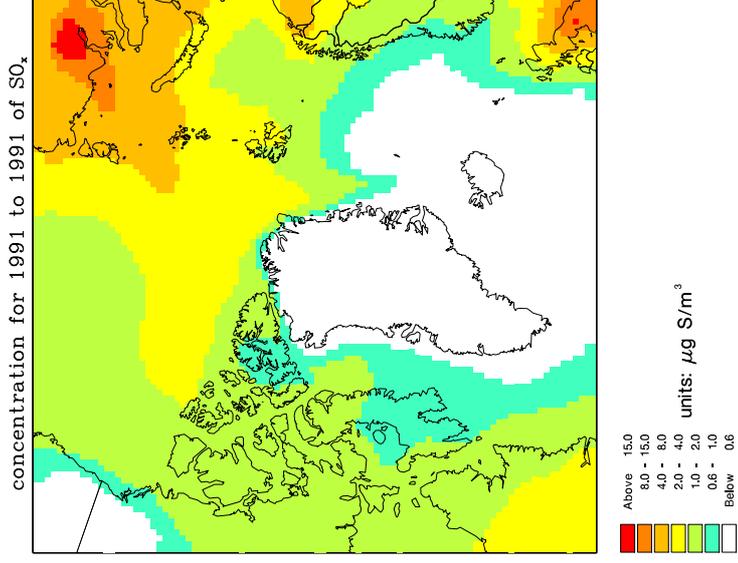
50 km resolution



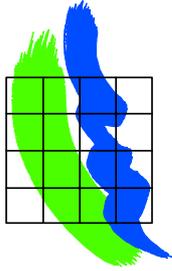
# The monthly mean concentrations for SO<sub>x</sub>



150 km resolution

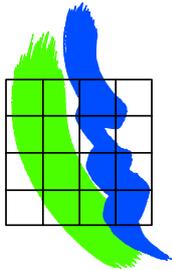


50 km resolution



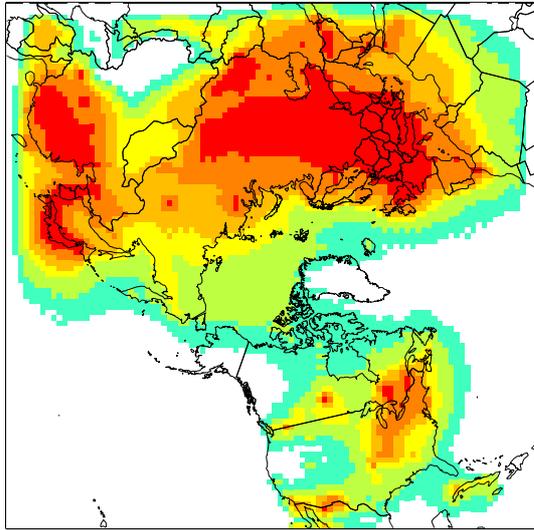
## Lead version of DEHM

- Based on current version of DEHM with simplified sulphur chemistry
- GEIA Global Lead Emissions Inventory, Version 1, for 1989
- removal parametrizations equal to  $\text{SO}_4^{2-}$

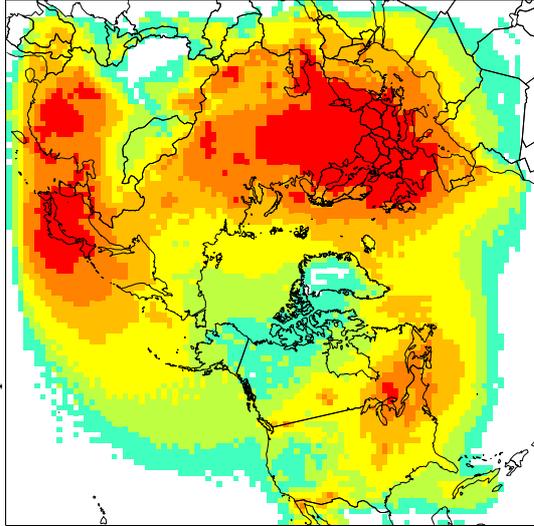


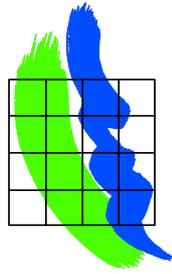
# Lead version of DEHM

concentration for 1990 to 1997 of Pb

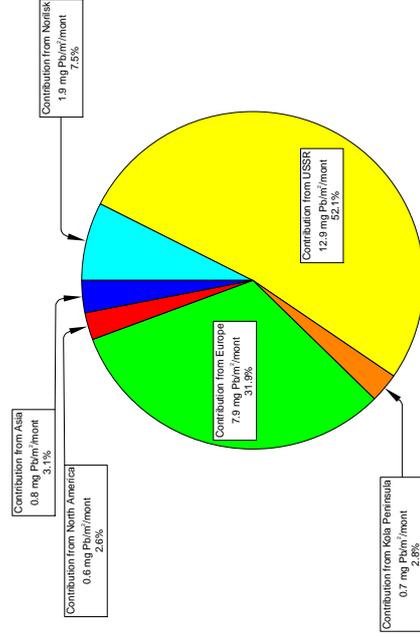
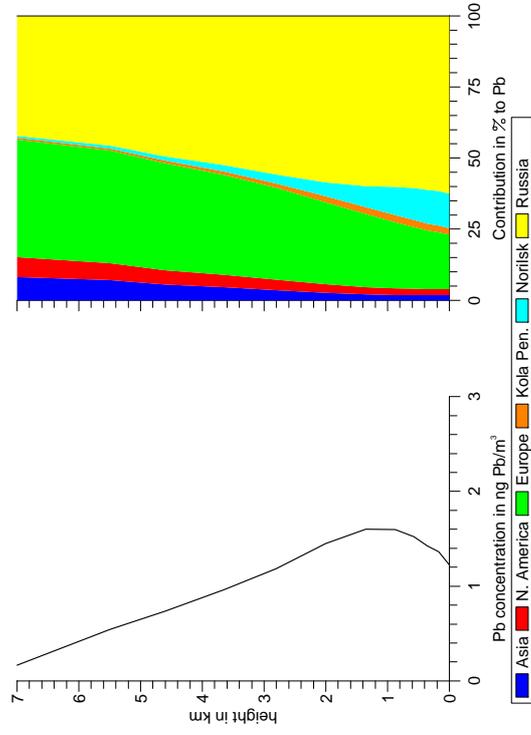


Total deposition of Pb for 1990 to 1996

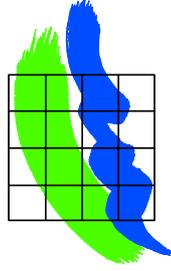




# Lead version of DEHM

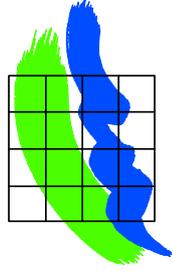


The total deposition is 25  $\mu\text{g Pb/m}^2/\text{month}$



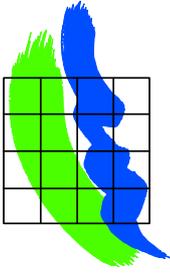
## Photochemical version of DEHM

- Pollutants: 54 species, more than 110 chemical reactions (17 photolyse, 94 gas-phase and 2 aqueous phase reactions), chemistry scheme similar to the EMEP oxidant model
- Emissions: Global GEIA emissions of anthropogenic emissions of SO<sub>x</sub> and NO<sub>x</sub>, NO<sub>x</sub> from lightning and soil and Isoprene from vegetation, all on 1°x 1°
- global EDGAR inventory on 1°x 1° for anthropogenic hydrocarbons
- SO<sub>x</sub> and NO<sub>x</sub> for Europe from EMEP
- Has been run for whole 1998

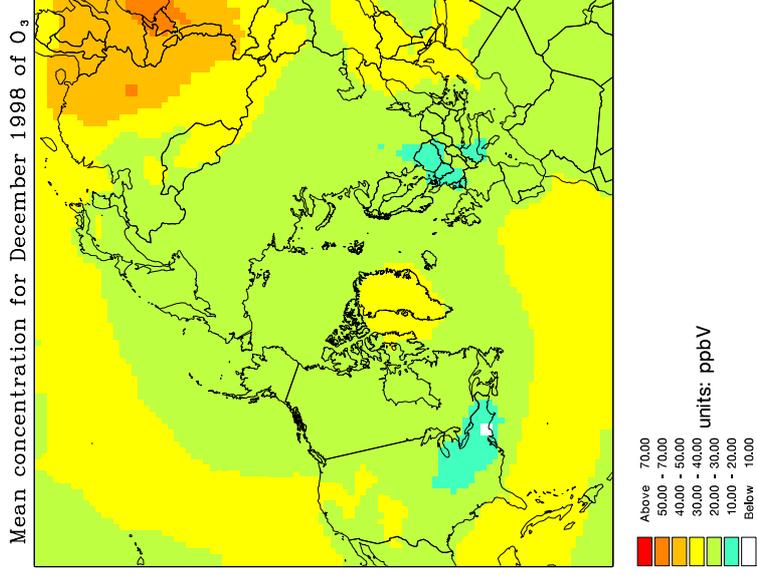
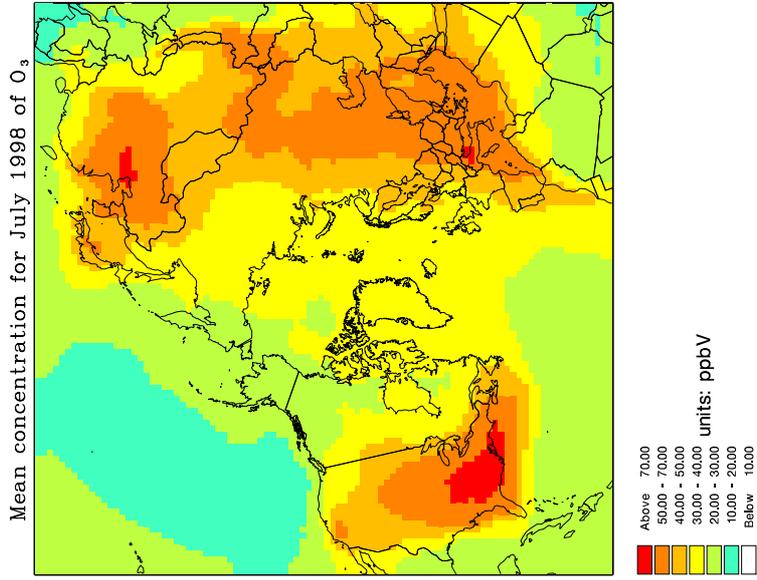


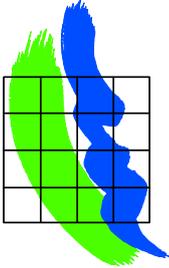
## Purpose with the photochemical version

- Improvement of the parameterization of the chemistry compared to the simple sulfur model
- Provided necessary input concentrations ( $O_3$ ,  $SO_2$ ) for the Mercury model
- Be a useful contribution for the understanding of the atmospheric chemistry in the Arctic, especially during the Polar Sunrise in connection with field measurements

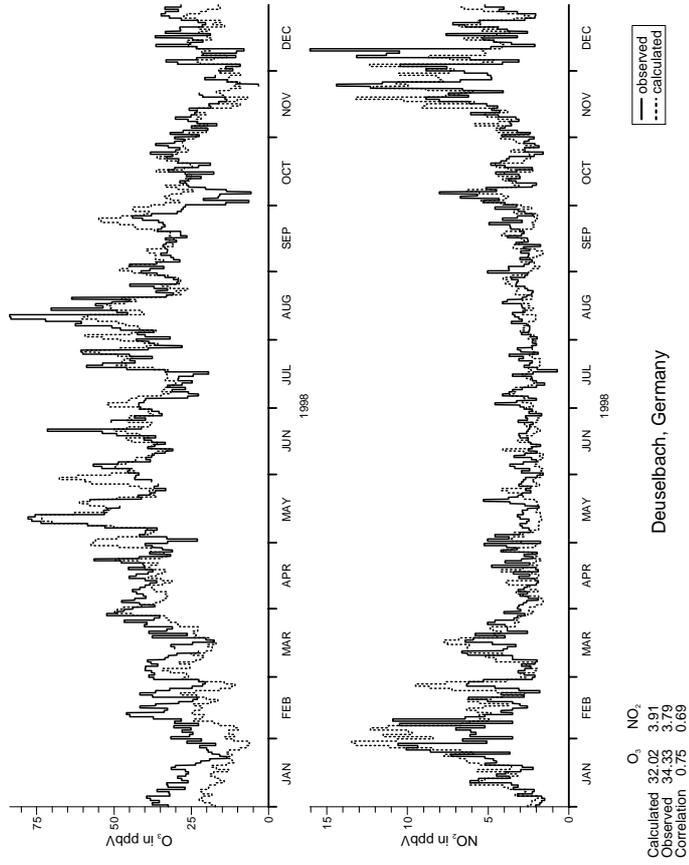


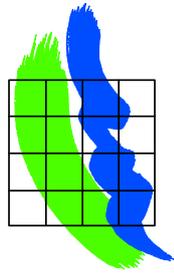
# Ozone mean concentrations



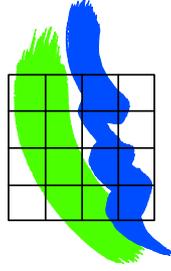


# Some validation



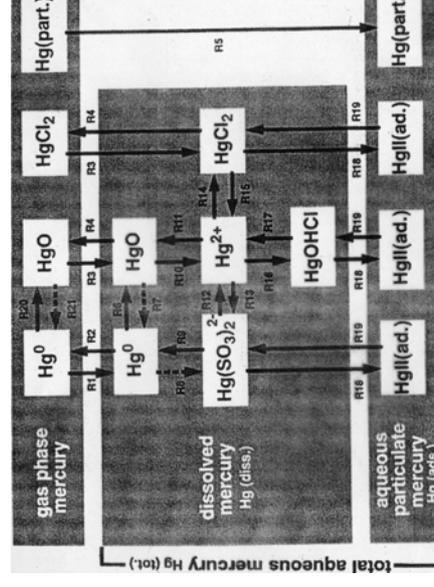


## Ozone in the Arctic

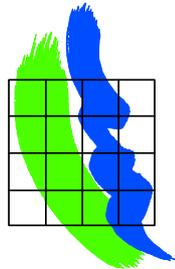


## Mercury version of DEHM

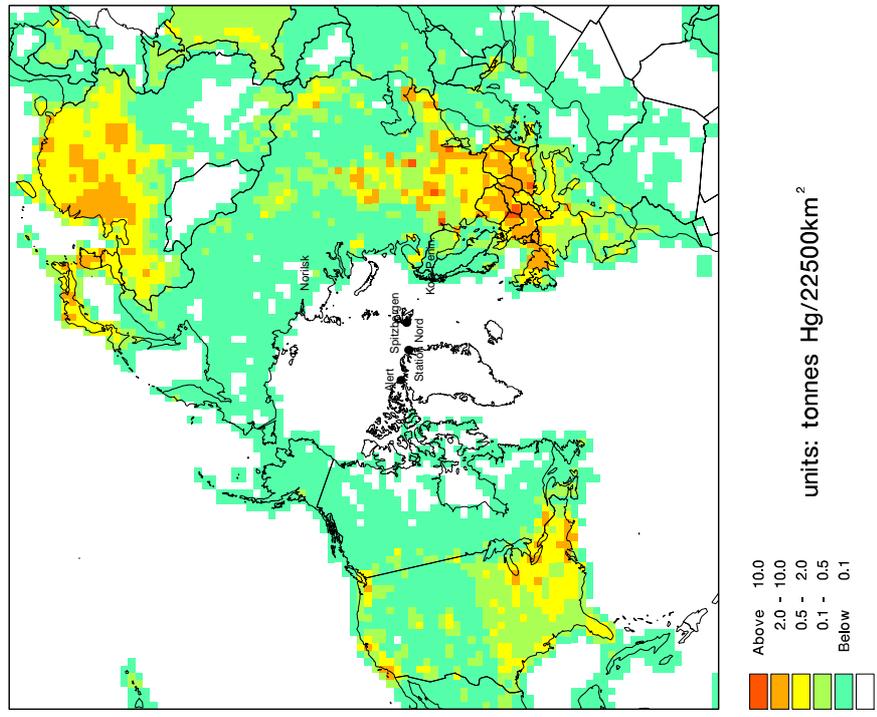
- Mercury model with GKSS chemistry
  - Gas phase pollutants:  $\text{Hg}^0$ ,  $\text{HgO}$ ,  $\text{HgCl}_2$  and particulate Hg
  - 9 aqueous phase pollutants
  - Chemistry depending on  $\text{O}_3$ ,  $\text{SO}_2$ ,  $\text{Cl}^-$  and Soot
  - During the polar sunrise in the Arctic an additional fast oxidation rate of  $\text{Hg}^0$  to  $\text{HgO}$  is assumed
- Wet removal rates for all aqueous phase pollutants as for Sulphate
- Dry deposition velocity for  $\text{HgO}$  and  $\text{HgCl}_2$  as for  $\text{HNO}_3$  and for particulate Hg as for Sulphate

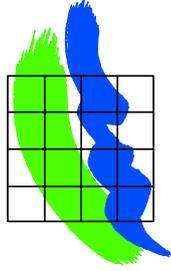


From Petersen et al. (1998)



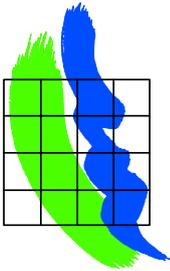
- GEIA Global Mercury Emissions Inventory for 1989  
80 % emitted as Hg<sup>0</sup> and 20 % as particulate Hg



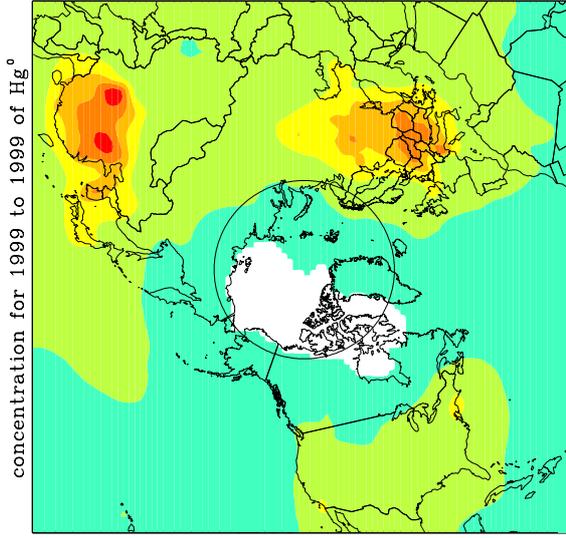
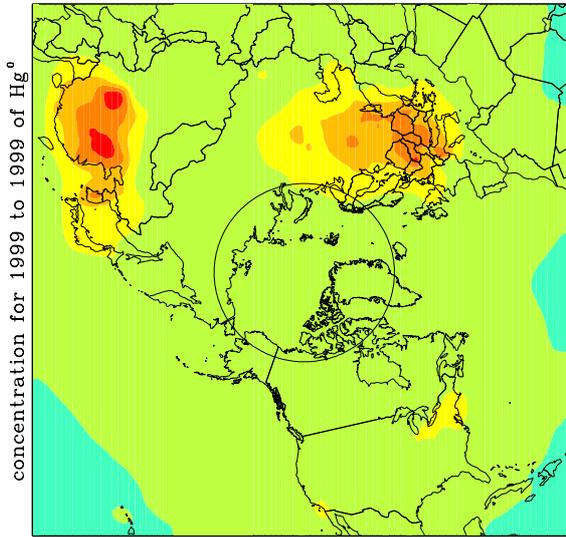


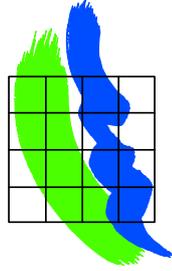
## Model simulations

- Model calculations for October 1998 to May 2000 with no nesting. All model runs with no re-emissions from land and oceans, instead a background concentrations on  $1.5 \text{ ng/m}^3$  of  $\text{Hg}^0$
- Basic Model with GKSS chemistry. During the polar sunrise in the Arctic an additional fast oxidation rate of  $\text{Hg}^0$  to  $\text{HgO}$  is assumed: Inside the boundary layer over sea ice during sunny conditions it is assumed that there is an additional oxidation rate of  $\frac{1}{4} \text{ hour}^{-1}$ . The fast oxidation stops, when surface temperature exceeds  $-4^\circ\text{C}$ .

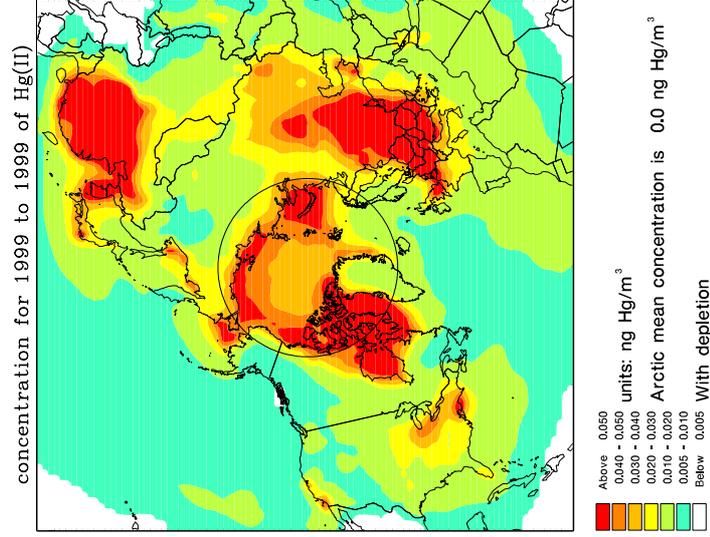
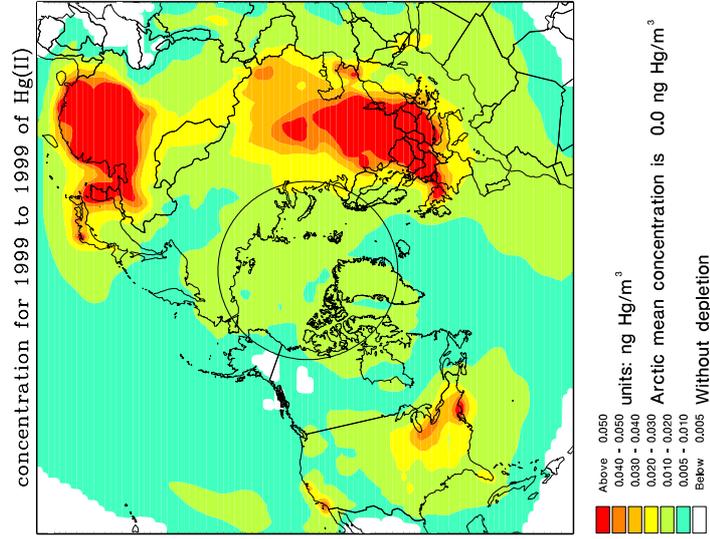


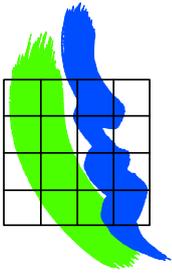
# Run without and with depletion



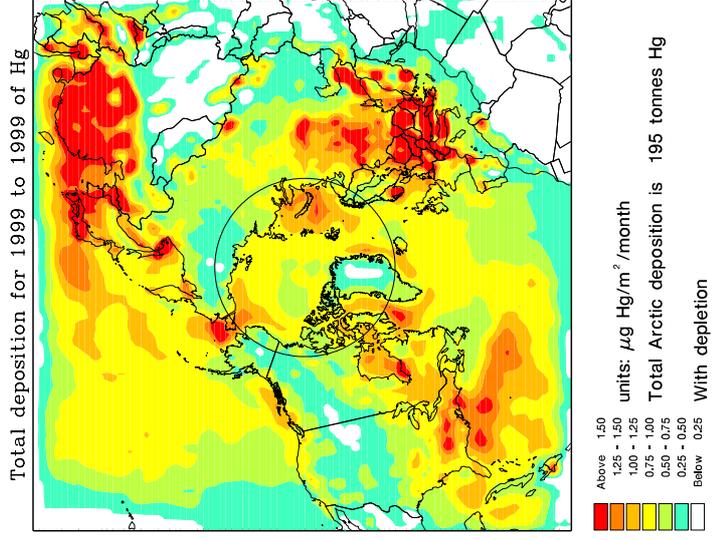
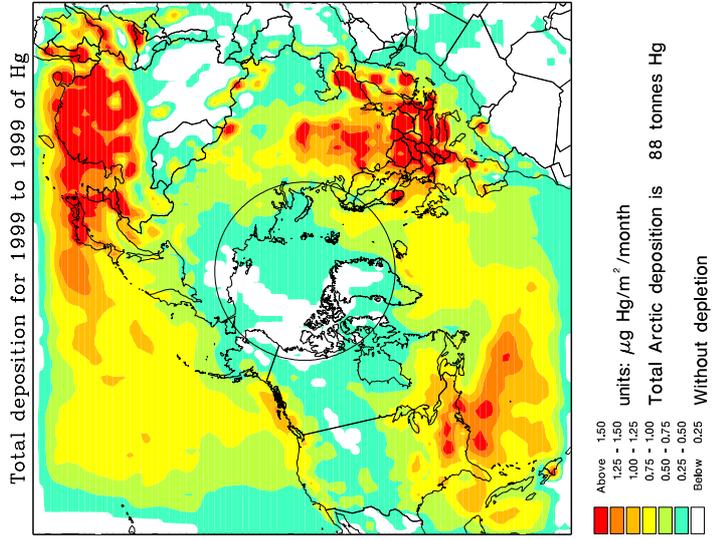


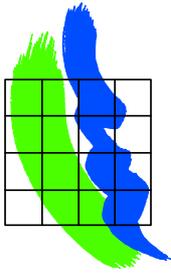
# Run without and with depletion



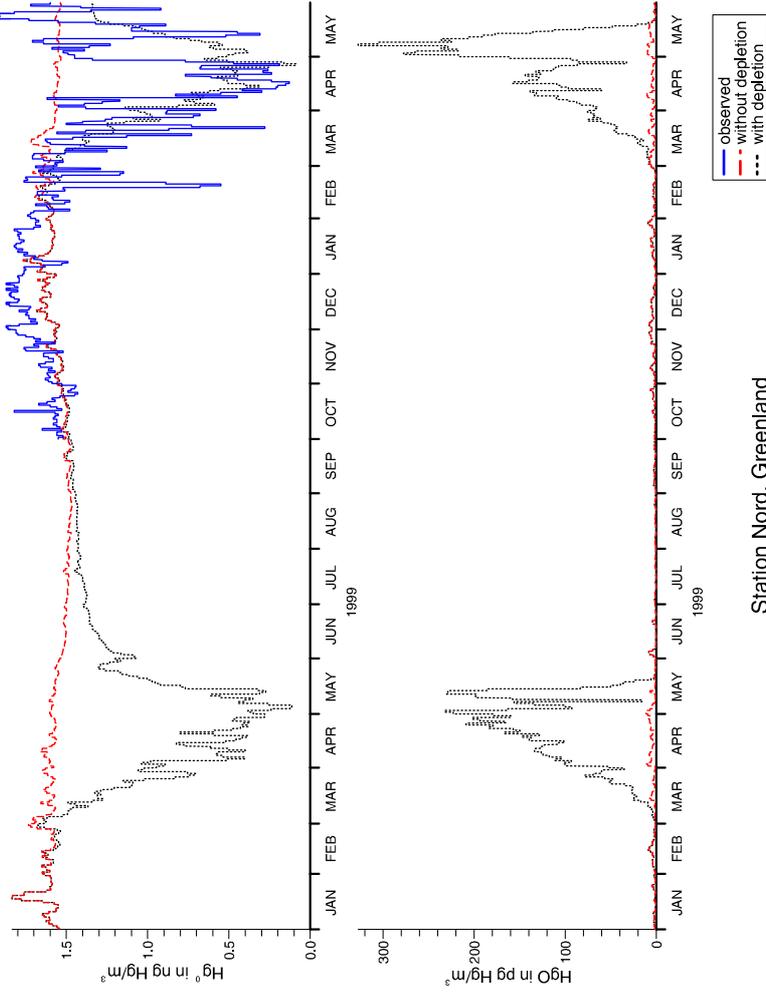


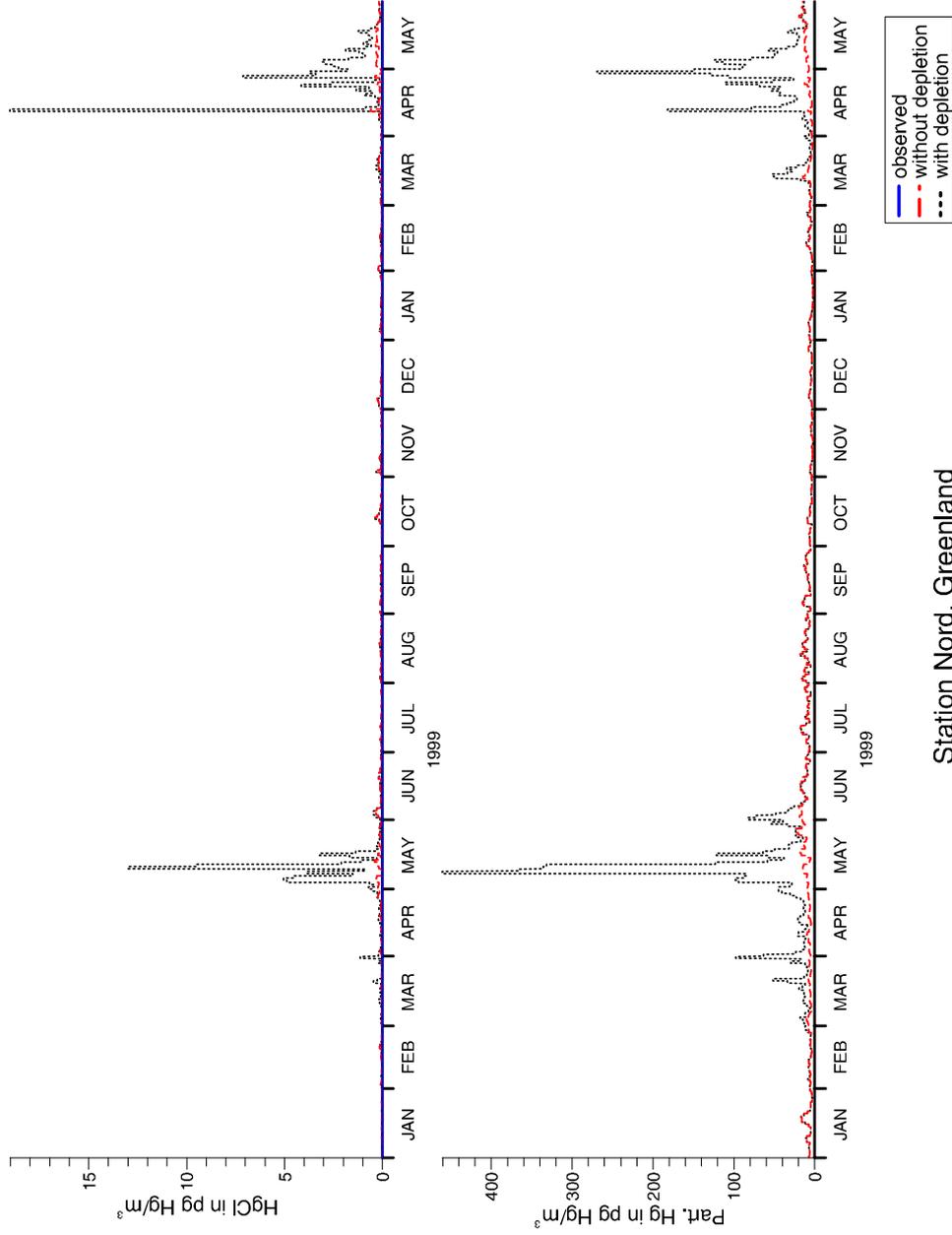
# Run without and with depletion



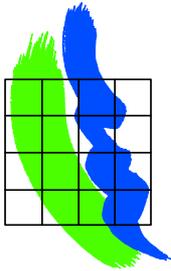


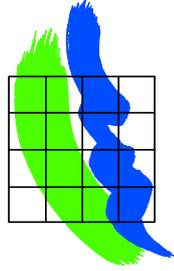
# Comparisons with measurements





Station Nord, Greenland

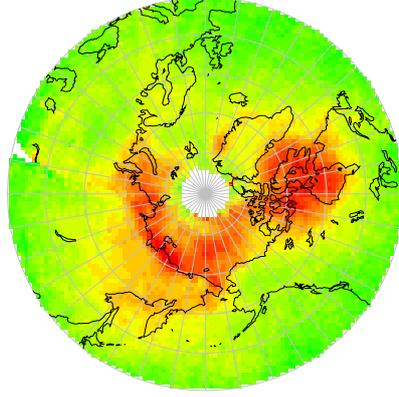




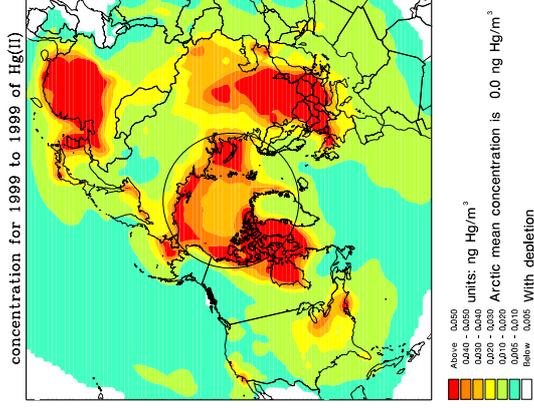
# Ongoing activities and future work

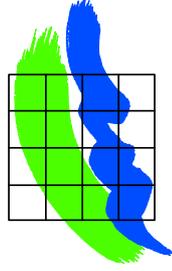
Improve parameterization of Arctic chemistry, coupling with GOME measurement of BrO, coupled to measurements of mercury in the Arctic in order to understand the spatial and temporal distribution of the depletion

March 1998



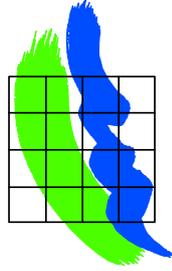
From Richter et al. (1997)





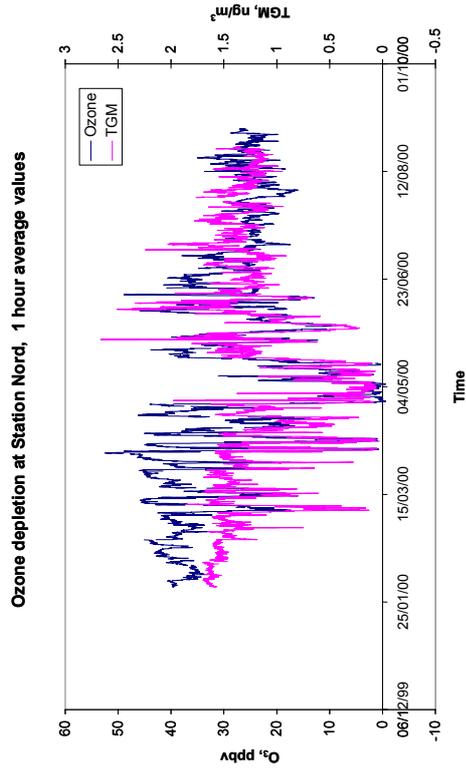
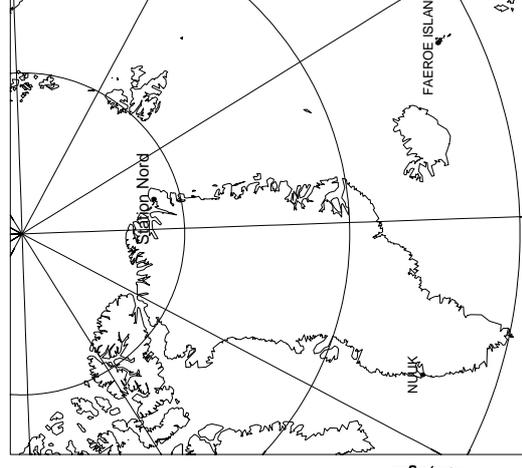
## Ongoing activities and future work

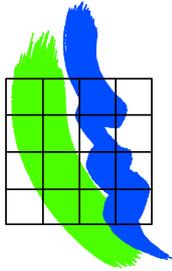
- Continuing the work with parameterization of background chemistry, coupling with aqueous chemistry and other oxidation pathways of  $\text{Hg}^0$
- Coupling of mercury chemistry with soot model
- Re-emissions from oceans, sea-ice and land



## Ongoing activities and future work

- Further validation of the model system
- RGM flux measurements with REA system





## ACKNOWLEDGEMENTS

The model work is financially supported by the Danish Environmental Protection Agency with means from the MIKA/DANCEA funds for Environmental Support to the Arctic Region



## 6.2.2 Alexey Gusev:



UN/ECE Convention on Long-Range  
Transboundary Air Pollution

**EMEP**

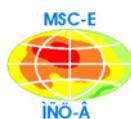
Co-operative programme for monitoring  
and evaluation of the long-range  
transmission of air pollutants in Europe

AMAP Workshop on Emissions, Sources and Discharges

### MODELLING OF HM AND POP TRANSPORT WITH AIR MASSES

Alexey Gusev, Oleg Travnikov

Meteorological Synthesizing Centre - East

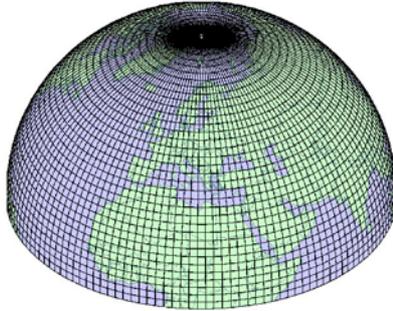


[www.msceast.org](http://www.msceast.org)



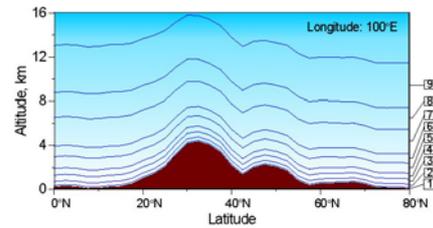
## Hemispheric model for Hg, PCBs, HCHs

Computation domain



Resolution:  $2.5^\circ \times 2.5^\circ$  (1 x 1)

Vertical grid structure



9 terrain-following layers



## Comparison with regional MSCE-HM model

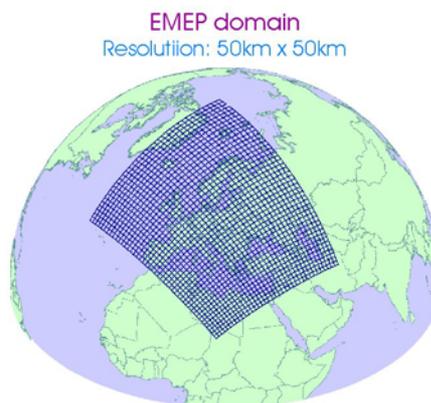
**Objective:** Lead airborne transport in 1990

### Regional model

**Domain:** EMEP domain  
**Resolution:** 50km x 50 km  
**Emission:** official emission data from the EMEP network for 1990

### Hemispheric model

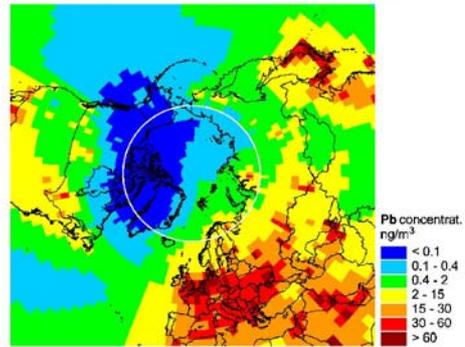
**Domain:** Northern Hemisphere  
**Resolution:** 2.5° x 2.5°  
**Emission:** reduced GEIA inventory for 1989



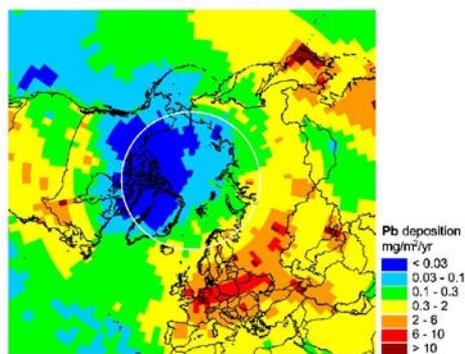


## Preliminary results for Pb in 1990

Mean annual Pb concentration in air



Total annual Pb deposition

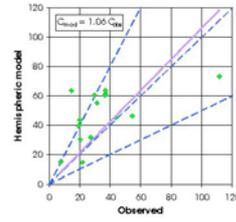
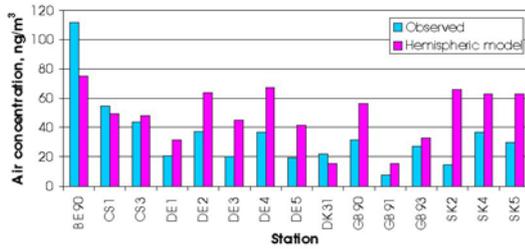




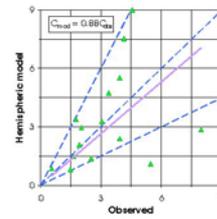
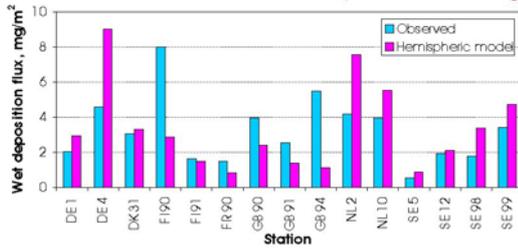
## Comparison with measurements

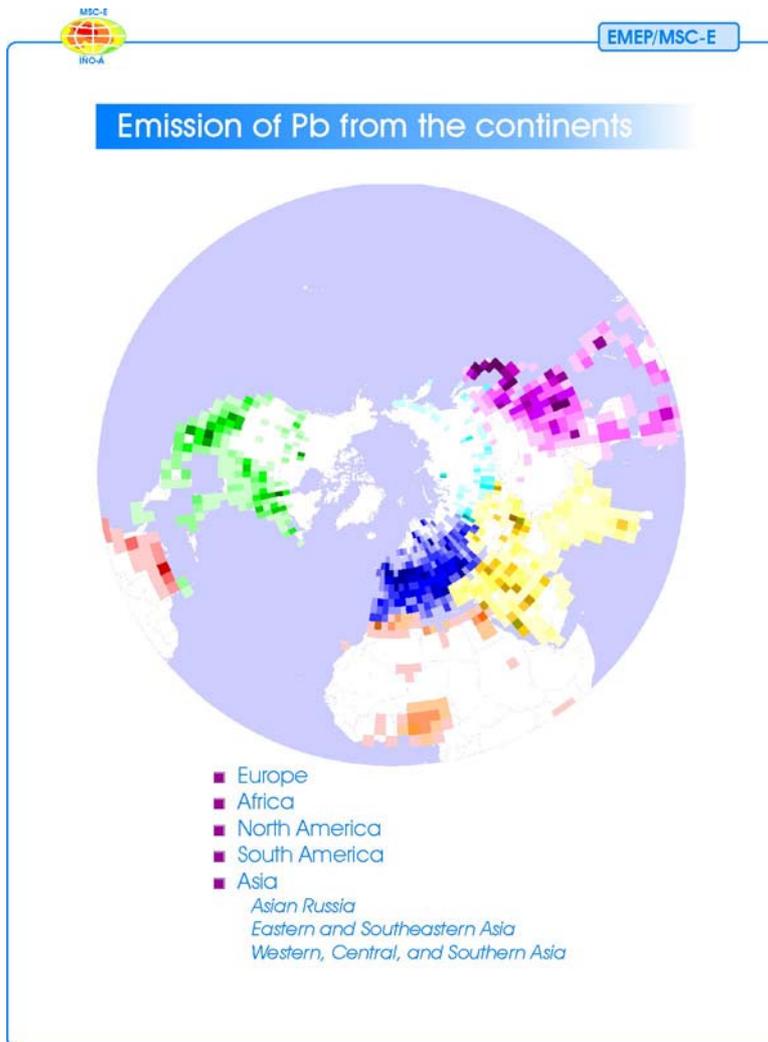
Emission: GEIA inventory of Pb in 1989  
 Measurements: EMEP monitoring network

Pb concentration in air,  $\text{ng/m}^3$



Wet deposition flux,  $\text{mg/m}^2/\text{yr}$



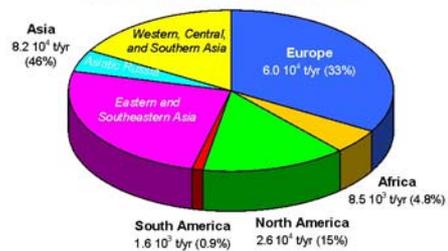




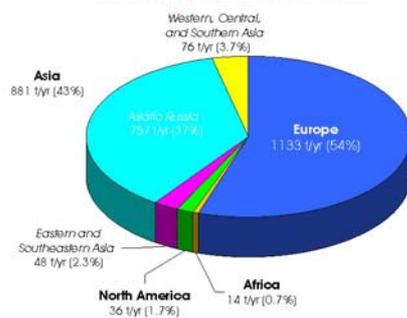
EMEP/MSC-E

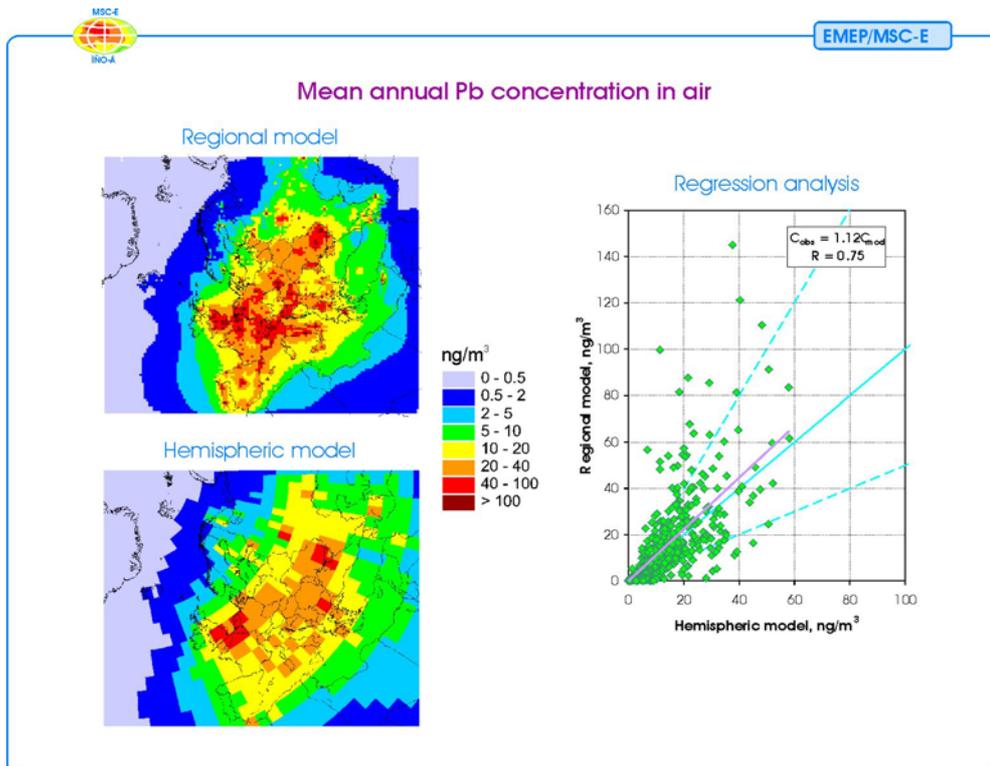
## Deposition to Arctic from continents

### Emission from continents



### Total deposition to Arctic

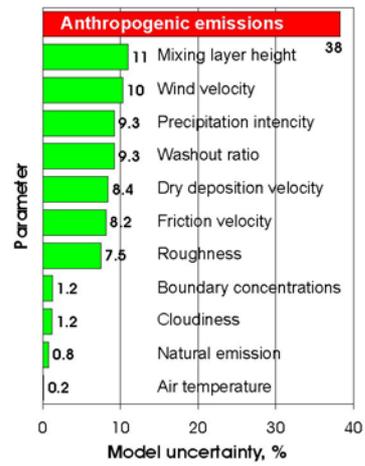




**Reliability of HM modeling results.  
Main activities of MSC-E:**

- Sensitivity study and uncertainty analysis
- Models intercomparison campaigns
- Comparison with measurements

**MSCE-HM model uncertainty due to individual parameters - air concentration of Pb**



Input uncertainty:  $\pm 40-50\%$



## Mercury intercomparison campaign

### Stages of mercury intercomparison

- Stage I** Comparison of chemical modules
- Stage II** Comparison with observations of short-term episodes
- Stage III** Comparison with monthly and annual mean measurements
- Stage IV** Comparison of transboundary transport results

### Participants

| Institution           | Model          |
|-----------------------|----------------|
| <b>EMEP/MSC-E</b>     | <b>MSCE-HM</b> |
| <b>GKSS, Germany</b>  | <b>TCM</b>     |
| <b>NIMH, Bulgaria</b> | <b>EMAP</b>    |
| <b>US EPA, USA</b>    | <b>CMAQ</b>    |
| <b>AER, USA</b>       | <b>AER</b>     |
| <b>IVL, Sweden</b>    | <b>IVL</b>     |
| <b>NERI, Denmark</b>  | <b>NERI</b>    |
| <b>EC/MSC, Canada</b> | <b>EC/MSC</b>  |

## EMEP/MSC-E

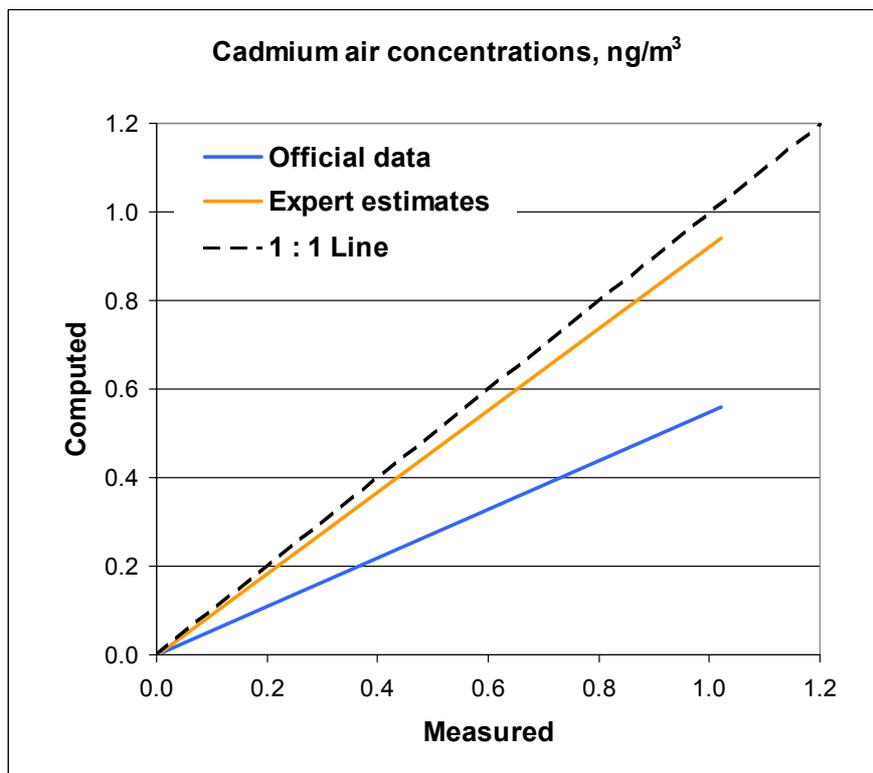
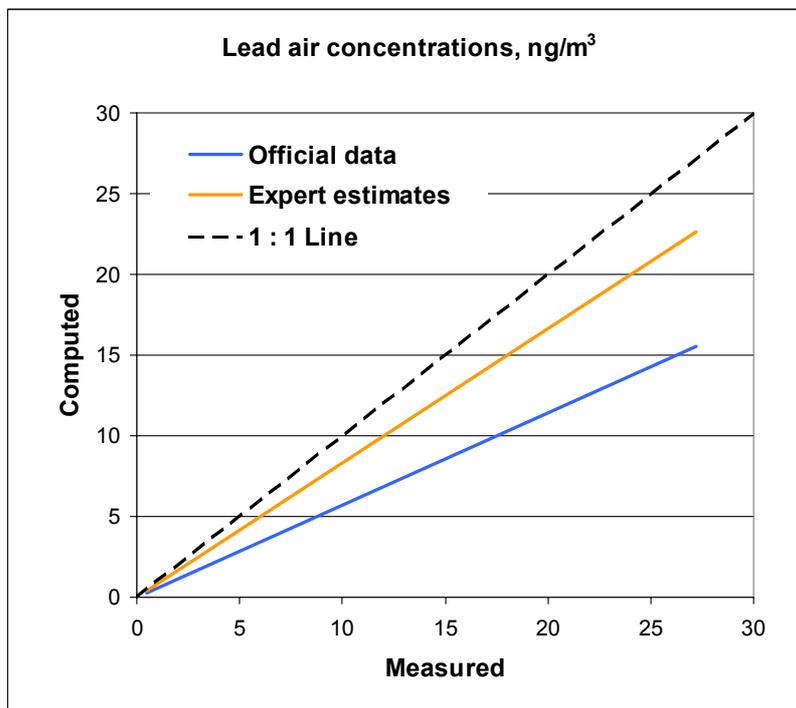
Emission data (t/y) for 1999 used in calculations  
(as in EMEP report 3/2001)

| Country                | Pb   | Cd   | Hg        |
|------------------------|------|------|-----------|
| Albania                | 24   | 0.6  | 0.46      |
| Armenia                | 60   | 1.5  | 0.00<br>1 |
| Austria                | 34   | 1.5  | 1.19      |
| Azerbaijan             | 144  | 3.1  | 0.61      |
| Belarus                | 38   | 1.4  | 0.38      |
| Belgium                | 212  | 3.3  | 3.02      |
| Bosnia and Herzegovina | 5    | 0.3  | 0.21      |
| Bulgaria               | 224  | 13.6 | 4.06      |
| Croatia                | 183  | 1.1  | 0.32      |
| Cyprus                 | 75   | 0.2  | 0.3       |
| Czech Republic         | 157  | 2.7  | 3.66      |
| Denmark                | 7    | 0.7  | 1.98      |
| Estonia                | 45   | 0.8  | 0.61      |
| Finland                | 14   | 0.6  | 0.4       |
| France                 | 868  | 12.1 | 16.6<br>3 |
| Georgia                | 112  | 2.4  | 0.47      |
| Germany                | 542  | 11   | 31        |
| Greece                 | 470  | 3    | 13        |
| Hungary                | 39   | 2.9  | 4.26      |
| Iceland                | 0    | 0.2  | 0.05      |
| Ireland                | 88   | 1.6  | 1.92      |
| Italy                  | 2174 | 29.9 | 13.2<br>3 |

| Country     | Pb   | Cd   | Hg    |
|-------------|------|------|-------|
| Kazakhstan  | 20   | 0.4  | 0.09  |
| Latvia      | 12   | 0.3  | 0.18  |
| Lithuania   | 19   | 2    | 0.25  |
| Luxembourg  | 2    | 0.05 | 0.29  |
| Netherlands | 35   | 1    | 0.53  |
| Norway      | 6    | 1.1  | 1.2   |
| Poland      | 694  | 55.1 | 25.1  |
| Portugal    | 392  | 3.4  | 4.92  |
| Moldova     | 11   | 0.1  | 0.18  |
| Romania     | 517  | 20.6 | 6.65  |
| Russia      | 2339 | 50.9 | 9.9   |
| Slovakia    | 53   | 7.5  | 1.66  |
| Slovenia    | 50   | 1.6  | 0.6   |
| Spain       | 944  | 13.5 | 18.04 |
| Sweden      | 38   | 0.8  | 0.9   |
| Switzerland | 131  | 2.2  | 2.63  |
| Macedonia   | 119  | 5.2  | 1.06  |
| Turkey      | 774  | 14   | 4.3   |
| Ukraine     | 2564 | 43.2 | 30.6  |
| U. K.       | 534  | 7    | 8.7   |
| Yugoslavia  | 358  | 6.3  | 3.36  |
|             |      |      |       |

Grey boxes indicate officially submitted data by EMEP countries,  
other boxes - available expert estimates

**Official emission data and expert estimates of Pb and Cd emissions within the EMEP region for 1996**



**Comparison of measured and computed air concentrations of Pb and Cd**

*EMEP/MSC-E*

Expert estimates of POP emission for 1970-1995 prepared within the project POPCYCLING-Baltic cover **PCBs, B[a]P, HCHs, PCDD/Fs, DDT, HCB** with resolution 50x50 km<sup>2</sup> for the EMEP grid

Officially submitted data on emissions of PCDD/Fs (g I-TEQ/y), PCBs (kg/y), HCB (kg/y) in European countries for 1998

| Country                | PCDD/Fs | PCBs | HCB       | Country            | PCDD/Fs | PCBs | HCB  |
|------------------------|---------|------|-----------|--------------------|---------|------|------|
| Albania                |         |      |           | Kazakhstan         |         |      |      |
| Armenia                |         |      | 0         | Latvia             |         |      |      |
| Austria                | 53.8    |      | 0         | Lithuania          | 5.97    | 14   |      |
| Azerbaijan             |         |      |           | Luxembourg         | 8       |      |      |
| Belarus                | 15.7    |      | 0         | Netherlands        | 43.8    |      | 0    |
| Belgium                | 116     |      | 25        | Norway             |         |      | 50   |
| Bosnia and Herzegovina |         |      |           | Poland             | 290     | 2312 | 0    |
| Bulgaria               | 288     | 253  |           | Portugal           |         |      |      |
| Croatia                | 111     |      |           | Moldova            |         |      | 0    |
| Cyprus                 |         |      |           | Romania            |         |      |      |
| Czech Republic         | 767     | 458  |           | Russian Federation | 606     |      | 0.95 |
| Denmark                |         |      |           | Slovakia           | 188     | 136  |      |
| Estonia                |         |      |           | Slovenia           | 3.53    | 180  |      |
| Finland                | 39.5    |      | 0         | Spain              |         |      |      |
| France                 | 1391    | 50   |           | Sweden             |         |      | 0    |
| Georgia                |         |      |           | Switzerland        |         |      | 0    |
| Germany                |         |      |           | Macedonia          |         |      |      |
| Greece                 |         |      |           | Turkey             |         |      |      |
| Hungary                | 93.6    | 92   | 0.71<br>2 | Ukraine            |         |      | 0    |
| Iceland                |         |      |           | United Kingdom     | 277     | 2747 | 893  |
| Ireland                |         |      |           | Yugoslavia         |         |      |      |
| Italy                  |         |      |           |                    |         |      |      |

# **Workshop on Emissions and Emission Factor Estimates**

**jointly organized  
by EMEP/MSC-E and UN/ECE Task Force on  
Emission Inventories and Projections in Moscow  
from 21 to 23 November 2001.**

## **Objective:**

- To assist countries to report emission data for HMs and POPs.
- To improve HM and POP emission estimates.



EMEP/MSC-E

## Organizations dealing with HMs and POPs

**AMAP**

**EEA**

**EMEP**

**EUROTRAC**

**HELCOM**

**OSPAR**

**OECD**

**UNEP**

**WMO**

6.2.3 *Mark Cohen*

**Source-Receptor Modeling in North America:  
Lessons Learned from Attempts to  
Model the Transport and Deposition  
of Atmospheric Toxics to the Great Lakes**

**Dr. Mark Cohen**  
NOAA Air Resources Laboratory  
SSMC3, R/ARL, Room 3316  
1315 East West Highway, Silver Spring MD, 20910  
ph: 301-713-0295 x122; fax: 301-713-0119  
*mark.cohen@noaa.gov*



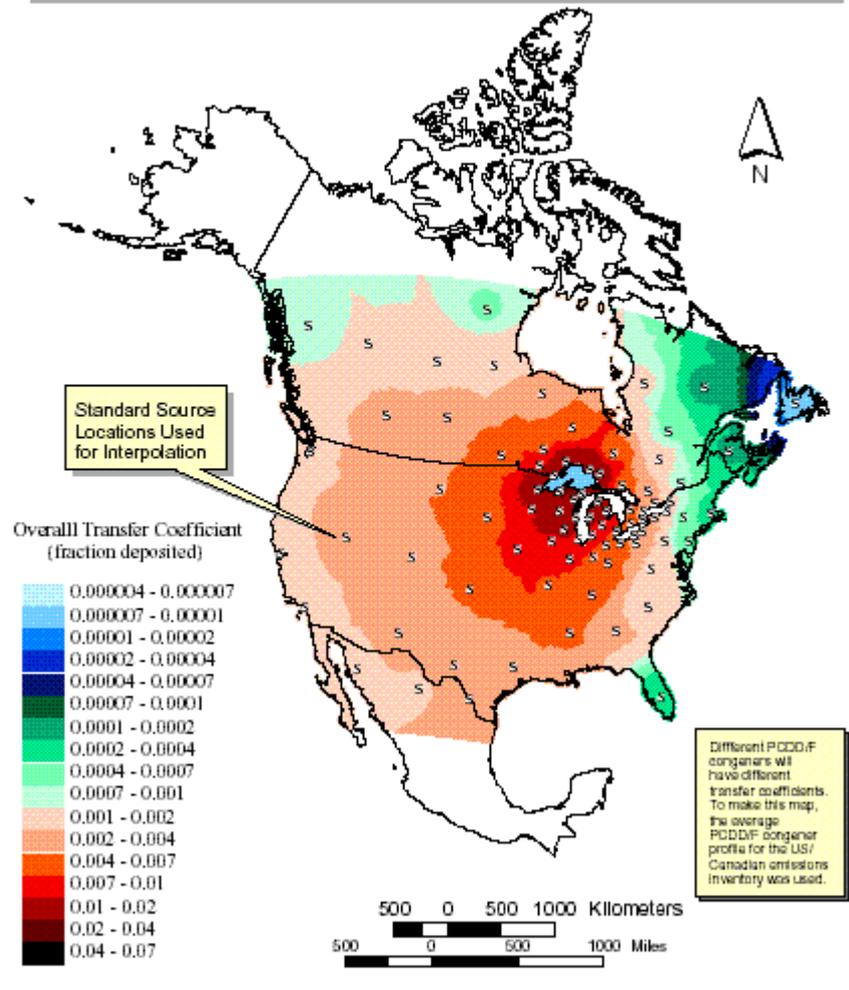
**Presentation at:**

**AMAP Workshop on Sources, Emissions and Discharges  
Norwegian Institute for Air Research (NILU)  
Kjeller, Norway, August 23-24, 2001**

## Comprehensive Modeling Can Be Used to Estimate the Impact of Emissions Sources on Receptors

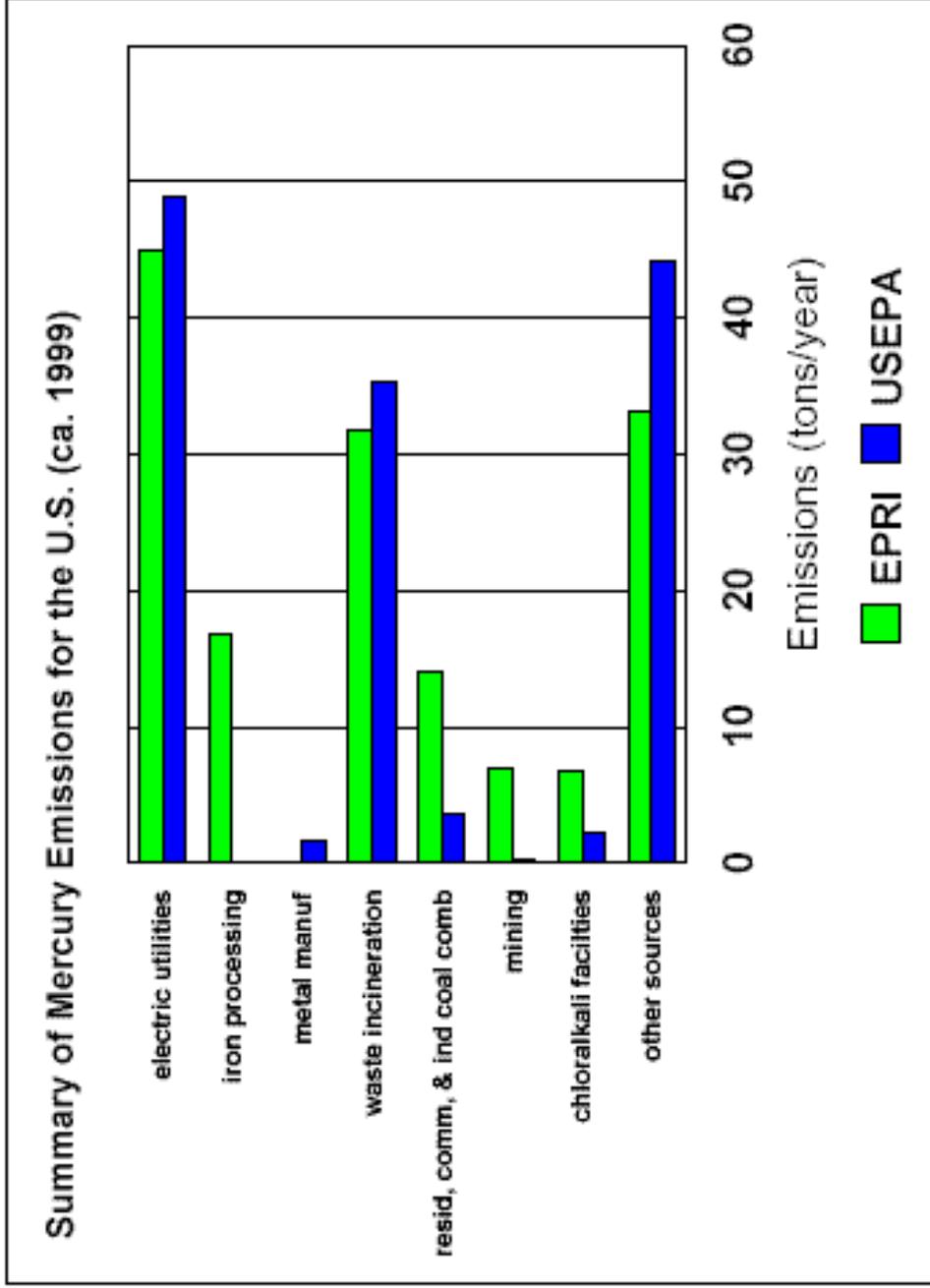
- regional and long-range transport can be very important
- ideally, one wants to get source-receptor information, but it is rarely attempted due to technical and political barriers
- *U.S. appears to be much more concerned with what others might be doing to it than with what it is doing to itself and others...*
- an interpolation-based approach has been developed for trace toxic species that allows source-receptor information to be obtained with some degree of numerical efficiency
- a comprehensive modeling analysis has been performed for the Great Lakes for dioxin and atrazine [current work: mercury]
- uncertainties in the modeling are large, but the largest uncertainty appears to be the emissions inventory
- monitoring and modeling are best together; alone they are not nearly as useful

**Fig. 3. Dioxin Transfer Coefficients: Theoretical Fraction of 1996 Dioxin Emissions Deposited in Lake Superior (grams TEQ deposited per year/ grams TEQ emitted per year) from Various Regions in the U.S. and Canada**



## North American Emissions Inventories For Many Pollutants Are Generally of Rather Poor Quality

- other than the criteria pollutants (e.g., sulfur, nitrogen, etc.), for most (if not all) toxic pollutants, there is no systematic approach to developing a comprehensive inventory...
- often, large source categories are omitted, major uncertainties remain unaddressed for long periods of time (or are never addressed)...  
need *omissions* inventory in addition to emissions inventory
- the emissions inventory development process is highly politicized
- confidentiality, secrecy, non-transparency, and non-cooperativeness remain a problem for government and industrial/commercial emissions sources
  - *activity factors & emissions factors are rarely documented or even provided*
  - *emissions records are of uncertain quality: they may be ok or they may be terrible; with most inventories, there is no way to really know the difference unless you start from the very beginning and recreate the inventory yourself!*
  - *example of current U.S. mercury inventory: latest information is being held secret while regulatory decisions being made*
- tyranny of "old" inventories -- all that is available are very old inventories; then, when one uses them in an analysis, it is said that the results are obsolete, and that the current situation is much better... need more up-to-date information so that analyses can be policy-relevant.
- even if you are not doing modeling, you still need an accurate emissions inventory to do almost anything about a pollutant...



## WHAT DO YOU WANT TO KNOW?

1. Is a given pollutant causing a *problem*?
2. If so, is the atmospheric pathway *significant* relative to other loading pathways, and what is the *contribution* from the atmospheric pathway?
3. **SOURCE RECEPTOR RELATIONSHIPS** -- If the atmospheric pathway is significant, what is the *geographic scale* of the problem -- what fraction of the loading is due to local contributions? what fraction due to regional contributions? what fraction due to more distant sources? What are the most significant *source categories* contributing to the atmospheric loading?

Obviously, the reason why you want to know about source-receptor relationships is that you want to eventually be able to do something about the problem.

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**The answers to the above questions will obviously depend on**

- what pollutant you are considering,
- where you are considering it, and
- when you are considering it...

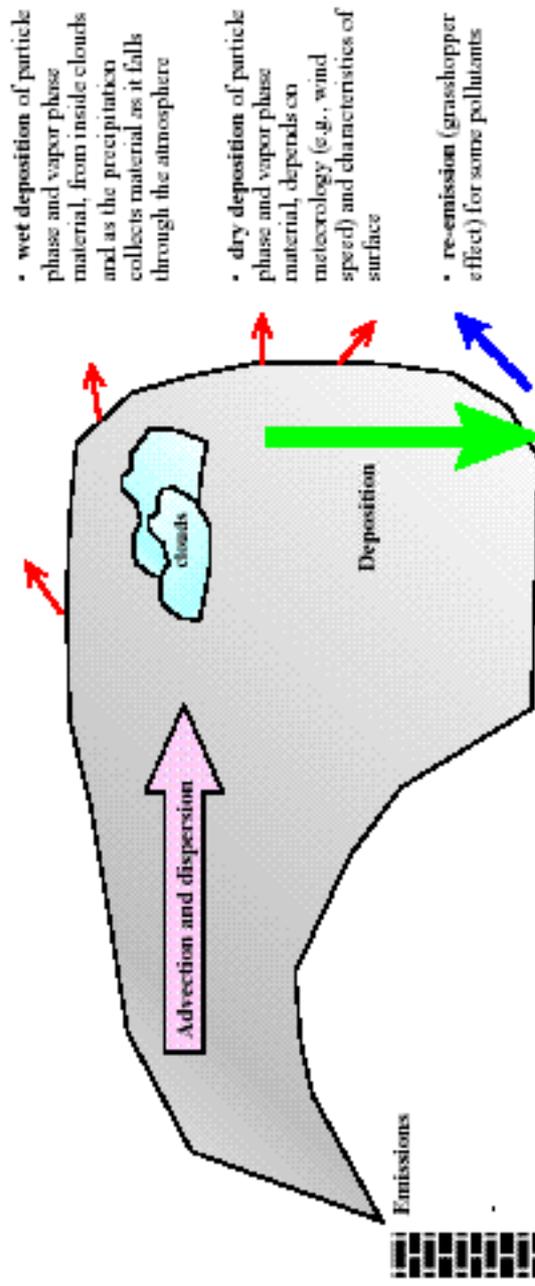
**And, you might be interested in trends in the above information –**

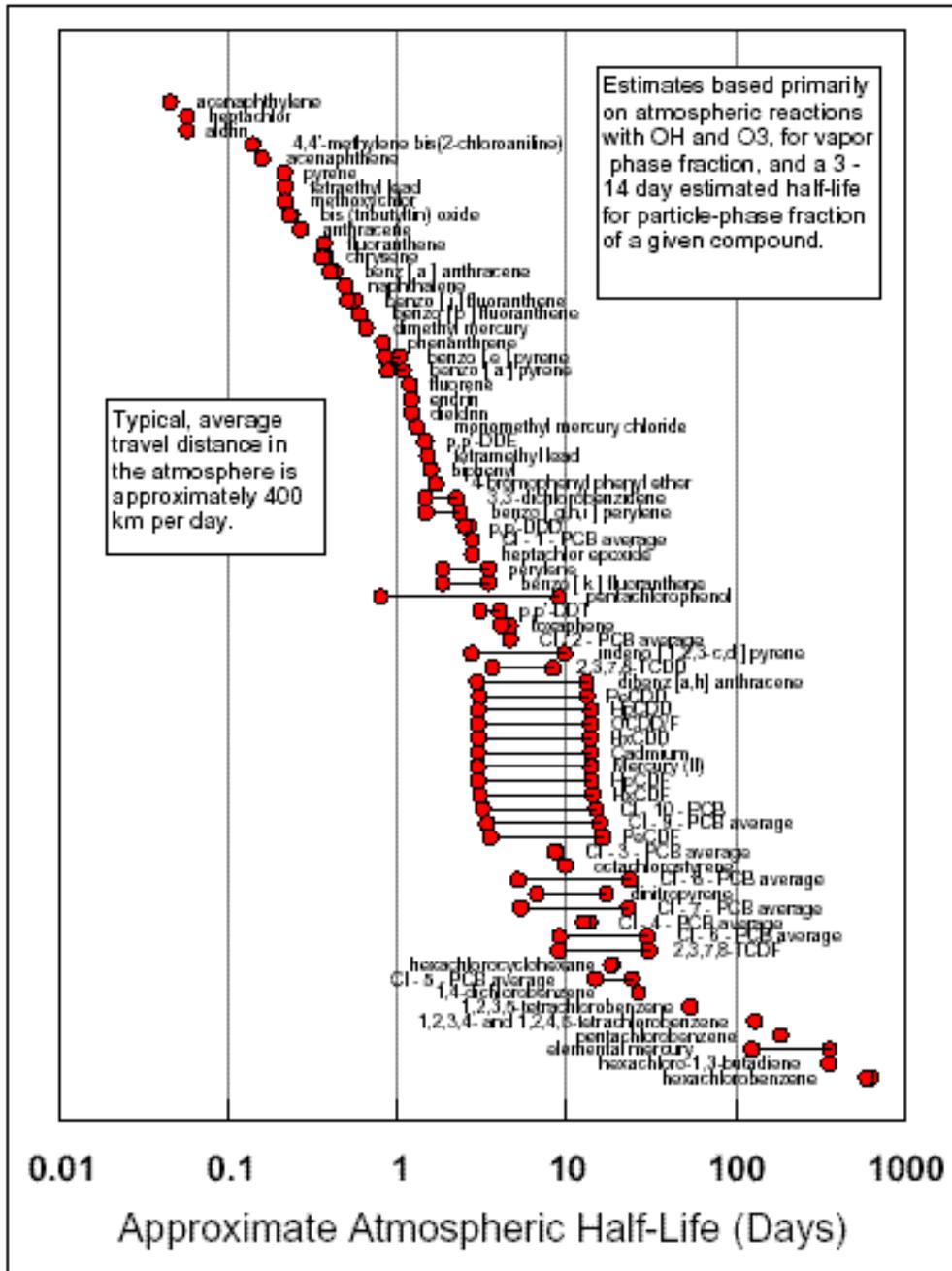
- has deposition been increasing or decreasing?
- what might the impact be of a change in the emissions from one or more sources in the future?

| Estimates of the Percent of Great Lakes Loadings<br>Attributable to the Atmospheric Deposition Pathway  |   |  |                  |                     |   |
|---|---|--|------------------|---------------------|---|
| Pollutant   | Lake Superior   | Lake Michigan  | Lake Huron       | Lake Erie           | Lake Ontario  |
| DDE   | 97 <sup>a</sup>   | 98 <sup>a</sup>  | 97 <sup>a</sup>  | 22 <sup>a</sup>     | 31 <sup>a</sup>                                     |
| Lead  | 97 <sup>a</sup> ; 64 <sup>b</sup> ; 69 <sup>c</sup>     | 99 <sup>a</sup>  | 98 <sup>a</sup>  | 48 <sup>a</sup>     | 73 <sup>a</sup>                                     |
| Mercury   | 73 <sup>d</sup>   | > 80 <sup>e</sup>  | <sup>a</sup>     | <sup>a</sup>        | <sup>a</sup>  |
| PCB's   | 90 <sup>a</sup> ; ~ 95 <sup>b,c</sup> ; 82 <sup>d</sup> | 58 <sup>a</sup>  | 78 <sup>a</sup>  | 13 <sup>a</sup>     | 7 <sup>a</sup>                                      |
| PCDD/F  | -100 <sup>f</sup><br>-80 <sup>f</sup>                   | 50-100 <sup>g</sup><br>(PCDD)<br>5-35 <sup>g</sup> (PCDF)<br>88 <sup>f</sup> | 88 <sup>f</sup>  | -40 <sup>f</sup>    | 5-35 (PCDD) <sup>g</sup><br>< 5 (PCDF) <sup>g</sup> |
| Benzo(a)pyrene  | 96 <sup>a</sup>   | 80 <sup>a</sup>  | 80 <sup>a</sup>  | 79 <sup>a</sup>     | 72 <sup>a</sup>                                     |
| Hexachloro-<br>benzene  | 99 <sup>f</sup>   | 95 <sup>f</sup>  | 96 <sup>f</sup>  | > 17 <sup>f</sup>   | 40 <sup>f</sup>                                     |
| Atrazine  | 97 <sup>h</sup>   | -30 <sup>h</sup> ; 23 <sup>h</sup>   | -20 <sup>h</sup> | -10-20 <sup>h</sup> | -5 <sup>h</sup>                                     |
| Mirex   | <sup>a</sup>  | <sup>a</sup>   | <sup>a</sup>     | <sup>a</sup>        | -5 <sup>h</sup>                                     |
| <b>References and Notes</b><br>(a) Strachan and Eisenreich (1988), percentages of total inputs; (b) Hoff <i>et al.</i> (1996); (c) Net loss of PCB's to the atmosphere of 1600 kg/year; total non-atmospheric inputs of approximately 70 kg/year; (d) Dokan <i>et al.</i> (1993); (e) Pearson <i>et al.</i> (1998); (f) Cohen <i>et al.</i> (1995); (g) Rygwinski <i>et al.</i> (1999); (h) Schottler and Eisenreich (1997); (i) Mason and Sullivan (1997); (k) no estimates could be found |   |  |                  |                     |   |

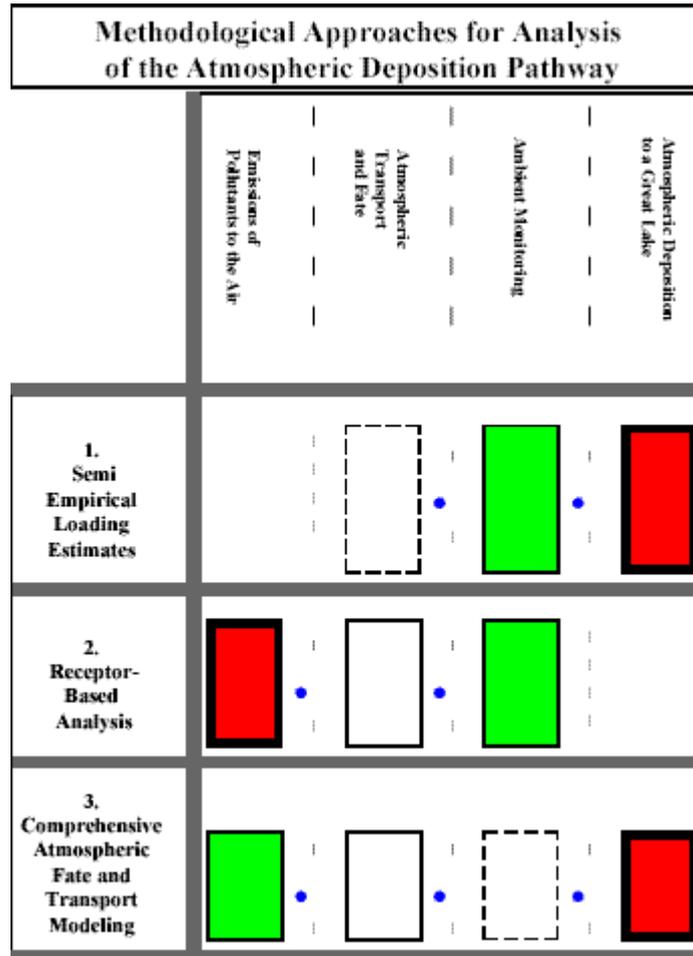
## TRANSPORT and FATE of ATMOSPHERIC POLLUTANTS

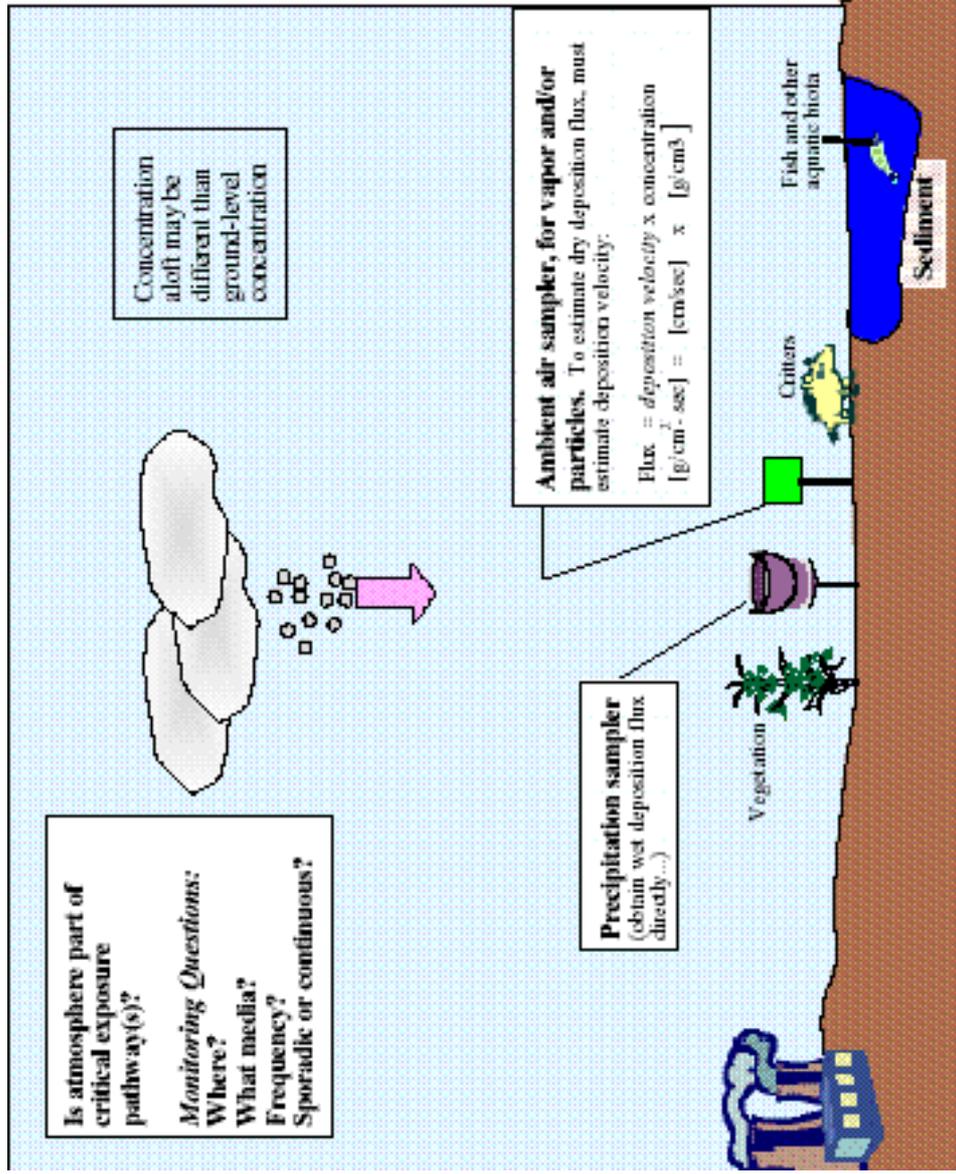
- vapor-particle partitioning at low relative humidity
- vapor-droplet partitioning in clouds (and/or at high relative humidity)
- chemical transformations producing new compounds (may be more or less toxic) due to reactions and photolysis in the *vapor phase*, *on the surface of particles*, and/or *within droplets* (note: this may involve interaction with compounds from other emissions sources)





| Illustrative Examples of Certain Aspects of Atmospheric Behavior of Pollutants  |                             |              |          |                           |              |      |                            |   |
|---|-----------------------------|--------------|----------|---------------------------|--------------|------|----------------------------|---|
| Pollutants  | Vapor-Particle Partitioning |              |          | Relative water solubility |              |      | Important Deposition Modes |   |
|   | vapor                       | intermediate | particle | low                       | intermediate | high | wet                        | dry   |
| <ul style="list-style-type: none"> <li>• hexachlorobenzene</li> <li>• other chlorobenzenes</li> <li>• light PAH's (e.g., anthracene)</li> <li>• elemental mercury</li> <li>• aldrin/dieldrin</li> <li>• mirex</li> <li>• endrin</li> <li>• heptachlor</li> <li>• hexachloro-1,3-butadiene</li> <li>• octachlorostyrene</li> </ul> | •                           |              |          | •                         |              |      |                            | •<br><br><i>NOTE:<br/>Two-way gas<br/>exchange at<br/>terrestrial<br/>and water<br/>surfaces can<br/>be important</i> |
| <ul style="list-style-type: none"> <li>• hexachlorocyclohexanes (e.g., lindane)</li> <li>• pentachlorophenol</li> </ul>   |                             |              |          |                           | •            |      | •                          | •   |
| <ul style="list-style-type: none"> <li>• reactive gaseous mercury (e.g., HgCl<sub>2</sub>)</li> <li>• 4,4'-methylene bis(2-chloroaniline)</li> </ul>  | •                           |              |          |                           |              | •    | •                          | •   |
| <ul style="list-style-type: none"> <li>• atrazine</li> </ul>  | •<br>?                      | •<br>?       | •<br>?   |                           |              | •    | •                          | •<br>?  |
| <ul style="list-style-type: none"> <li>• TCDD/F and PeCDD/F</li> <li>• DDT / DDD / DDE</li> <li>• toxaphene</li> <li>• some PAH's (e.g., fluorethene, chrysene)</li> <li>• methoxychlor</li> <li>• PCB's</li> </ul>   |                             | •            |          | •                         |              |      | •                          | •   |
| <ul style="list-style-type: none"> <li>• cadmium</li> <li>• particulate mercury</li> <li>• HxCDD/F, HpCDD/F, OCDD/F</li> <li>• heavy PAH's (e.g., benzo-a-pyrene)</li> </ul>  |                             |              | •        | •                         |              |      | •                          | •<br>?<br><i>(?d for small<br/>particles?)</i>  |





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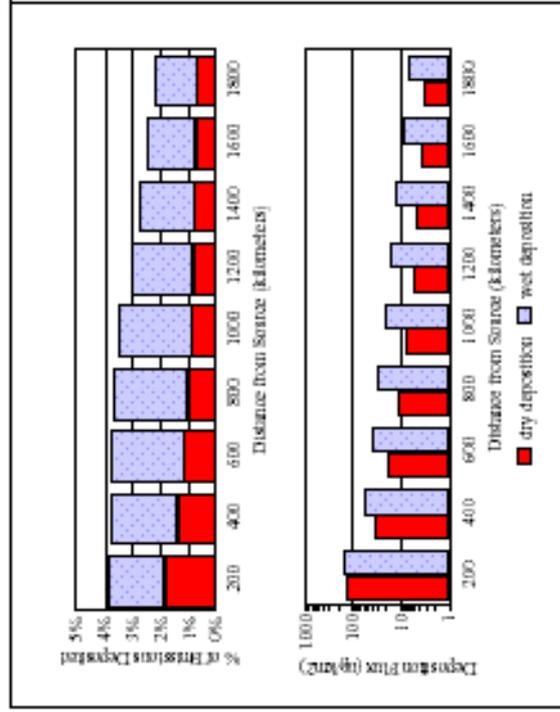
## SUMMARY

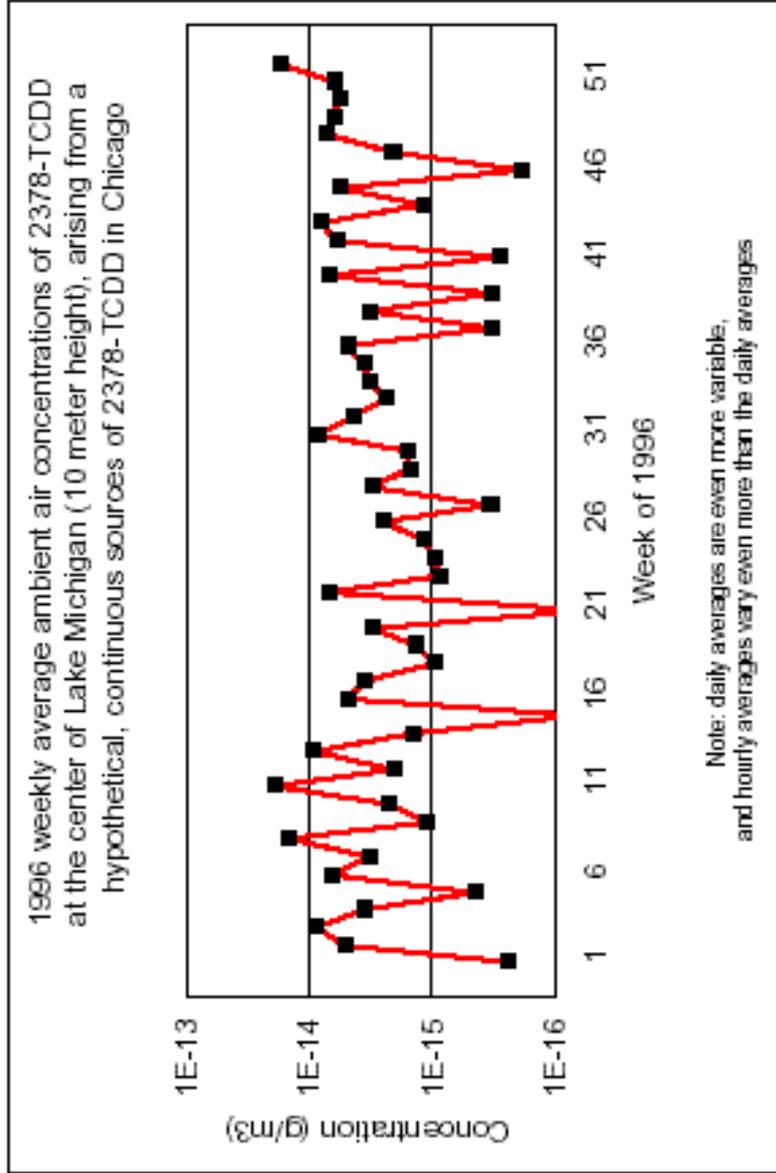
### Short Range • Long Range Transport

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- **From the Perspective of the Source** (e.g., a given incinerator)
    - dispersion, transformation, and deposition generally serve to reduce impacts as distance from the source increases:  
*but, for some pollutants, re-emission can lengthen the effective transport distance and even cause an eventual buildup at distances far from the source*
    - in general, the biggest impacts are nearby:  
*but, this represents only a small fraction of the emissions*
    - each pollutant behaves differently:  
*wide range of estimated lifetimes/transport distances/re-emissions rates*
    - we have some idea of the fate and transport of pollutants:  
*but, there are a lot of uncertainties*
- 
- **From the Perspective of the Receptor** (e.g., the Great Lakes)
    - the contributions from individual sources will tend to be very *episodic*
    - on a source by source basis, all things being equal, nearby sources will have a greater impact than far-away sources:  
*but the number of sources can increase as you go away from the receptor; so there is a competition – less impact per source, but more sources; [worldwide sources may be significant for some pollutants (e.g., HCB)]*
    - geographical distribution (distance/orientation) is important:  
*but for many pollutants of concern in the Great Lakes, our characterization of sources remains relatively uncertain...*
    - Dioxin examples for the Great Lakes  
*for Lake Michigan, about 40% comes from within 100 km of the lake, but more than half comes from much further away (100-1500 km). For the other Great Lakes, even less local impact.*

Figure 11. Deposition amount and flux of 2,3,7,8-TCDD in successive, concentric, annular 200-km-radius-increment regions away from a continuous 1996 year-long source at the center of the modeling domain. The deposition amount has been divided by the total amount emitted in the simulation to give the fraction of the emissions deposited in any given concentric region. The deposition flux for each region has been normalized to correspond to an emissions rate of 1 gram/year.





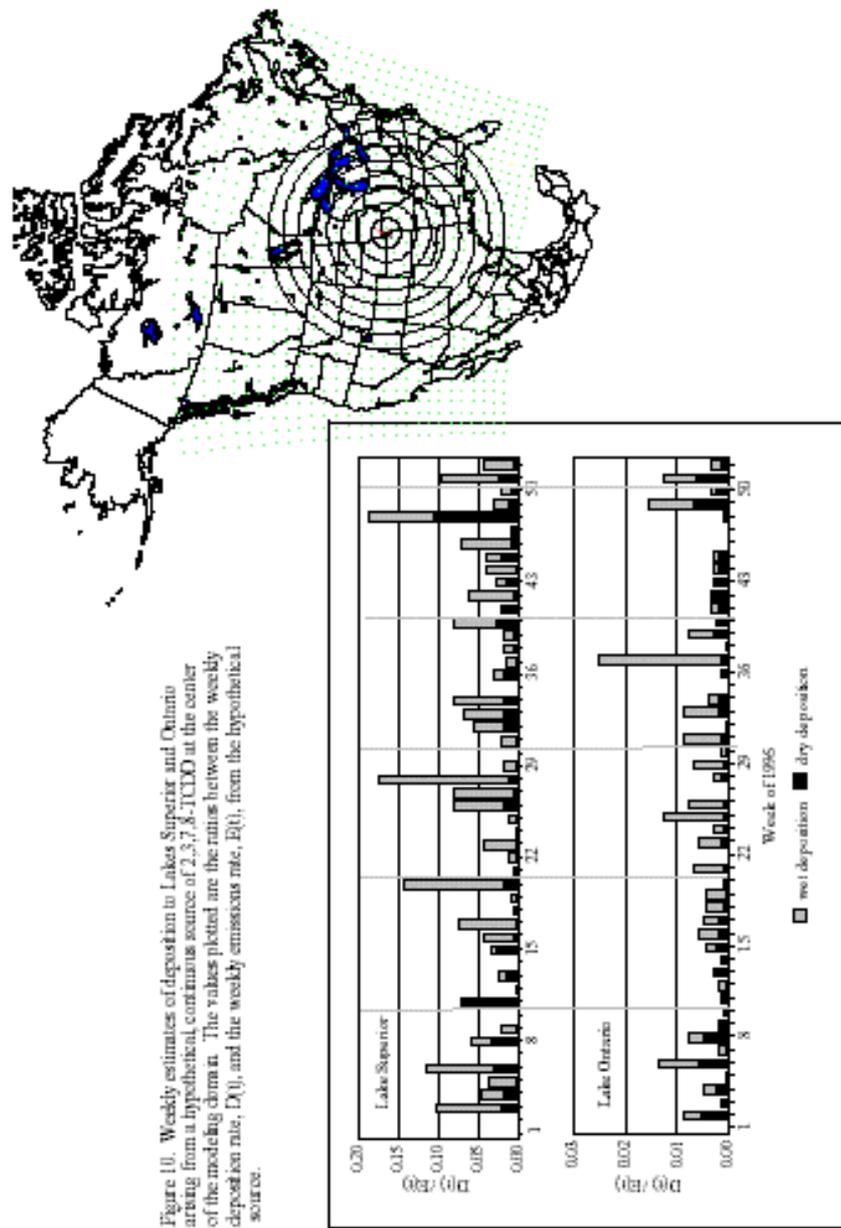


Figure 10. Weekly estimates of deposition to Lakes Superior and Ontario arising from a hypothetical, continuous source of 2.3, 7.8, 1000 at the center of the modeling domain. The values plotted are the ratios between the weekly deposition rate,  $D(t)$ , and the weekly emission rate,  $E(t)$ , from the hypothetical sources.

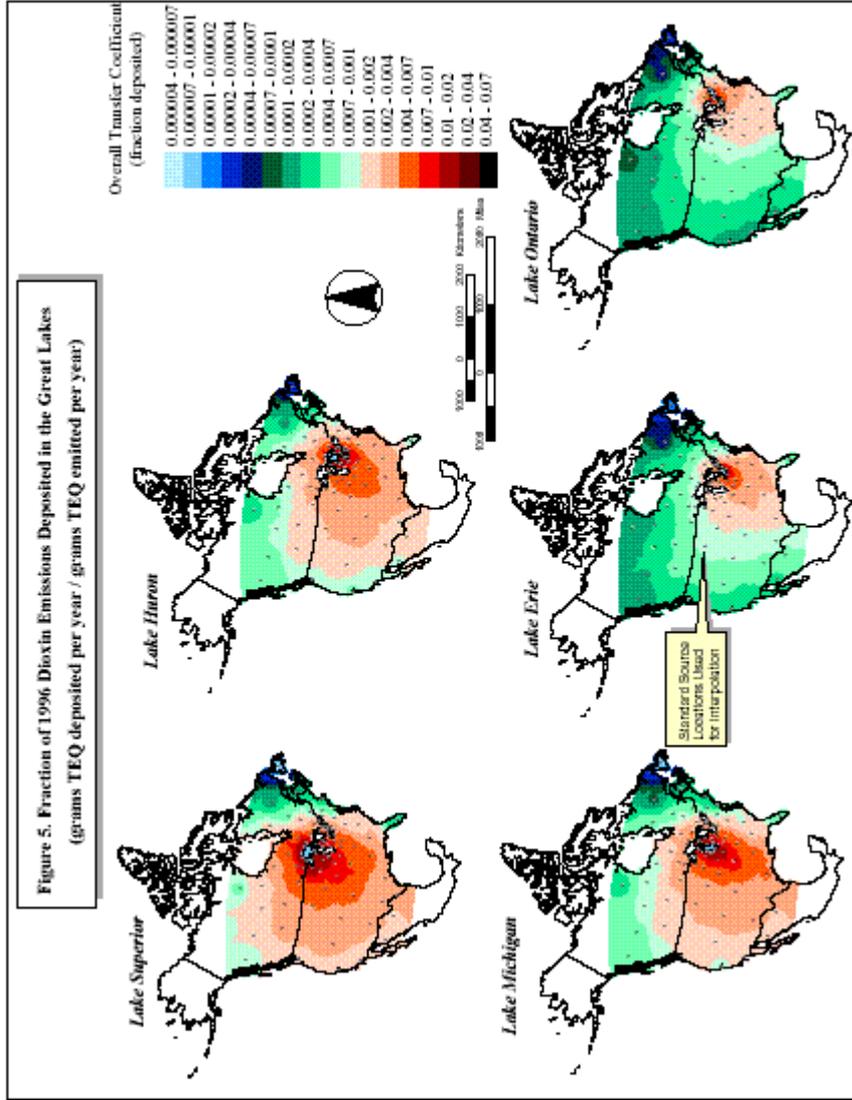
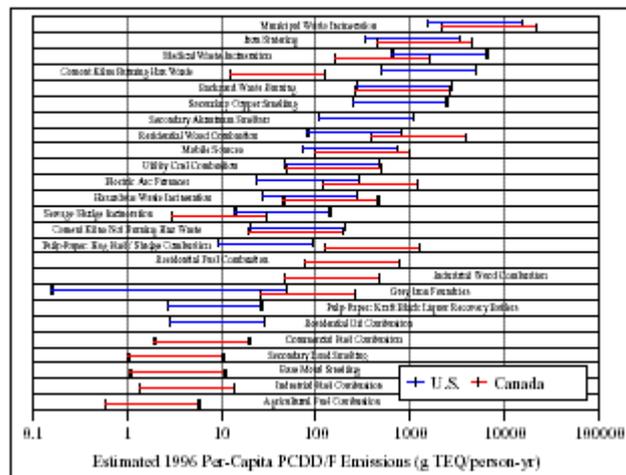
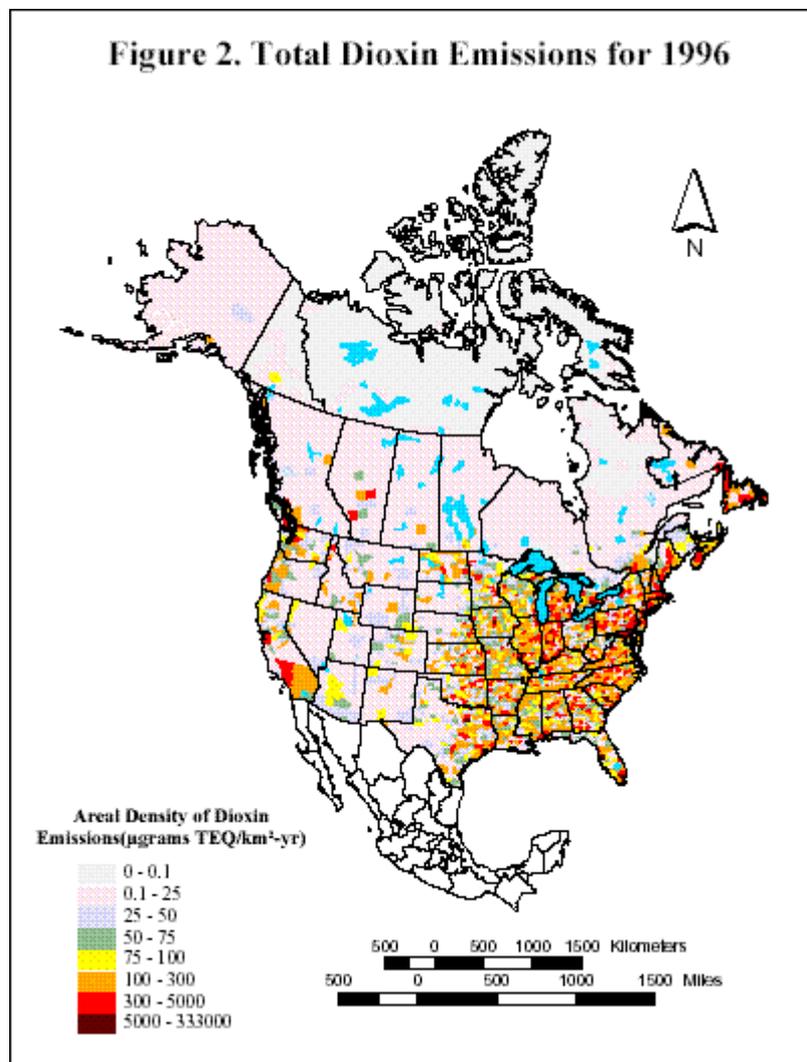


Figure 1. Estimated per-capita 1996 emissions from U.S. and Canadian source categories (g TEQ/person-yr)



Estimated 1996 Per-Capita PCDD/F Emissions (g TEQ/person-yr)

Figure 2. Total Dioxin Emissions for 1996



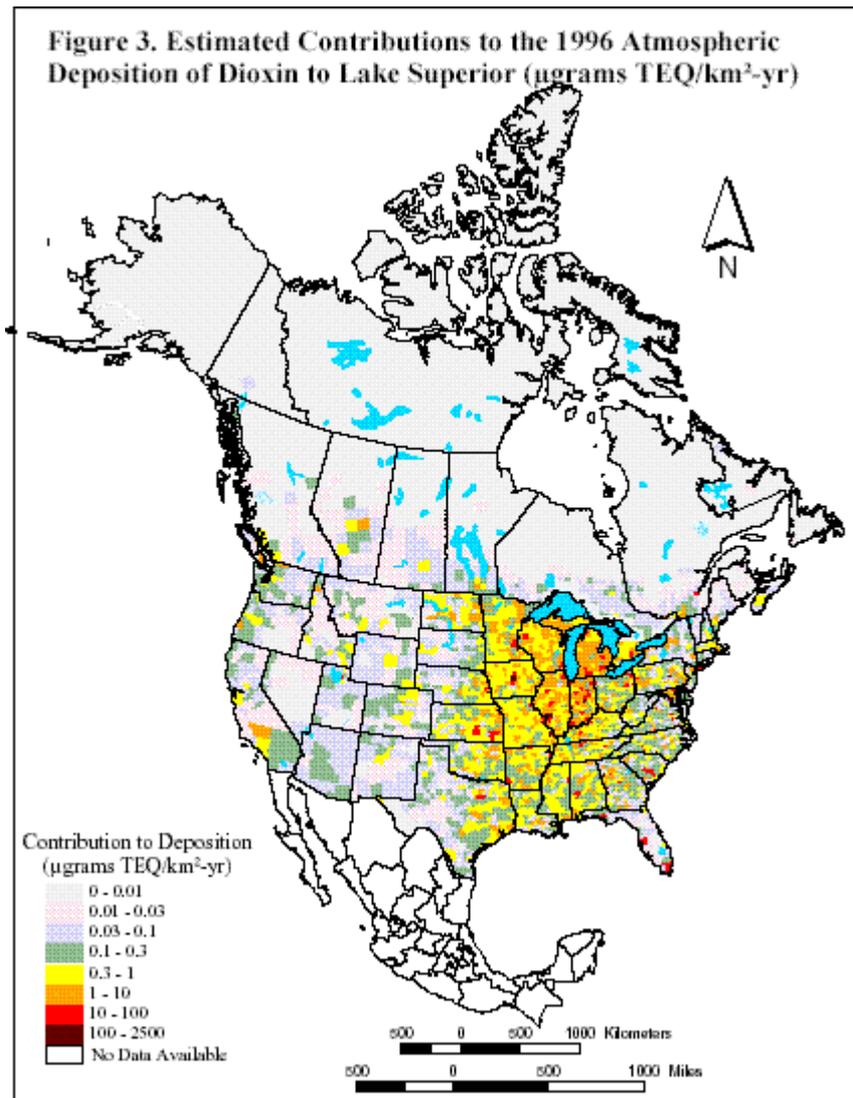


Figure 5. Percent of total 1996 emissions and deposition of dioxin arising from within different distance ranges from Lake Superior.

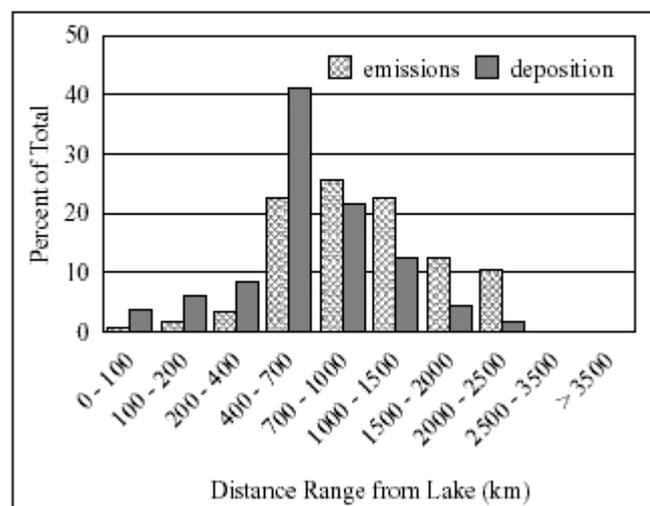


Figure 6. Percent of total 1996 emissions and deposition of dioxin arising from within different distance ranges from Lake Michigan.

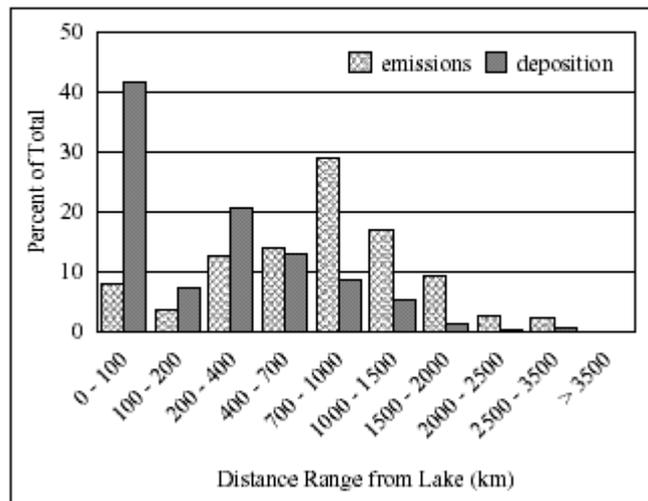
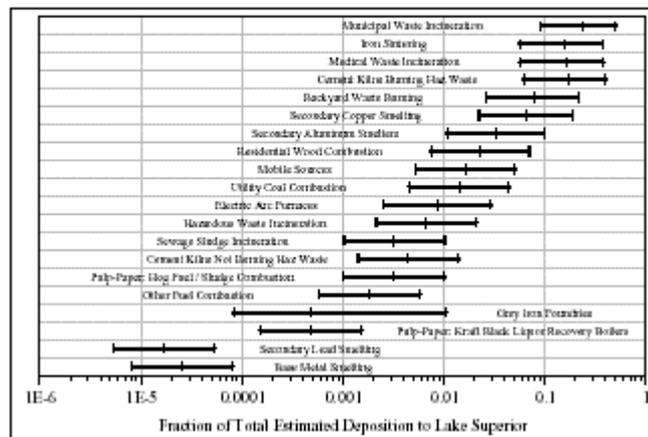


Figure 4. Fraction of estimated 1996 PCDD/F atmospheric deposition contributions to Lake Superior from U.S. and Canadian sources arising from different source categories.



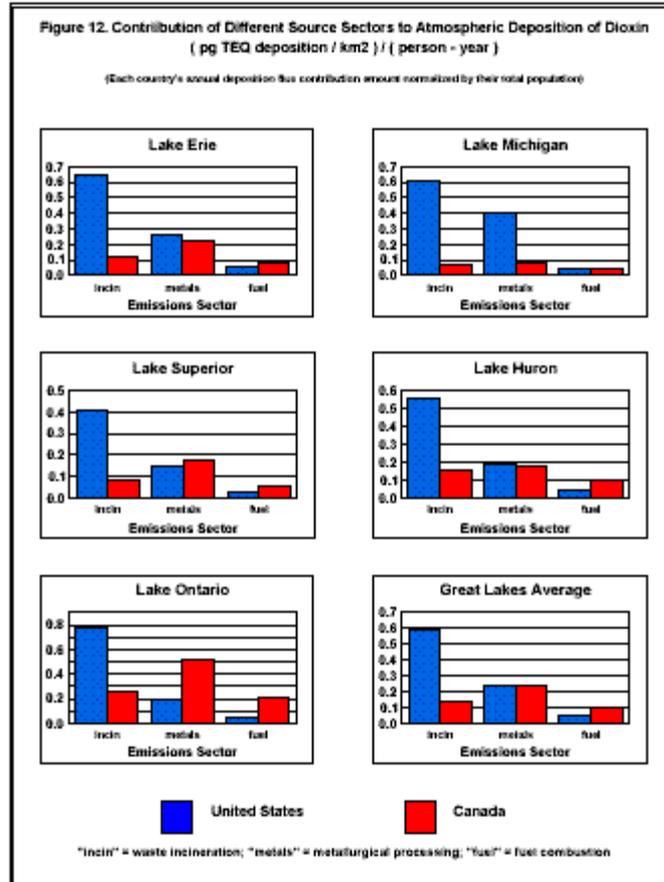


Figure 9. Model-estimated total 1996 deposition for different PCDD/F homologue groups to Lake Superior.

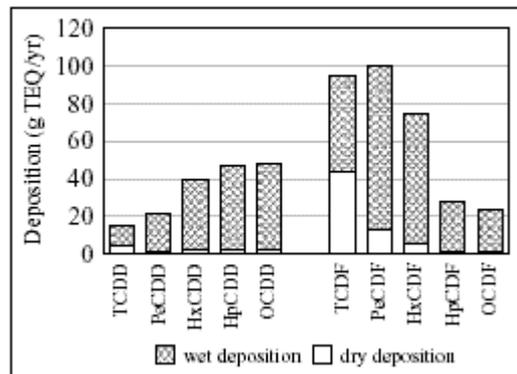


Figure 6. Comparison of model predictions with ambient measurements at month-long sample sites. Sample locations and dates (all dates 1996): A. Mohawk Mtn CT (5/14-6/13); B. Mohawk Mtn CT (8/14-9/13); C. Mohawk Mtn CT (10/22-11/22); D. Northern VT (8/1-8/28); E. Central VT (8/1-8/28).

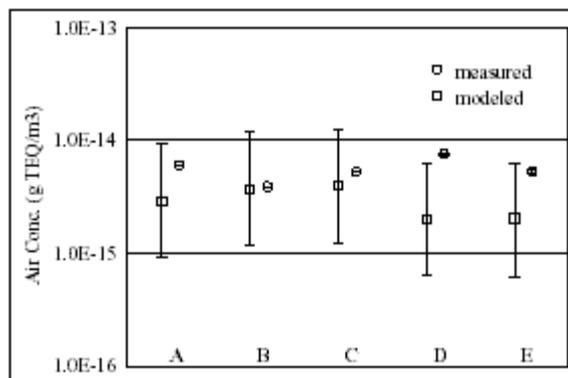


Figure 11. Relative contribution of the seventeen 2,3,7,8-substituted PCDD/F congeners to the overall model-predicted deposition to Lake Superior.

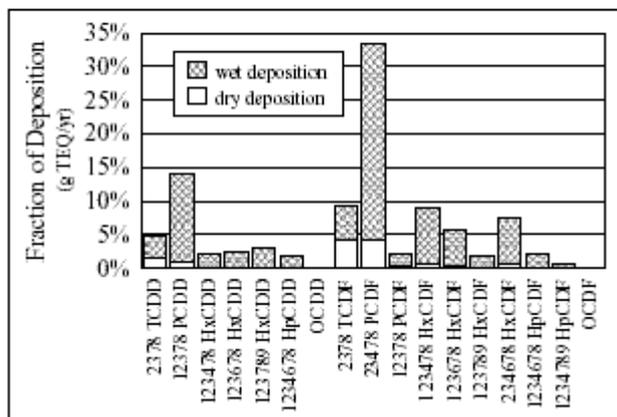
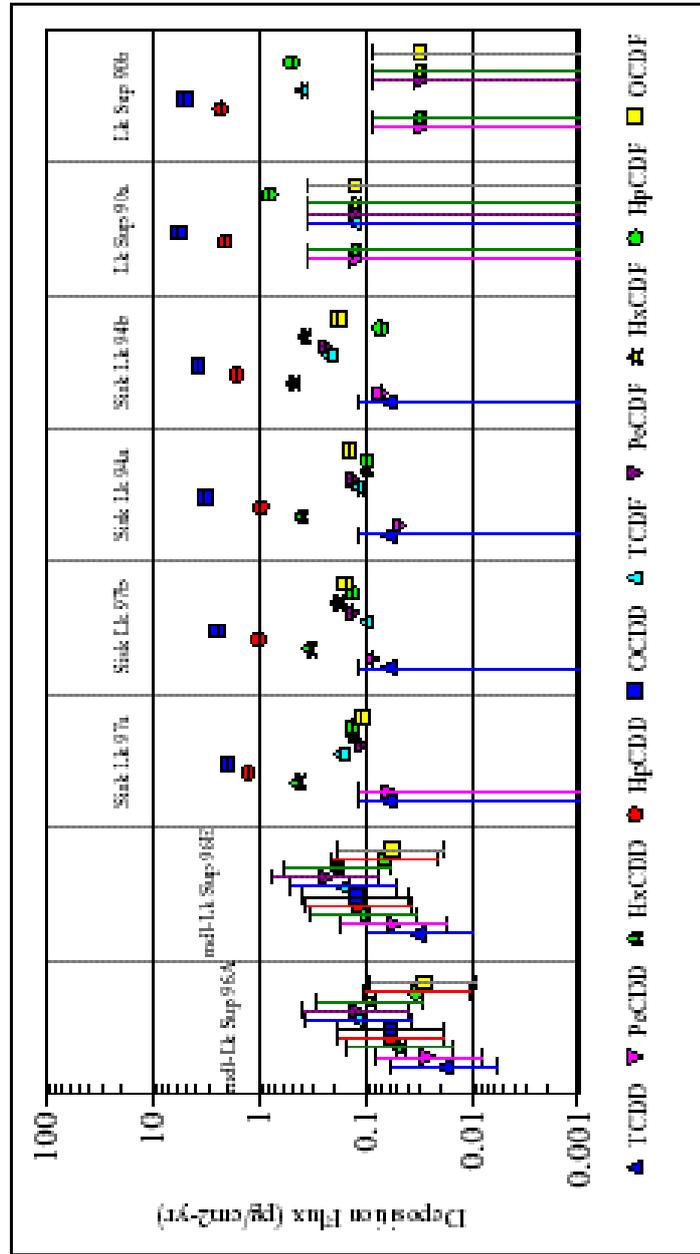
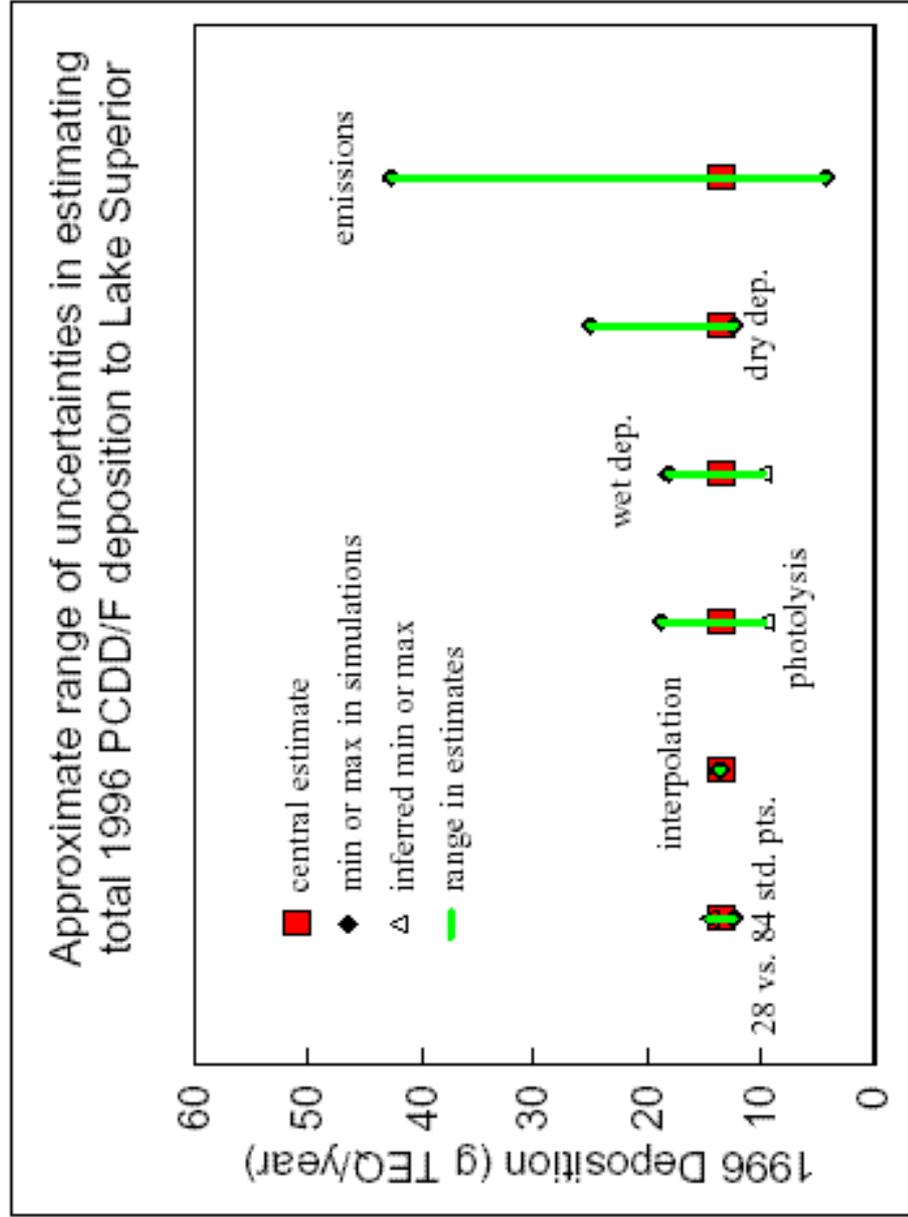
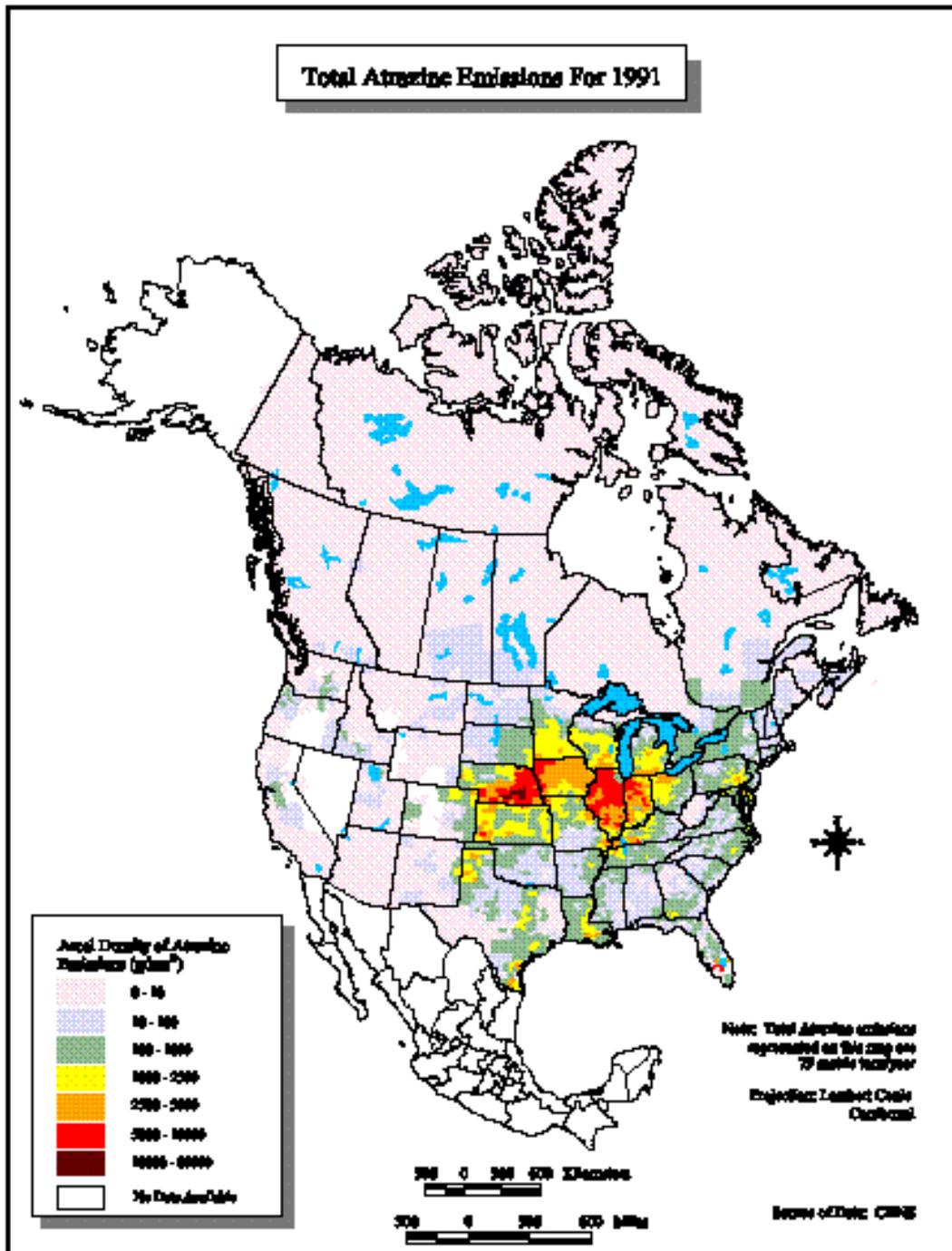


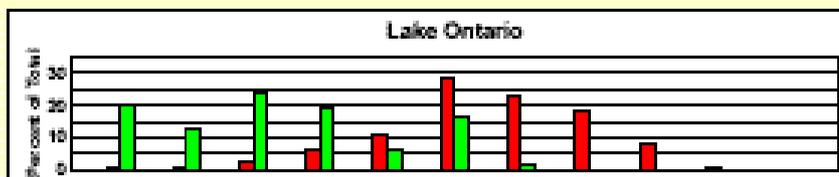
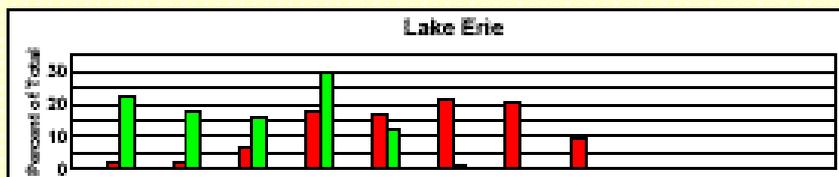
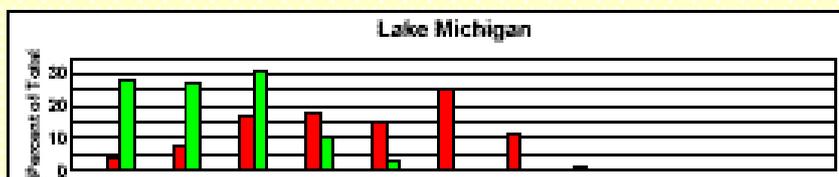
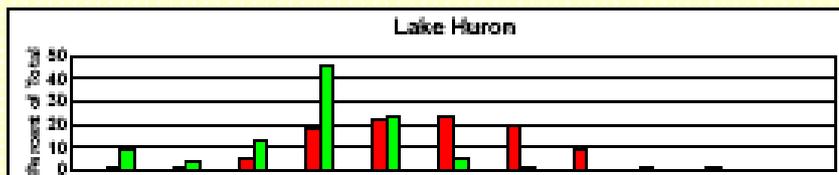
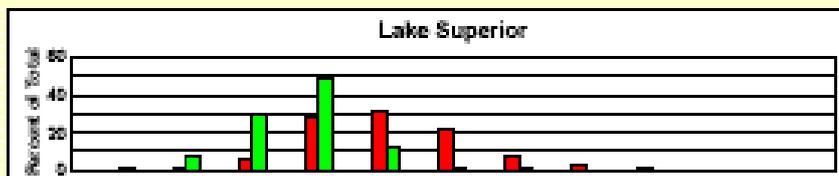
Figure 10. Measured 1994 and 1997 fluxes to Siskiwit Lake on Isle Royale (Baker and Hites, 2000) and 1990 fluxes to Lake Superior (Pearson et al., 1997, 1998), compared to 1996 modeled fluxes. Non-detects in the measurements are plotted with a mid-point value at 1/2 the detection limit (DL), with a range of 0 to the DL. Model estimates shown using dry deposition methodologies A and E. Measurements in Isle Royale and Lake Superior were each reported at 2 sediment sampling sites.







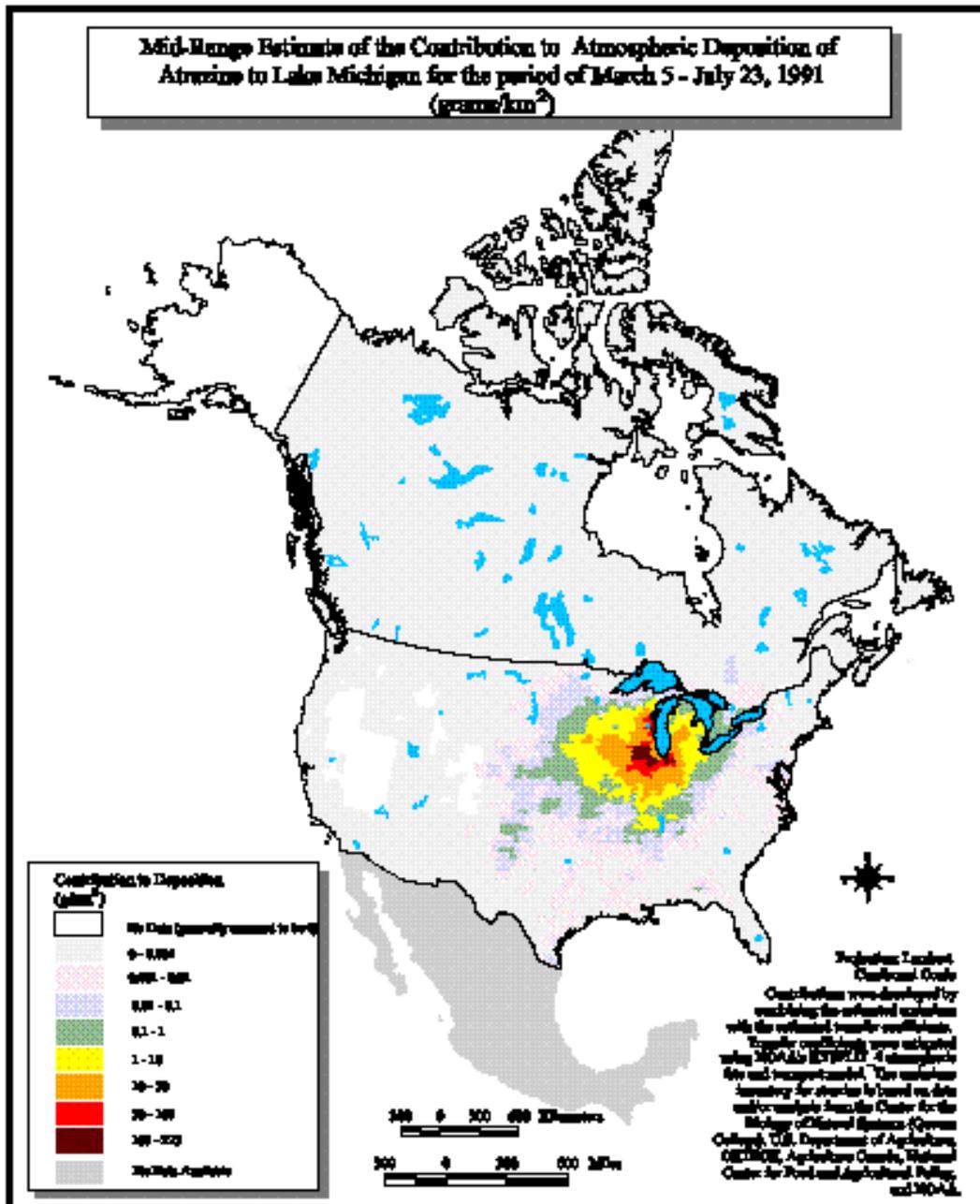
**Percent of Total 1991 Emissions or Deposition of Atrazine Arising From Within a Given Distance Range of Each Great Lake**

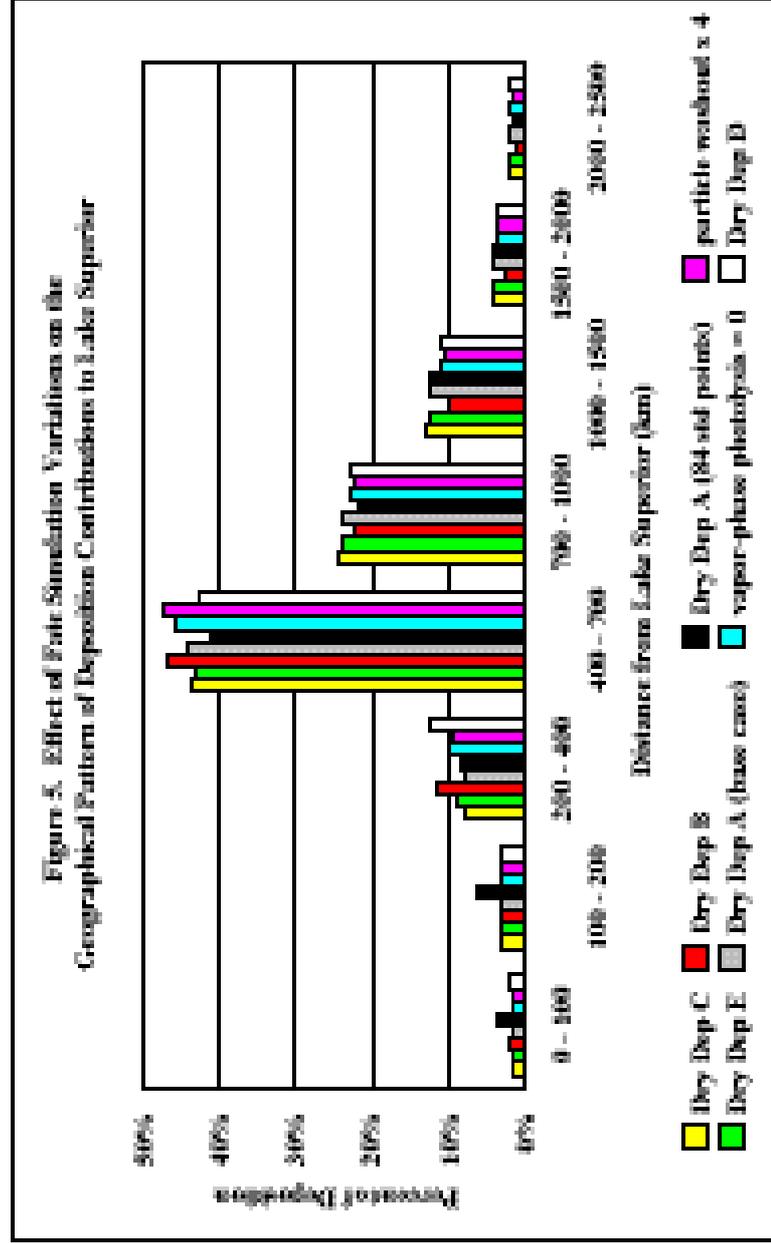


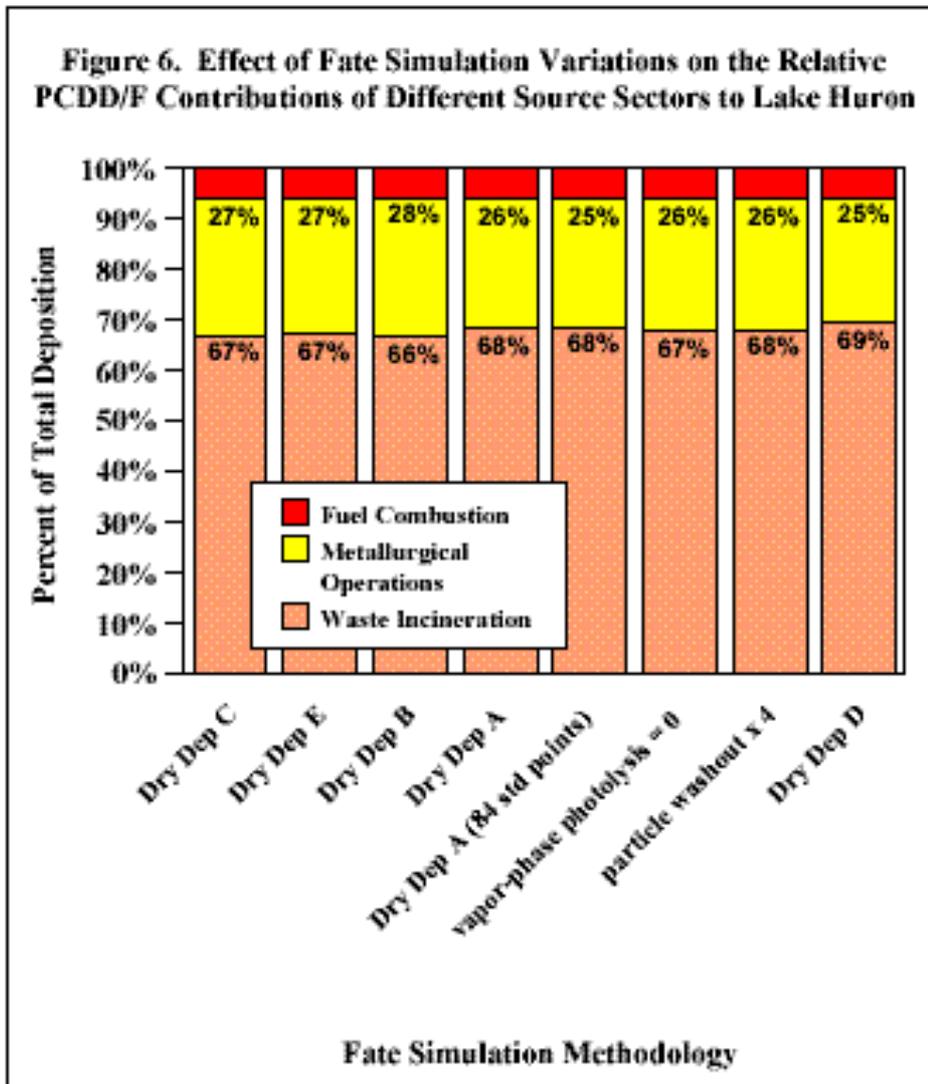
0-100 100-200 200-400 400-700 700-1000 1000-1500 1500-2000 2000-2500 2500-3500 3500-5000 5000-10000

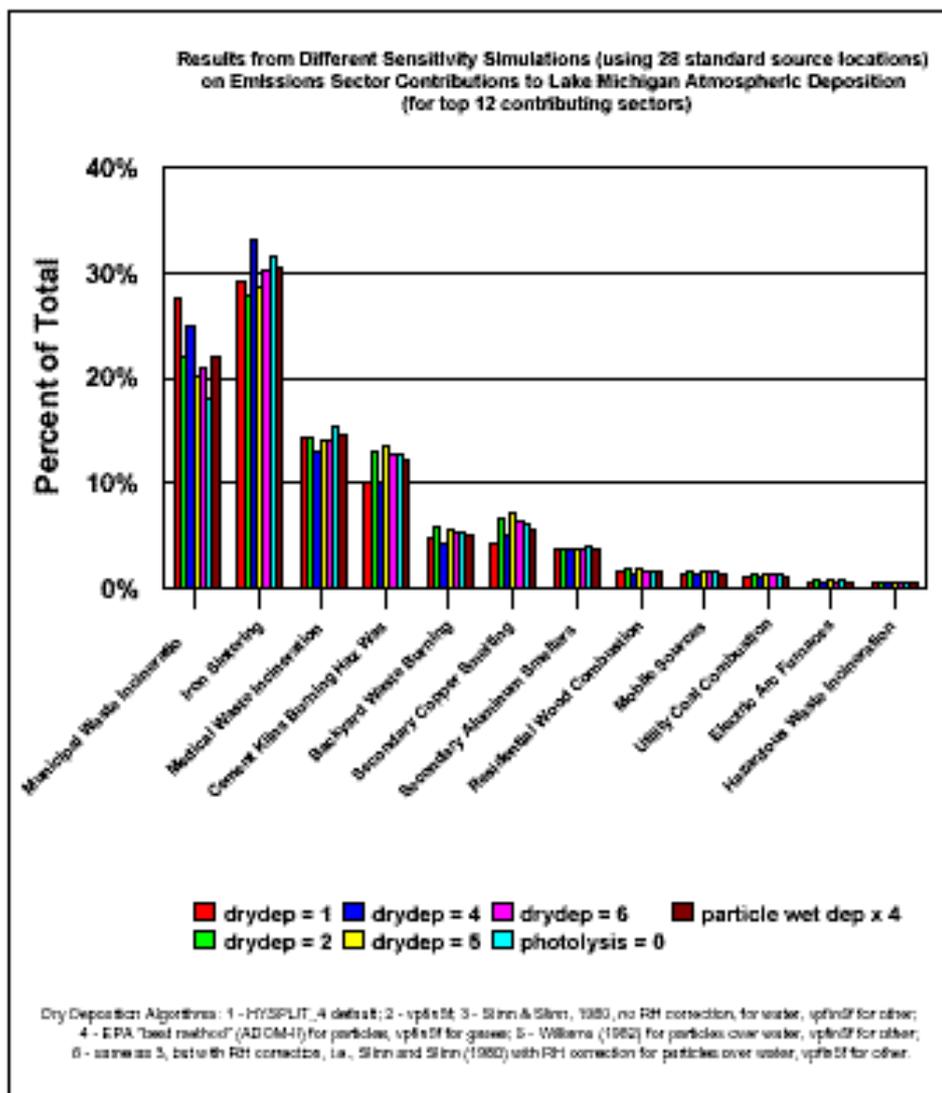
Distance Range from a Given Great Lake (kilometers)

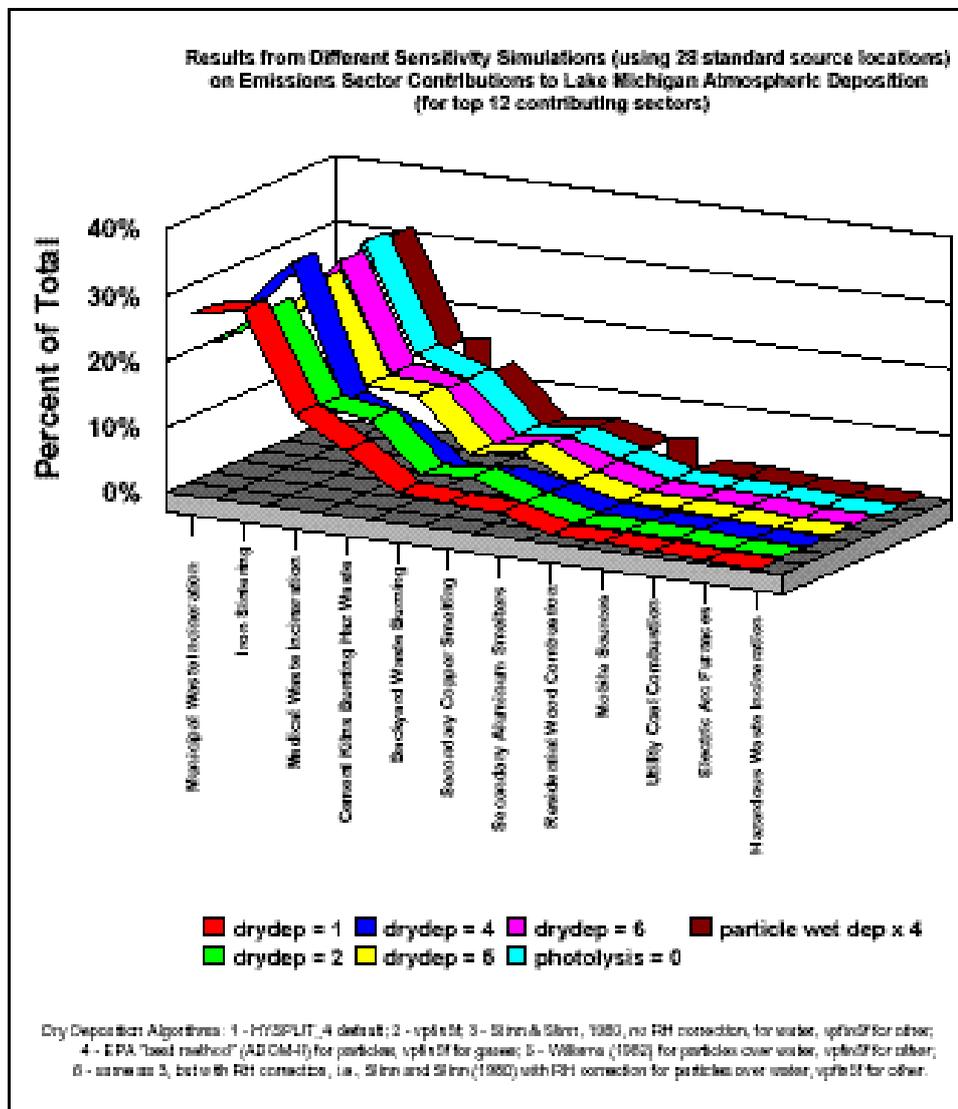
EMISSIONS DEPOSITION

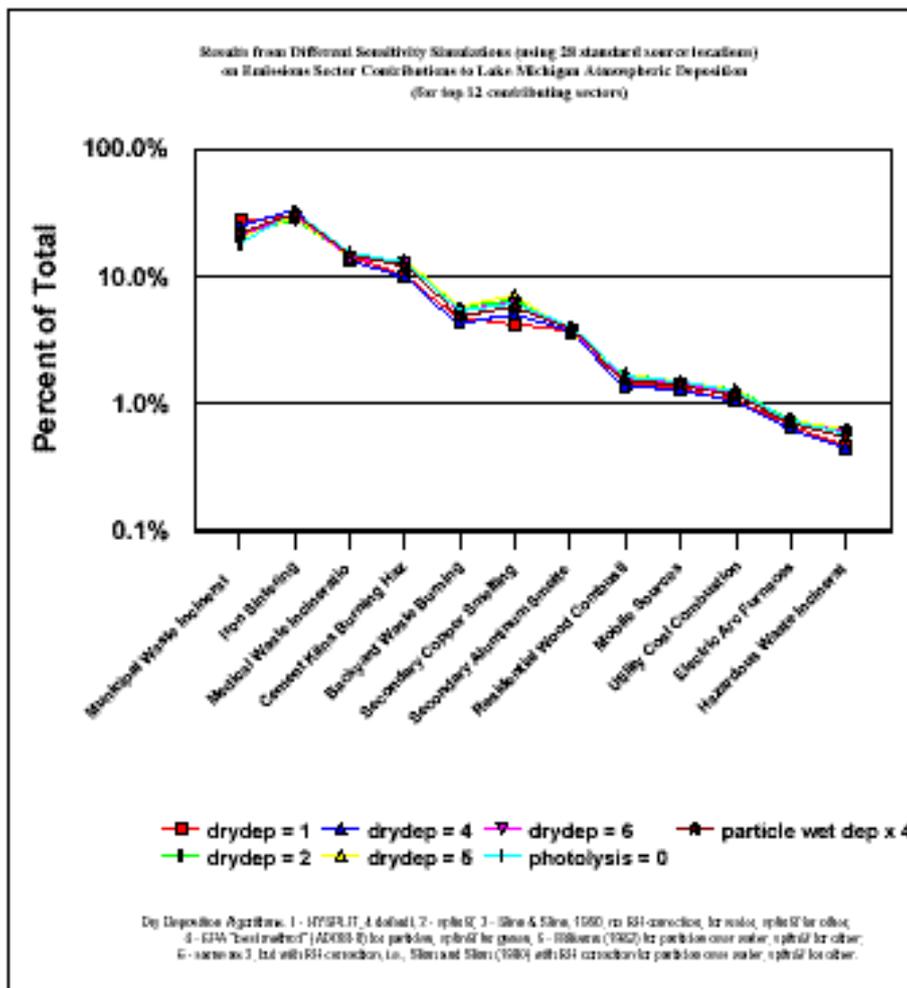










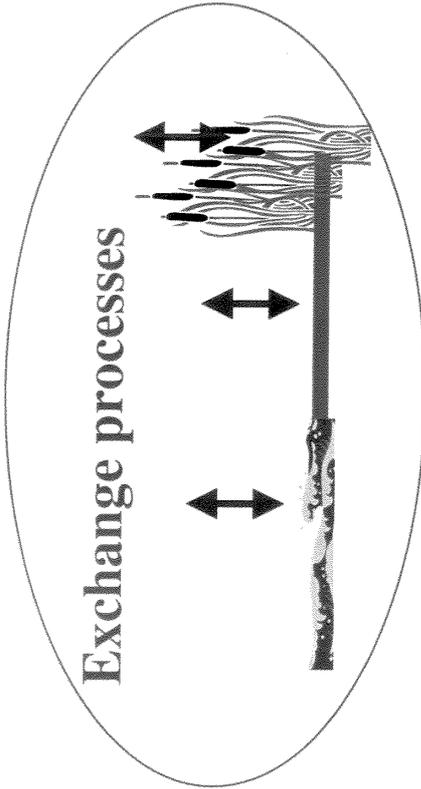
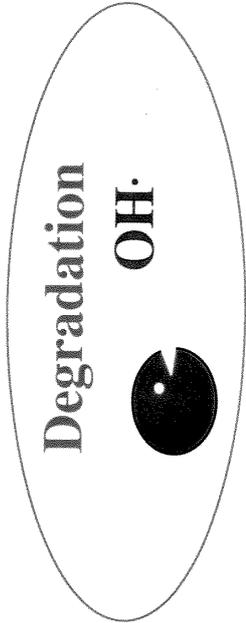




# Studying Global Fate.....



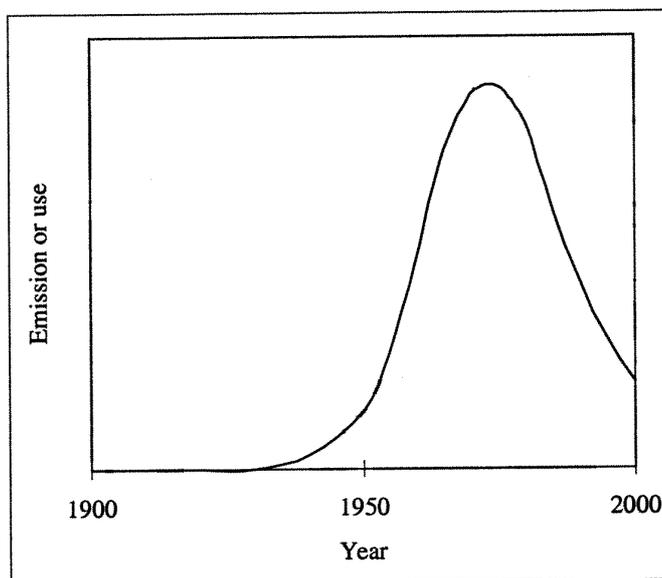
*Spatial  
&  
Temporal  
Studies*



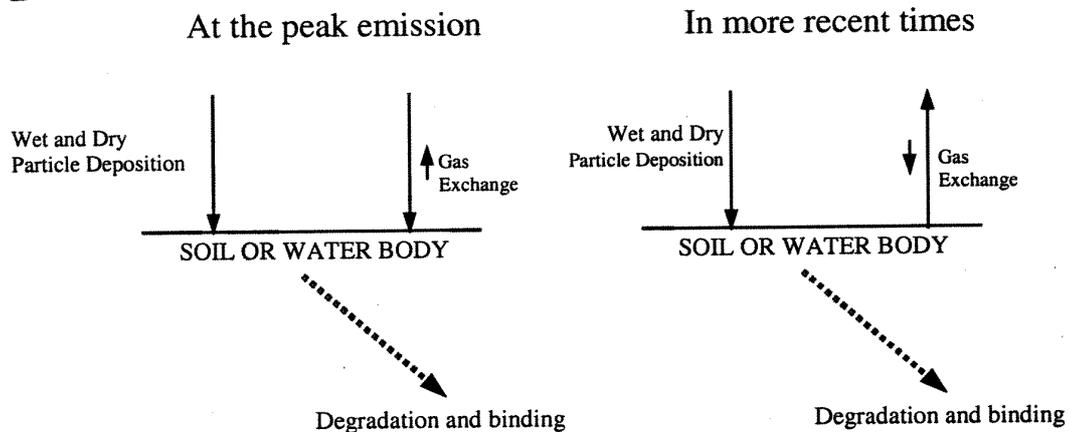
*Small Scale Process Studies*

**Figure 3: Typical POPs primary emission time trend (A) and air-surface exchange processes (B).**

**A**

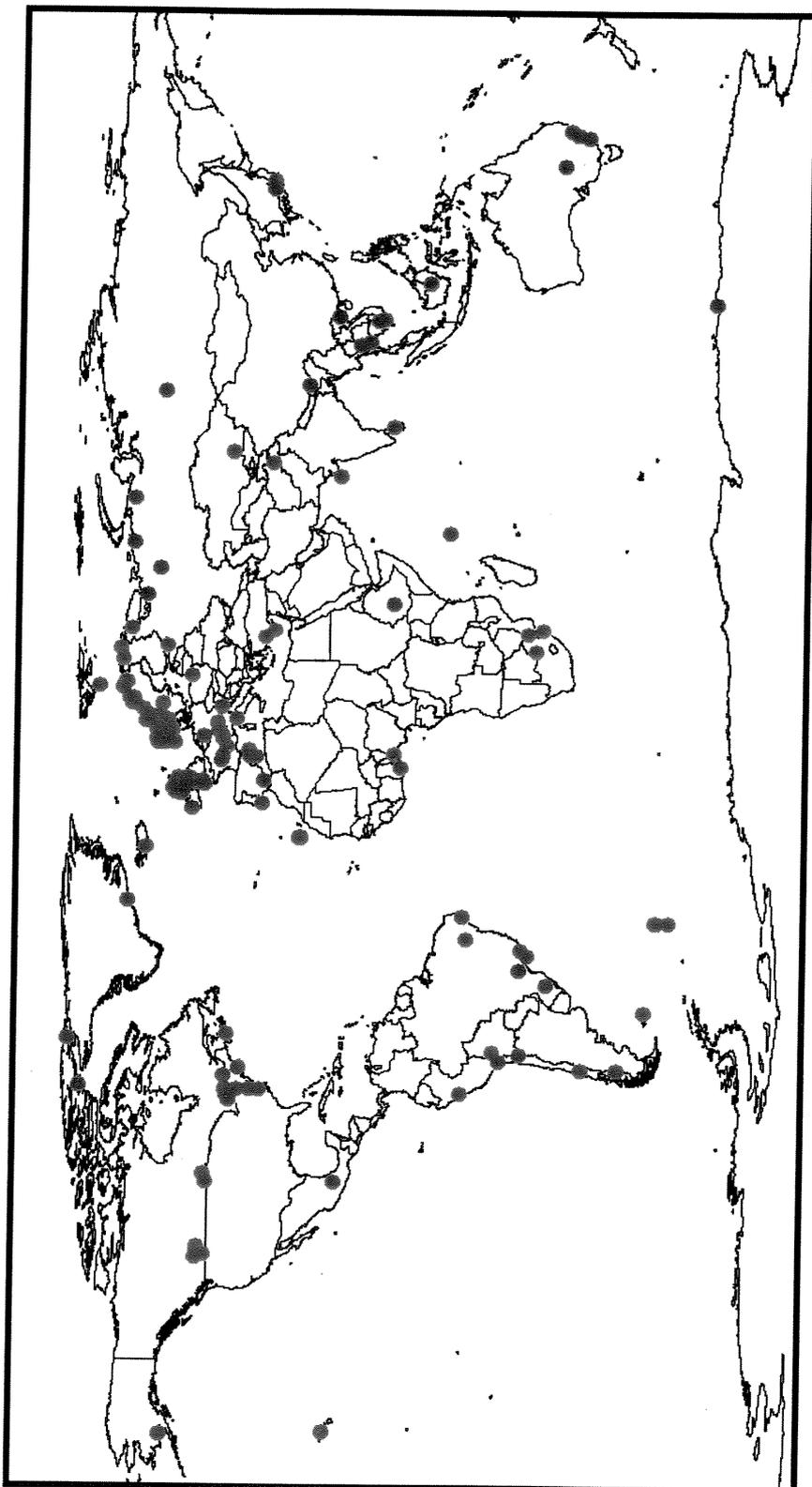


**B**

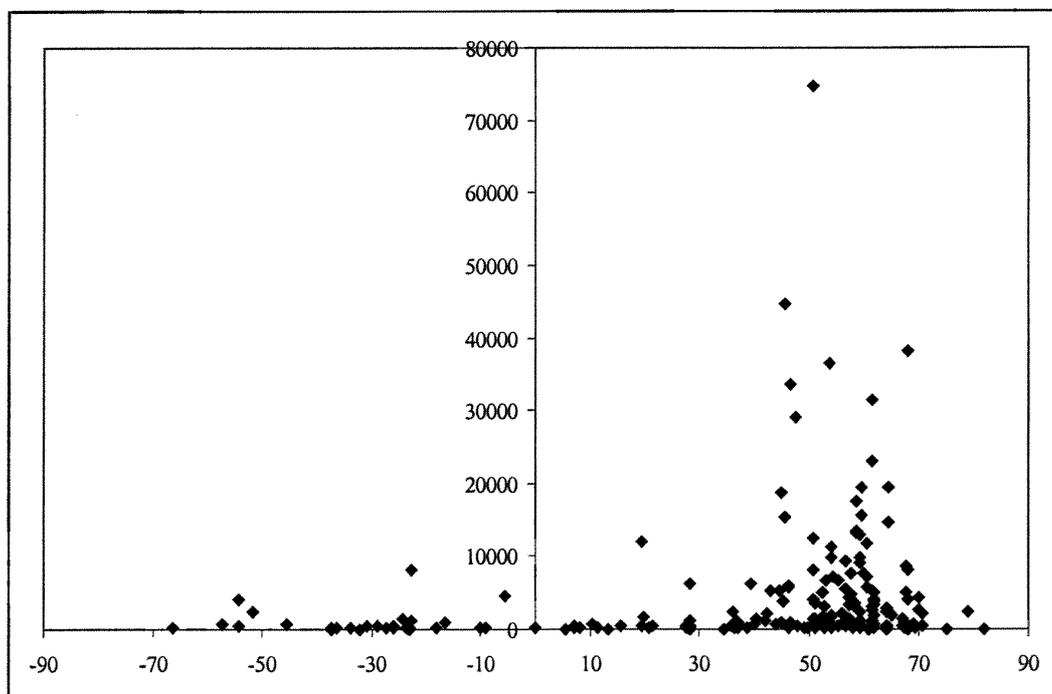
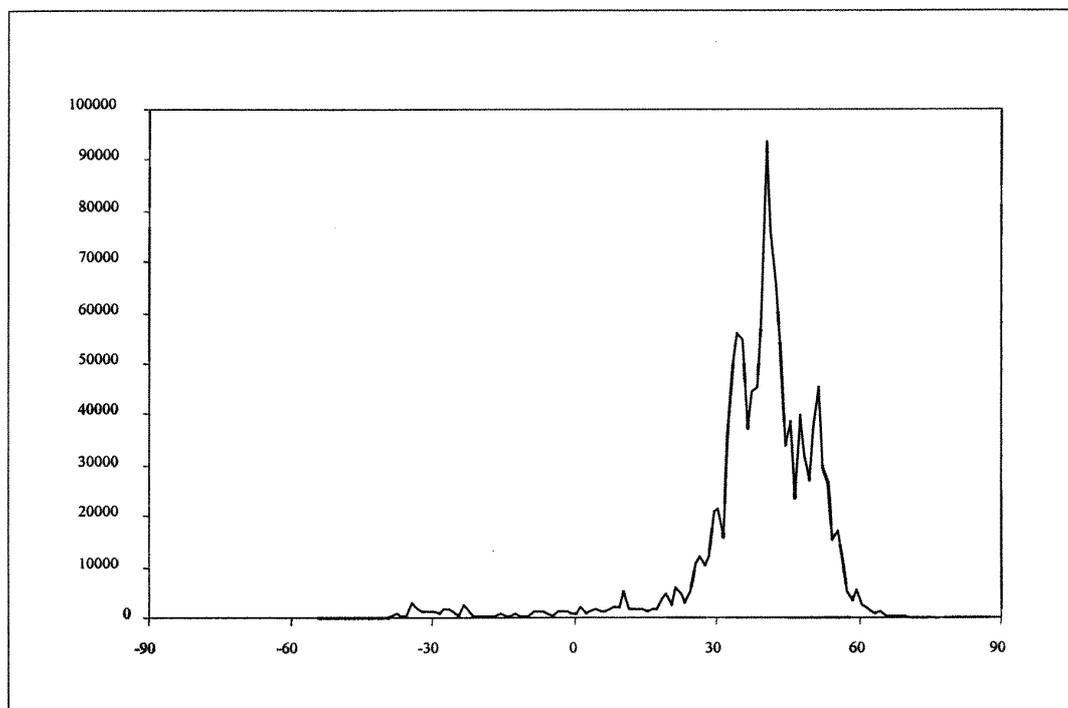


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Figure 1 – Global soil sampling locations.

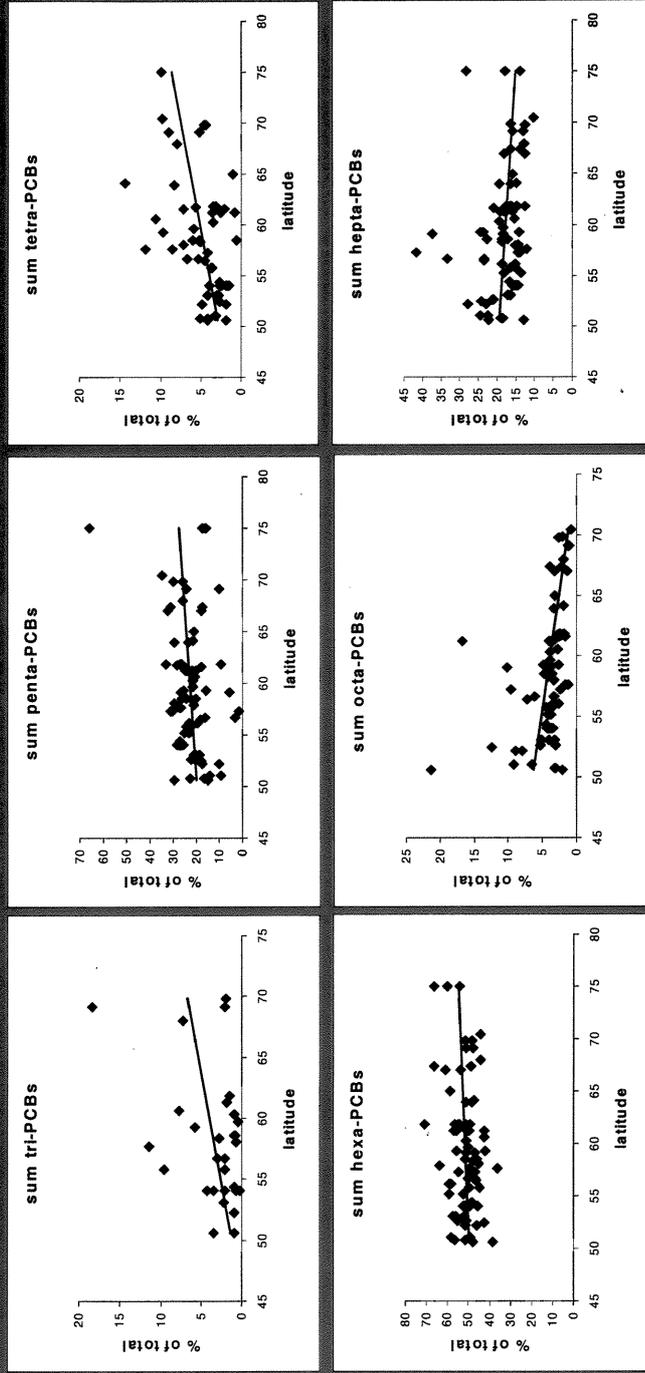


**Figure 1: The latitudinal distribution of: a. global PCB usage (tonnes); b. PCBs in surface soils (pg  $\Sigma$ PCB/g dry weight).**

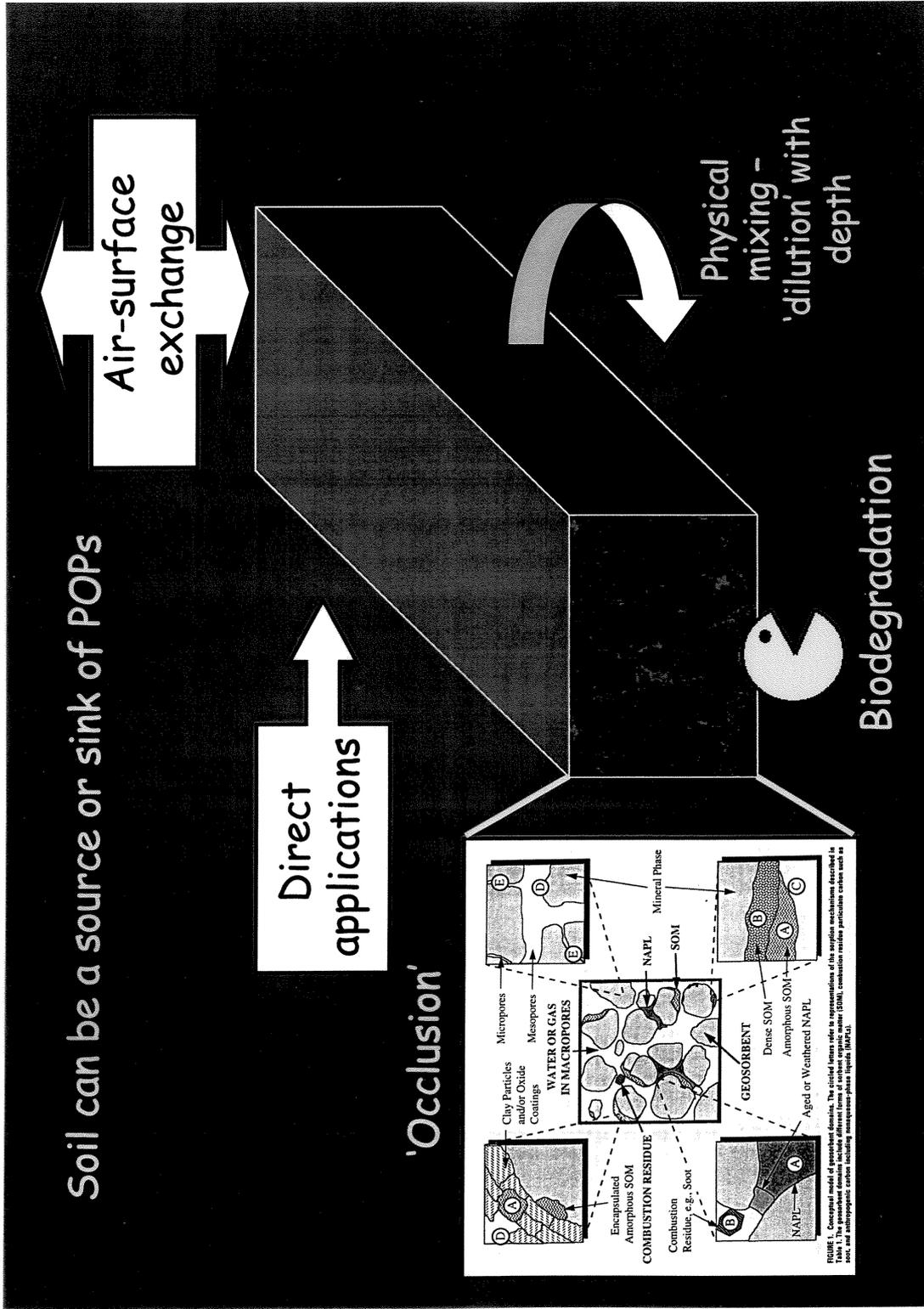


# Relative concentration vs. latitude

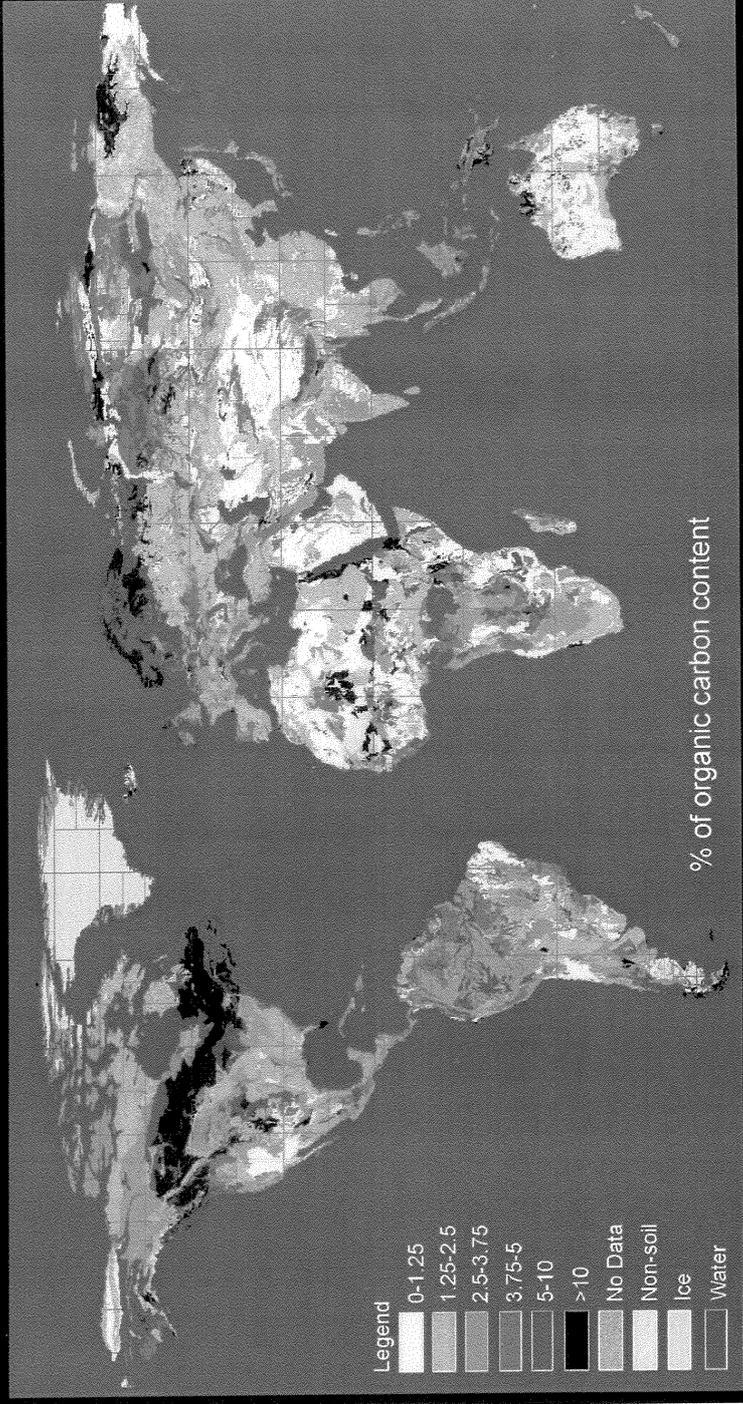
## Sum homologue groups - % of total

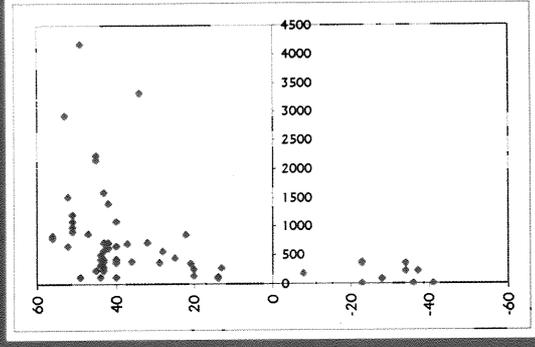
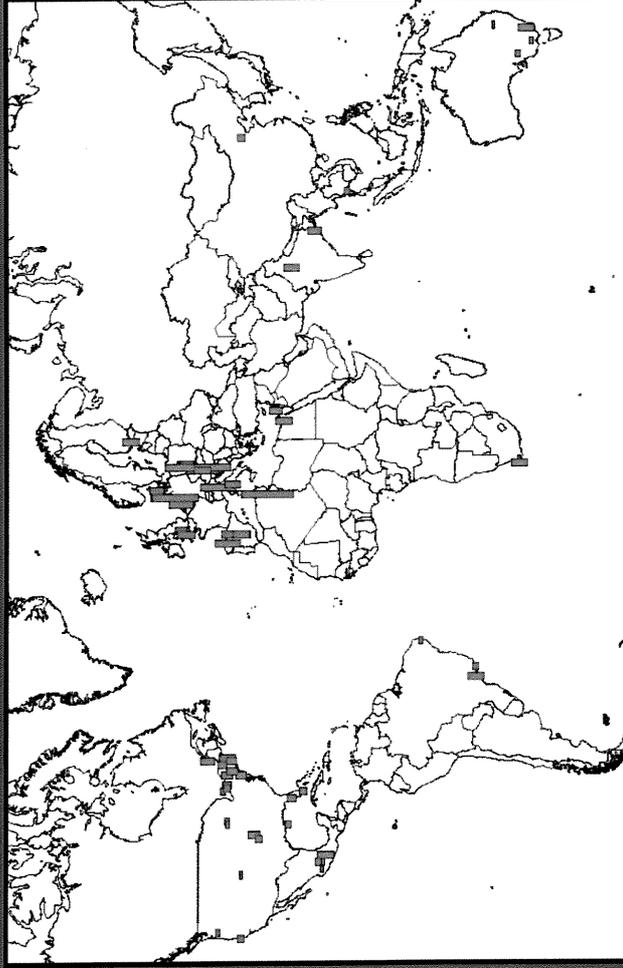


All except hepta - slope significant at  $P < 0.05$   
⇒ Indication of global fractionation

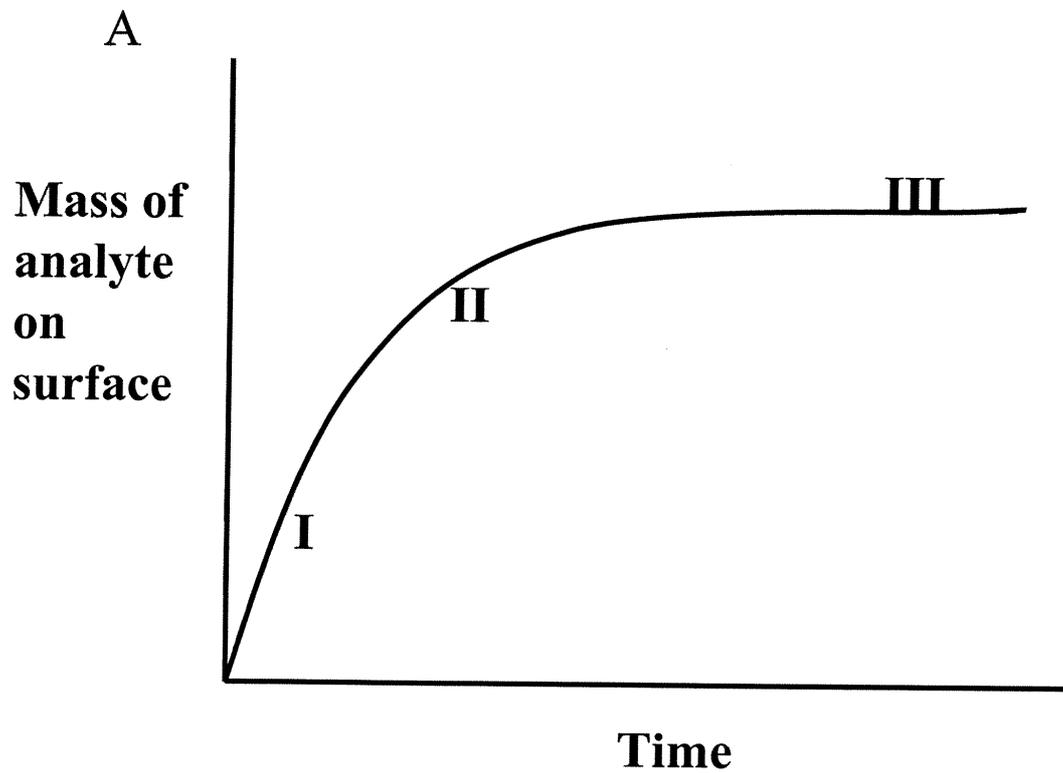


# Global soil organic carbon map



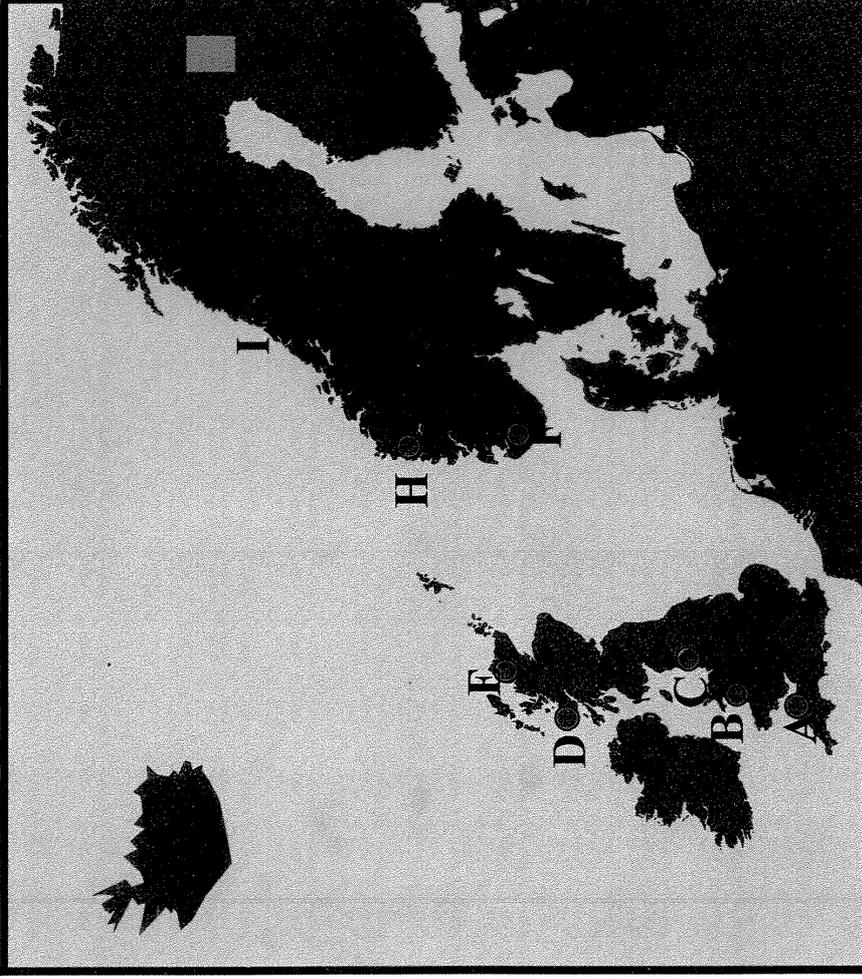


**PCB-138**  
Concentration range from 110-3330pg/g

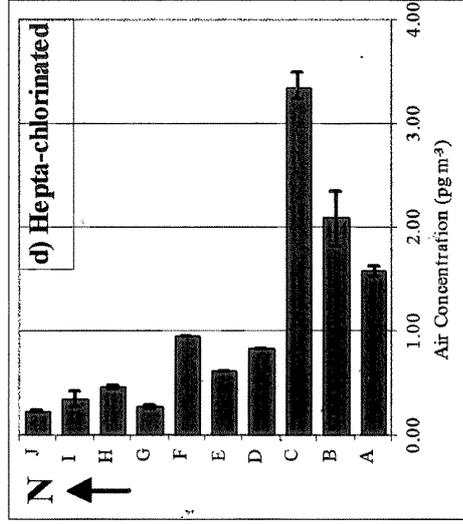
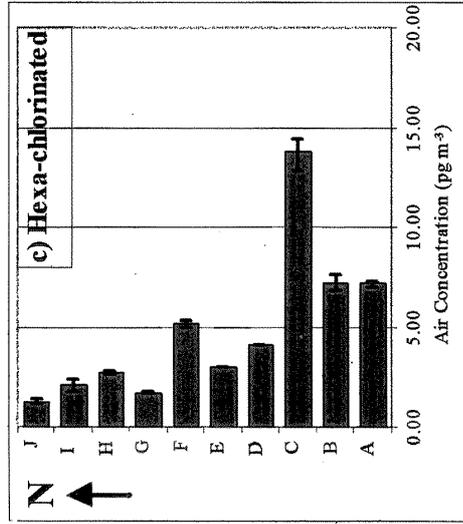
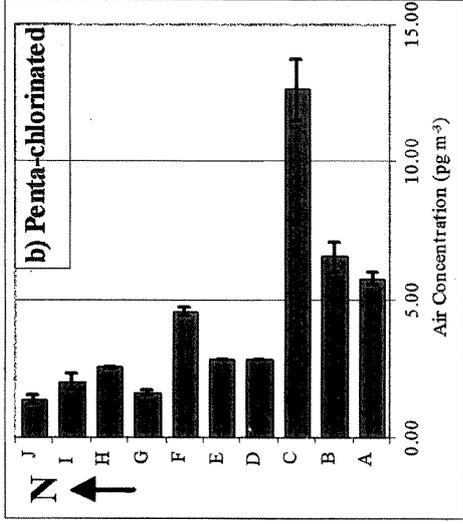
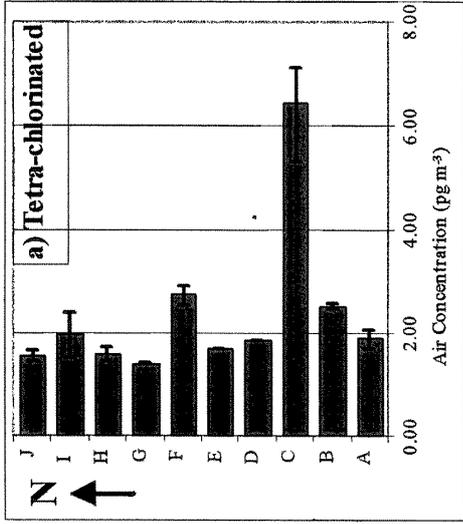


- I** Initial linear uptake
- II** Curvilinear portion as equilibrium partitioning is approached
- III** Equilibrium between the surface and gas phase concentrations

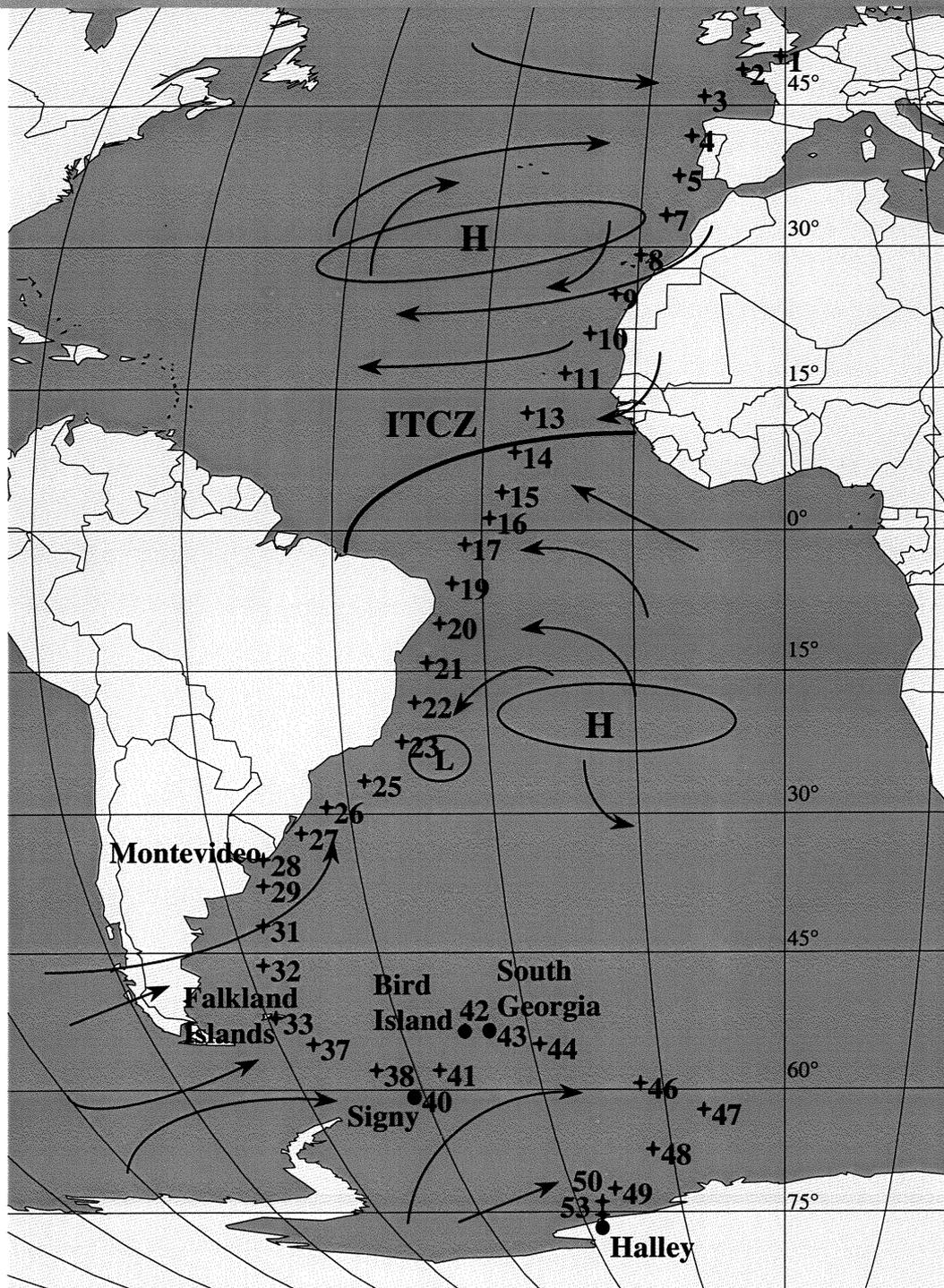
# Map of USGS-SPMD Sample Sites



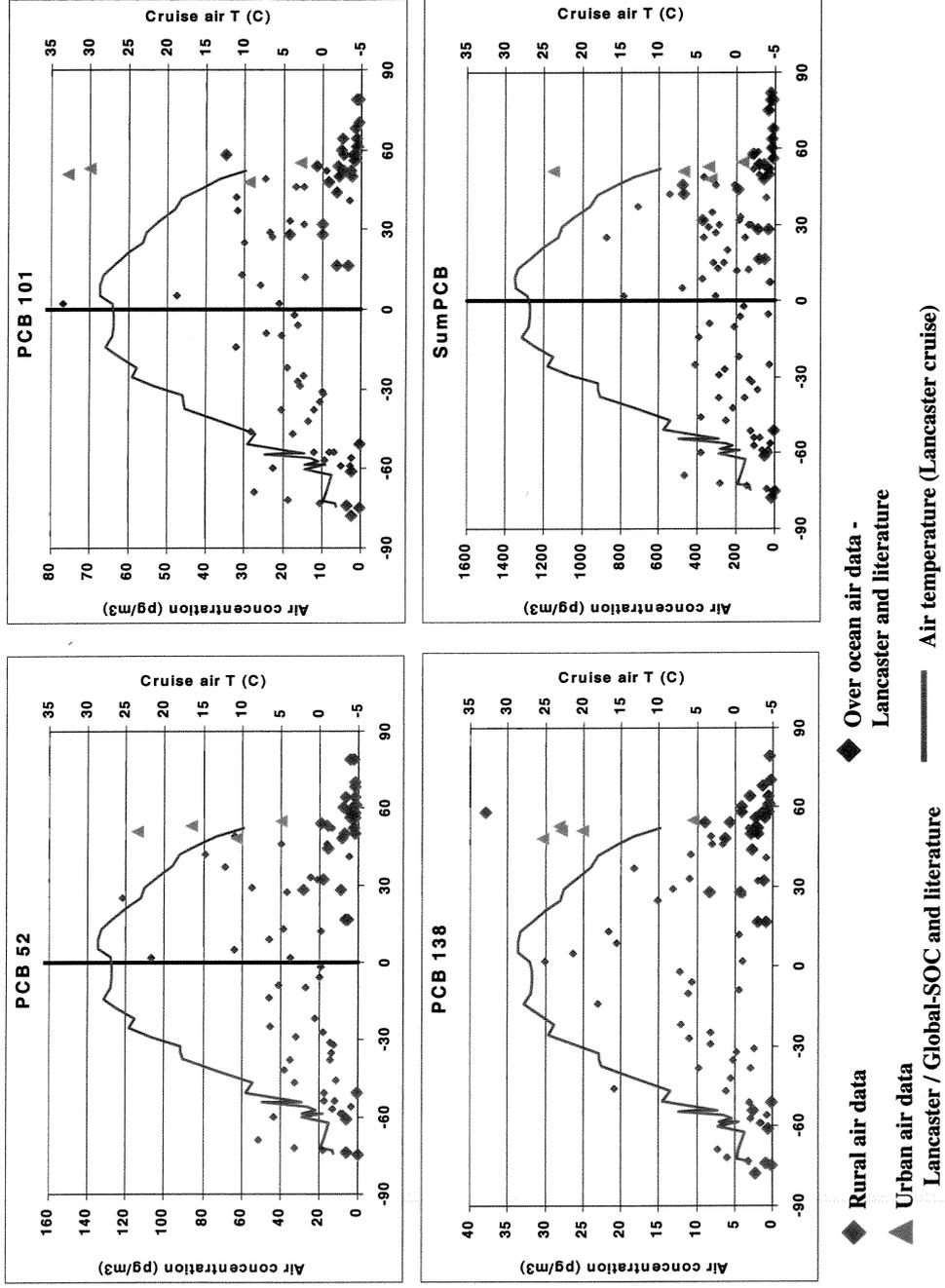
F4



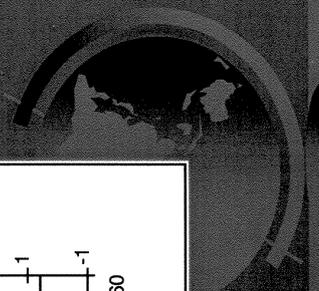
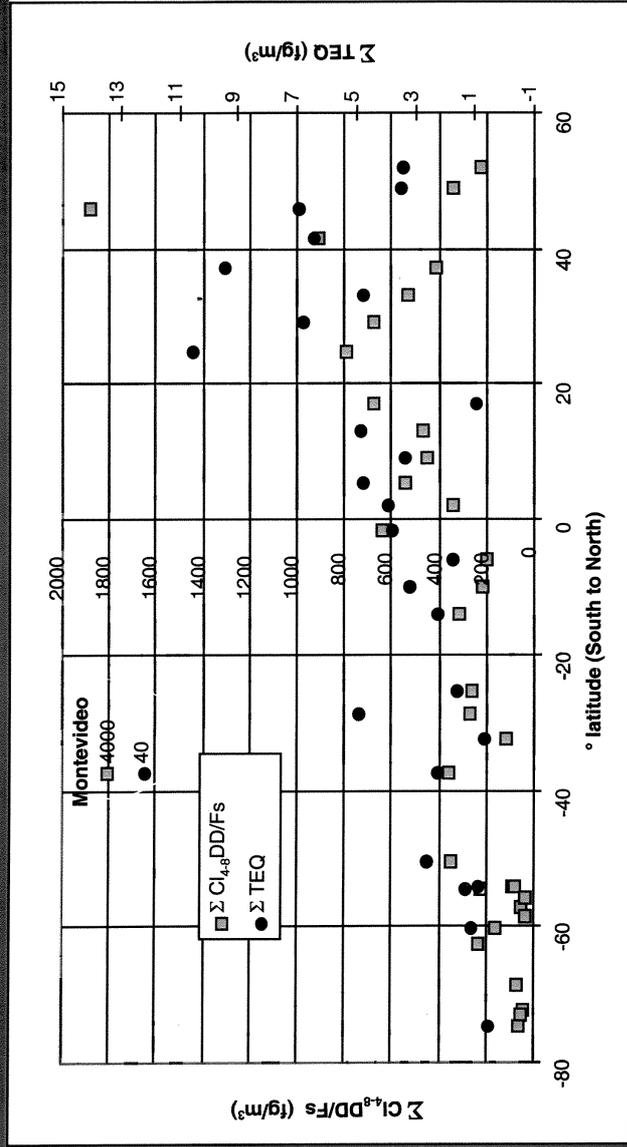
## North-South Transect - Sampling Stations

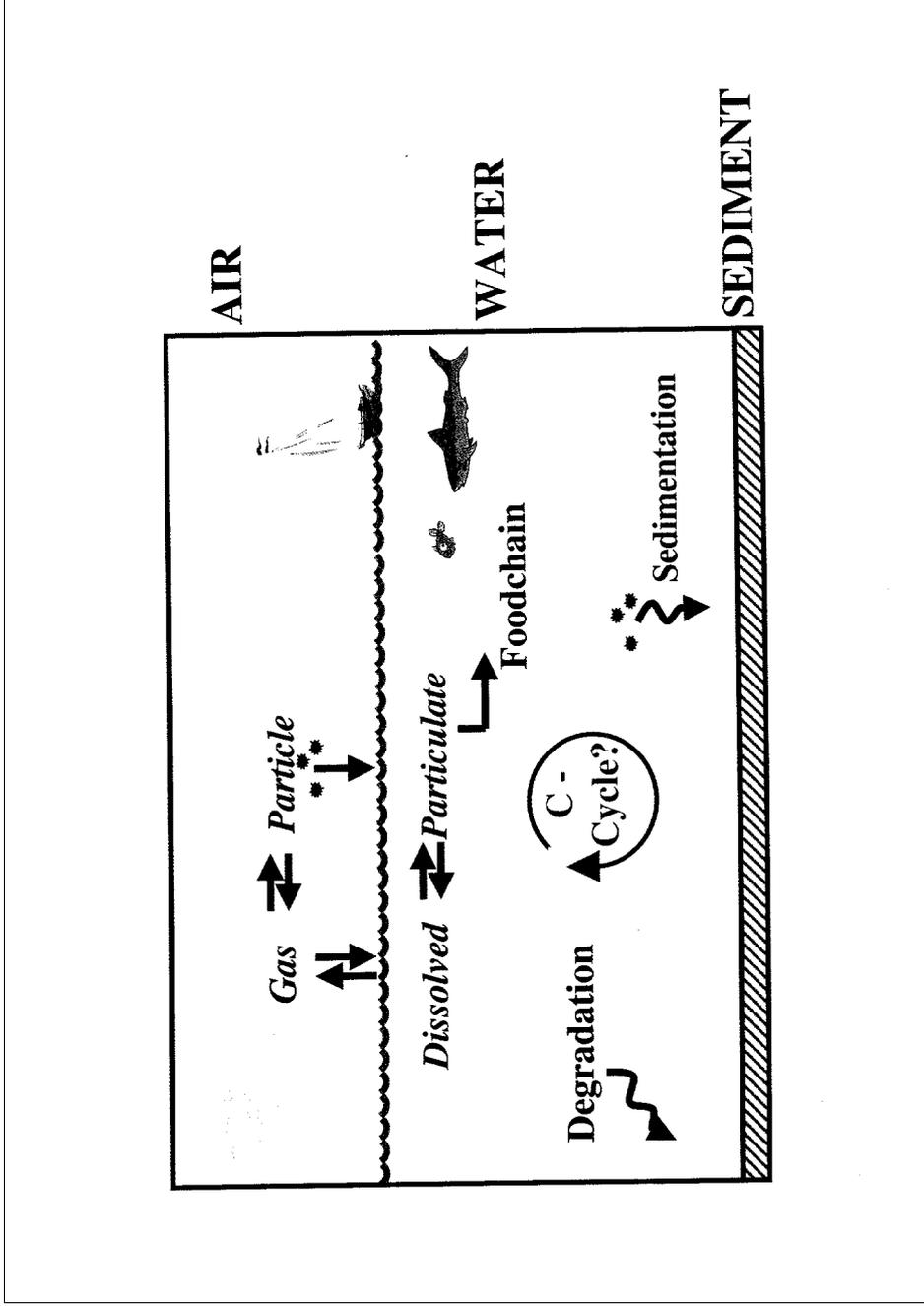


General circulation after D. Jacob, adjusted for local backtrajectories



# $\Sigma\text{Cl}_{4,8}\text{DD/Fs}$ and $\Sigma\text{TEQ}$ South to North





## Interesting 'global' questions (1):

- What is 'controlling' the rate of change in air?  
Implications for the future?
- Degradation in soils?
- How should we regard the soil?  
'permanent sink' / 'long-term store'

### 6.2.5 *Peter Outridge*

#### **Discriminating natural from anthropogenic sources of lead in Canadian Arctic lakes**

By P.M. Outridge, Geological Survey of Canada, 601 Booth St., Ottawa K1A 0E8, Canada. (E-mail: [outridge@nrcan.gc.ca](mailto:outridge@nrcan.gc.ca))

Stable Pb isotopic profiles in dated lake sediment cores were used to gauge the relative amounts and possible sources of anthropogenic Pb deposited from the atmosphere in different regions of the Canadian Arctic. A distinct north-south difference was found. In the four High Arctic lakes (i.e. north of 66°N) in this study, recent Pb isotopic shifts or concentration increases attributable to industrial Pb were negligible. The maximum possible contribution from industrial Pb was 0-19% of acid-leachable Pb in the 1980s or 1990s. In contrast, two lakes in the Hudson Bay region displayed significant reductions of Pb isotope ratios and three- to five-fold increases of Pb concentrations in modern sediments, corresponding to industrial Pb inputs of at least 72-91% of leachable Pb. Eurasian industrial Pb is known to dominate the High Arctic atmosphere. A possible explanation for its negligible influence on northern lake sediments is that atmospheric Pb deposition at those latitudes is reduced compared with southern regions, and is small compared to local geological inputs. <sup>210</sup>Pb deposition declines with increasing latitude, apparently because of declining precipitation rates; stable Pb deposition may be similarly affected. In Hudson Bay, the introduction of anthropogenic Pb into the western Hudson Bay lowlands began later (at ca. 1880 AD) than in southeast Hudson Bay (1800 AD). This difference, and variations in the post-1900 Pb isotope temporal trends, suggested that the predominant industrial Pb source region in western Hudson Bay is Eurasia while in southeastern Hudson Bay it is Canada/US with a minor Eurasian component.



## 6.2.6 JoLynn Carroll

### Fluxes and Biogeochemical Processes for Contaminants in Northern Seas

*JoLynn Carroll*  
*Akvaplan-niva as*  
*Polar Environmental Centre*  
*N-9100 Tromsø NORWAY*  
[Jolynn.carroll@akvaplan.niva.no](mailto:Jolynn.carroll@akvaplan.niva.no)

Knowledge of pollutants in the environment - their behaviour and distribution together with supporting information on ocean, ice and air transportation is vital to developing sound management plans for monitoring of the Arctic. Monitoring programs implemented by the Norwegian authorities will be based in part on available knowledge on transport pathways for contaminants gathered as part of the Norwegian Ministry of the Environment and Ministry of Foreign Affairs programme, "Transport and Fate of Contaminants in the Northern Seas." Through this programme, new information on contaminants has been gathered and assessed on several important topics with some highlights mentioned below.

- (1) An evaluation was conducted of the magnitude of contaminant fluxes in sediment-laden sea ice for a variety of contaminants (heavy metals, radionuclides, persistent organic pollutants). In the Kara Sea, contaminant fluxes associated with the process of frazil ice formation were found to be small. For example, realistic fluxes of the radionuclide Cs-137 determined for the years 1976, 1985, 1987 and 1988 are 2%-16% of the worst-case scenario value of  $8.3 \times 10^{12}$  Bq/yr (Figure 1).
- (2) Historical data were gathered and checked for quality assurance on contaminant concentrations in ice, snow and water for some key areas of the Russian arctic. The contaminants addressed were oil hydrocarbons, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenols (PCBs), organochlorine compounds (OCs), and heavy metals (HMs). The temporal coverage for the data is 1992-1995. For the Pechora system, contaminant and hydrological data were combined and estimates of contaminant fluxes from this river system were calculated. The largest fluxes for all contaminant classes occur during summer time. Polycyclic aromatic hydrocarbons (PAHs) dominate the exchanges from the River to the Barents Sea. The data provide baseline data so that changes in contamination levels will be more easily detected in future years (Figure 2).
- (3) Improved mathematical formulations for simulating geochemical processes in transport models, including (i) uptake, redistribution and release of contaminants by sea ice, (ii) chemical exchange processes between sea-ice and atmosphere, (iii) sediment-seawater interactions and (iv) chemical exchange reactions at the sea-floor. The future incorporation of improved algorithms into transport models are expected to lead to more realistic assessments of the transport pathways and impacts resulting from contaminants detected in the environment through monitoring programmes.
- (4) Discharge scenarios for model validation and testing including potential acute sources and chronic discharges/emissions/ atmospheric fallout. In total, the ability to test and validate transport models using the resources compiled in

this project is expected to lead to better knowledge of potential contamination 'hotspots.' Knowledge of 'hotspots' will be essential when planning long-term monitoring activities.

A summary of these results will be presented at the Workshop. The information discussed in this presentation is also available in a series of project reports and publications from the presenter.

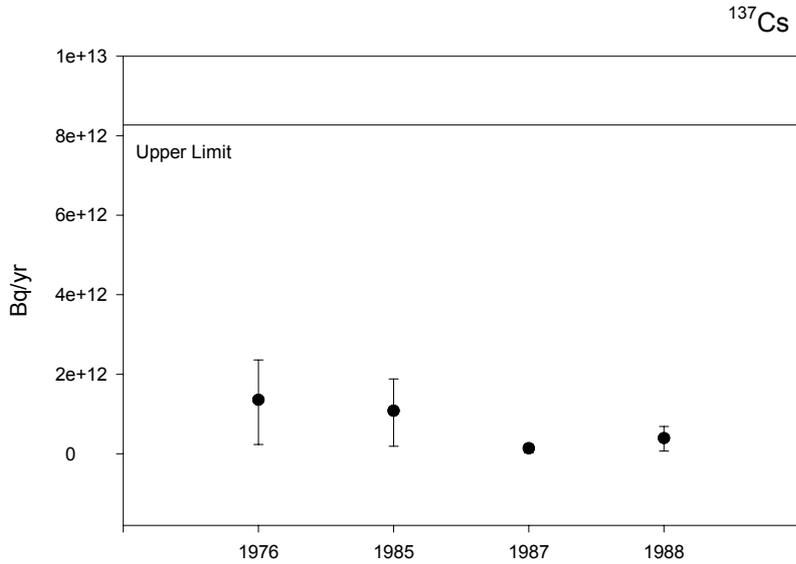


Figure 1: <sup>137</sup>Cs fluxes in sediment-laden sea ice from the Kara Sea for different years.

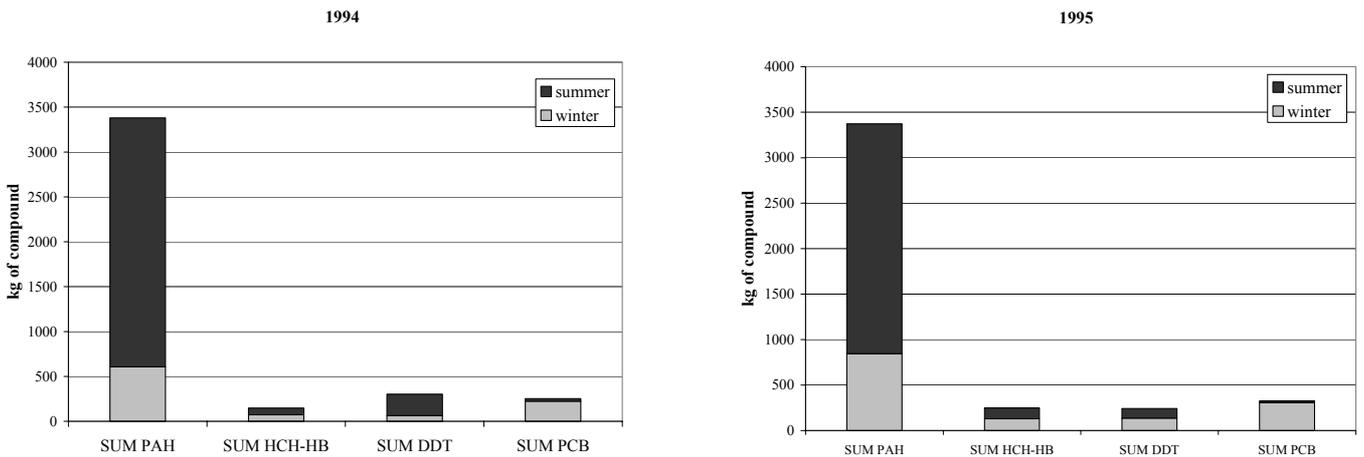


Figure 2: Seasonal fluxes of contaminants from the Pechora River in 1994 and 1995.

# Fluxes and Biogeochemical Processes for Contaminants in Northern Areas

JoLynn Carroll, Ph.D.

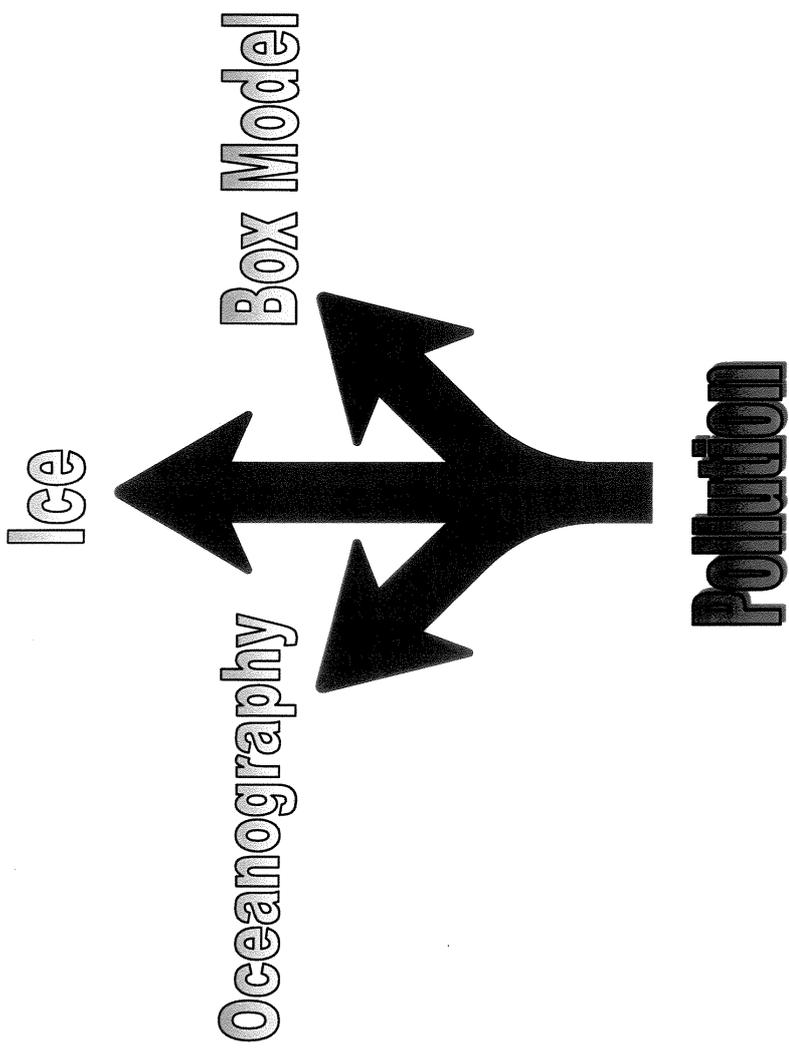
**Selected** results of the Norwegian Ministry of  
Environment and Ministry of Foreign Affairs  
Programme, “Transport and Fate of  
Contaminants in the Northern Seas.”

Akvaplan·niva  

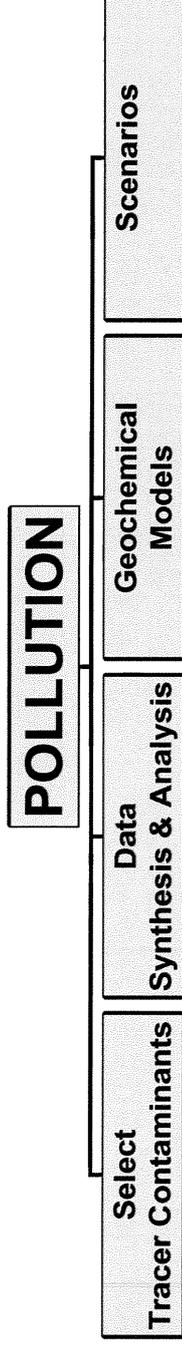

*AMAP workshop on sources, emissions and discharges, Oslo, 23-24 Aug 2001*

## Presentation Outline

- **I. Programme Overview**
- **II. Project Highlights**
  - Transport of contaminants in sediment-laden ice
  - Transport of contaminants through Russian rivers
- **III. The Next Phase**



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*Gather and synthesise information that will support the development of better models to predict the spread of contaminants in Arctic Seas.*

## Data Synthesis/Analysis

### *Historical data from the Russian Arctic*

- Contaminants in Ice
- Contaminants in Snow
- Contaminants in Water

Russian Federal Service for Hydrometeorology  
and Environmental Monitoring

- Regional Center for Monitoring of the Arctic

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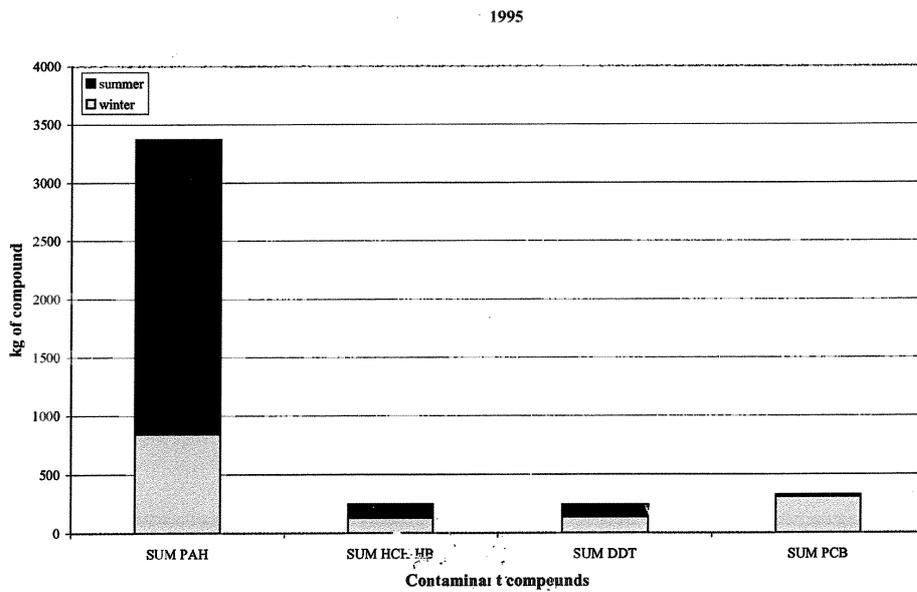
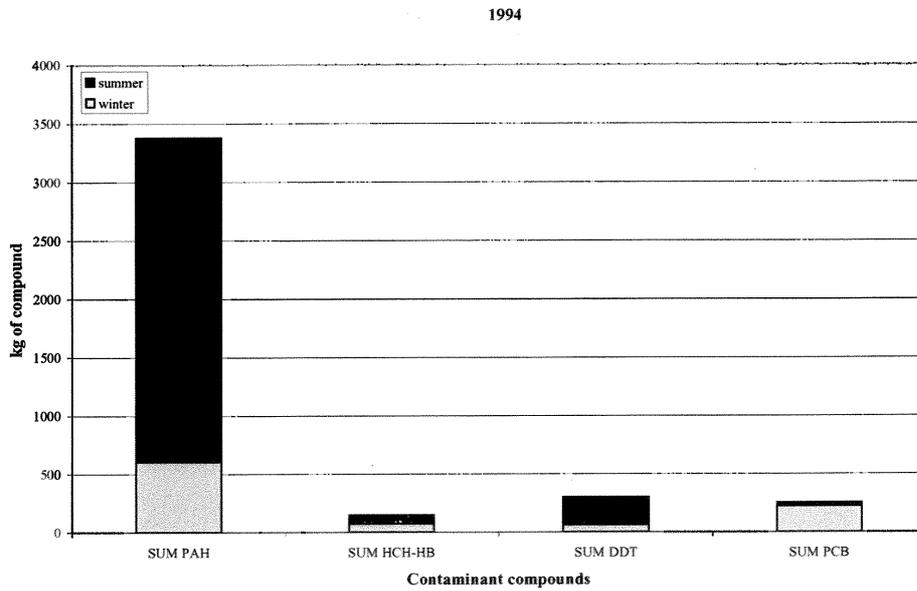

Table 1: Summary description of chemical compounds investigated in this project. Abbreviations used throughout this document conform to the conventions used in the RCMA report (Appendix II).

| Abbreviation                            | Chemical compound / Index | Abbreviation | Chemical compound / Index  |
|---|---------------------------|--------------|--|
| pH                                      | Hydrogen index            | OH           | Total oil hydrocarbons   |
| <b>ORGANOCHLORINES</b>                  |                           |              |  |
| QCB                                     | Pentachlorobenzene        | TNOC         | Trans-nonachlor  |
| HCHA                                    | $\alpha$ -HCH             | DDEPP        | p,p-DDE  |
| HCB                                     | Hexachlorobenzene         | TDEOP        | o,p-DDD  |
| HCHB                                    | $\beta$ -HCH              | TDEPP        | p,p-DDD  |
| HCHG                                    | $\gamma$ -HCH             | CNOC         | Cis-nonachlor  |
| HEPC                                    | Heptachlor                | DDTOP        | o,p-DDT  |
| ALD                                     | Aldrin                    | DDTPP        | p,p-DDT  |
| OCC                                     | Octachlorostyrene         | FMIR         | Fotomirex  |
| HCEPX                                   | Heptachlorepoxyde         | MIR          | Mirex  |
| TCDAN                                   | Trans-chlordane           | #28 ... #180 | PCB congeners #28, #52, #101, #105, #118, #138, #153, #156 and #180        |
| DDEOP                                   | o,p-DDE                   | SUMPCB       | sum of PCB congeners #28, #52, #101, #105, #118, #138, #153, #156 and #180 |
| CCDAN                                   | Cis-chlordane             |              |  |
| <b>POLYCYCLIC AROMATIC HYDROCARBONS</b> |                           |              |  |
| NAP                                     | Naphthalene               | BAA          | Benzo(a)anthracene   |
| ACNLE                                   | Acenaphthylene            | CHR          | Chrysene   |
| FLE                                     | Fluorene                  | BBF          | Benzo(b)fluoranthene   |
| ACNE                                    | Acenaphthene              | BKF          | Benzo(k)fluoranthene   |
| PA                                      | Phenanthrene              | BAP          | Benzo(a)pyrene   |
| ANT                                     | Anthracene                | DBAHA        | Dibenzo(a,h)-anthracene  |
| FLU                                     | Fluoranthene              | ICDP         | Indeno(1,2,3-cd)-pyrene  |
| PYR                                     | Pyrene                    | BGHIP        | Benzo(g,h,i)perylene   |
| <b>HEAVY METALS</b>                     |                           |              |  |
| FE                                      | Iron                      | NI           | Nickel   |
| MN                                      | Manganese                 | CO           | Cobalt   |
| ZN                                      | Zinc                      | PB           | Lead   |
| CU                                      | Copper                    | CD           | Cadmium  |

## Results

- **Contaminants in the Pechora Bay and adjacent areas**
- **Hydrological information for the Pechora, and Ob-Yenisey river systems**
- **Contaminants in snow and ice for the Ob-Yenisey Bays**
- **Snow and ice distributions in the Ob-Yenisey system**

Figure 6: Total Discharges of compounds 1994 and 1995.



## Geochemical Models

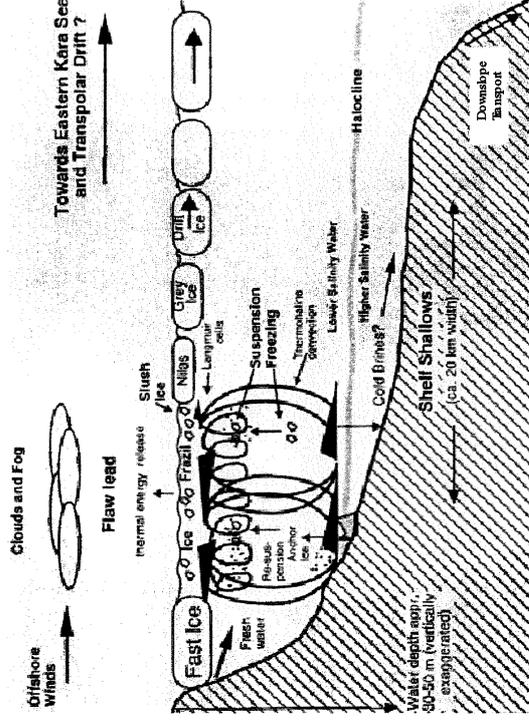
*Recommend mathematical formulations for  
key processes*

- Sea-surface Processes
- Sea-floor Processes

Institute for Air Research  
Agricultural University  
Radiation Protection Authority  
Institute of Marine Research  
Polar Institute

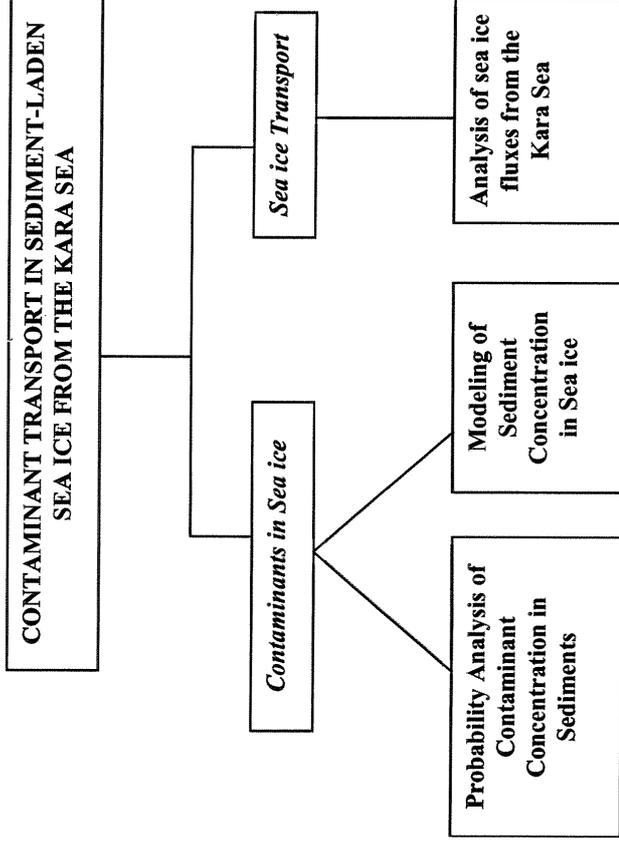
**Akvaplan·niva**  


# Flaw Lead Processes



Possible hydrodynamic processes of turbid ice formation over Arctic shelves (after Dethleff et al., 2000).



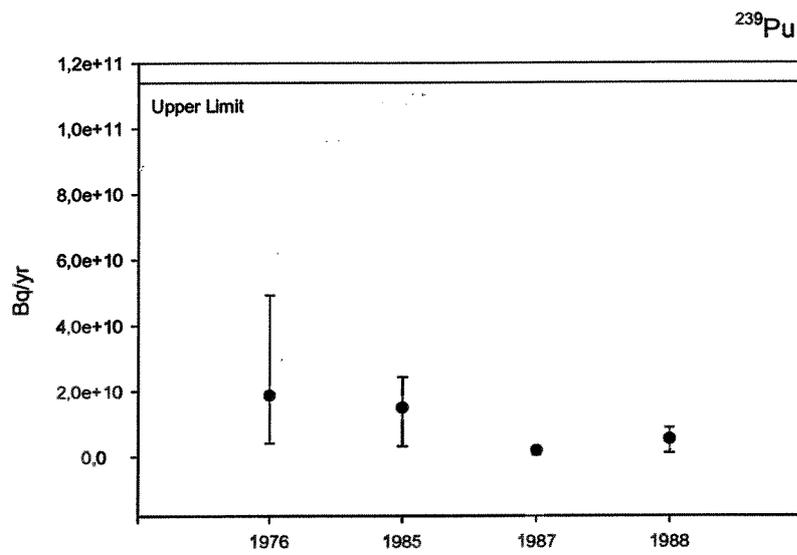
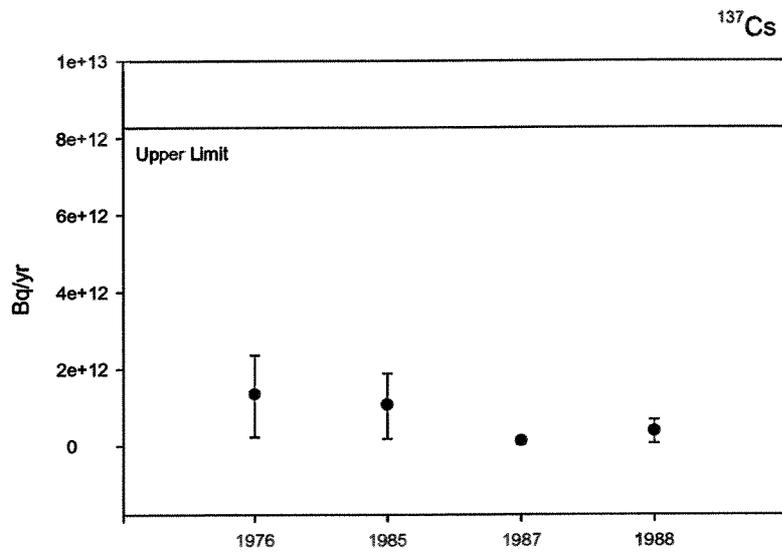


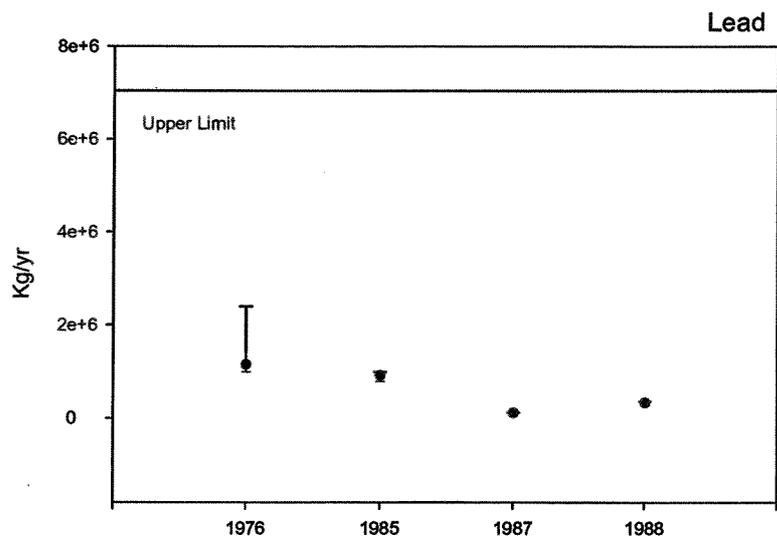
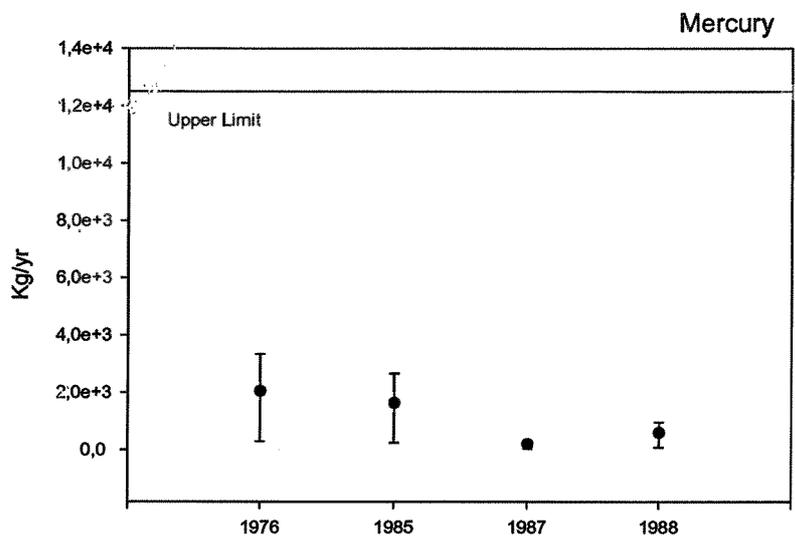
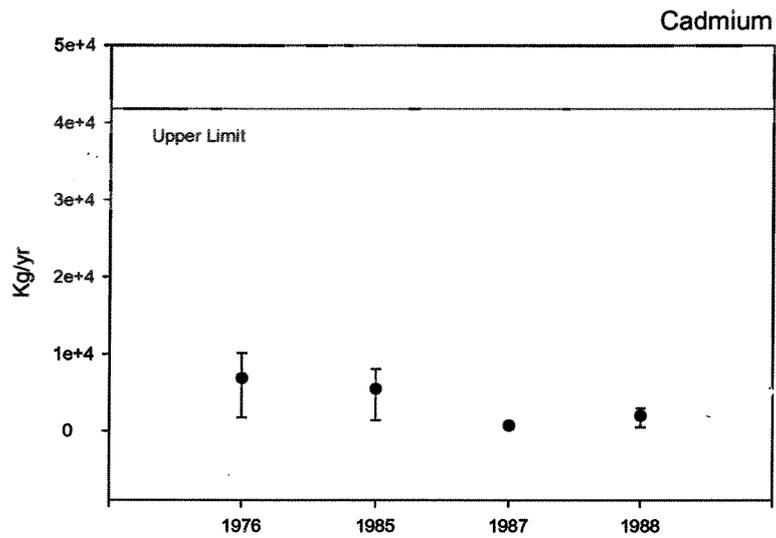
Components used in this analysis of contaminant transport via sediment-laden sea ice from the Kara Sea to the Eurasian Basin.



## Mean Monthly Fluxes of Sediment In Sea Ice (millions of tons/year)

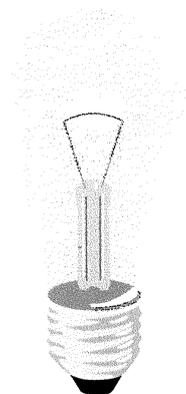
|              | 1985      | 1987       | 1988      | 1976      | Worst<br>Case<br>Scenario |
|--------------|-----------|------------|-----------|-----------|---------------------------|
| October      | 4,3       | 4,3        | 0,1       | 0,1       | 17                        |
| November     | 22        | 0,1        | 0,1       | 0,1       | 30                        |
| December     | 9,8       | 0,4        | 9,8       | 56        | 94                        |
| January      | 0,4       | 0,4        | 0,4       | 0,4       | 104                       |
| February     | 6,0       | 0,2        | 6,0       | 0,2       | 52                        |
| March        | 3,2       | 0,1        | 0,1       | 0,1       | 28                        |
| April        | 0,1       | 0,1        | 0,1       | 0,1       | 19                        |
| May          | 0,1       | 0,1        | 0,1       | 0,1       | 11                        |
| <b>Total</b> | <b>46</b> | <b>5,8</b> | <b>16</b> | <b>58</b> | <b>355</b>                |





## PHASE 2

- Where should monitoring be carried out?
- What sample types should be collected?



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## Phase 2

- Identify key areas in the Kara and Barents Seas where priority contaminants accumulate in the environment.
- Identify key habitat areas for the polar bear, Atlantic cod, and glaucous gull.



*Areas where contaminant pathways intersect with the primary habitat areas of the selected animals.*

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6.2.7 *S. Venkatesh*

**The  $\beta$ -HCH Story  
Global Emissions, Pathways to the Arctic and  
Other Issues**

**Yi-Fan Li and S. Venkatesh  
Air Quality Research Branch  
Meteorological Service of Canada  
Environment Canada  
4905 Dufferin Street  
Downsview, ON.M3H 5T4  
Canada**

**August 2001**

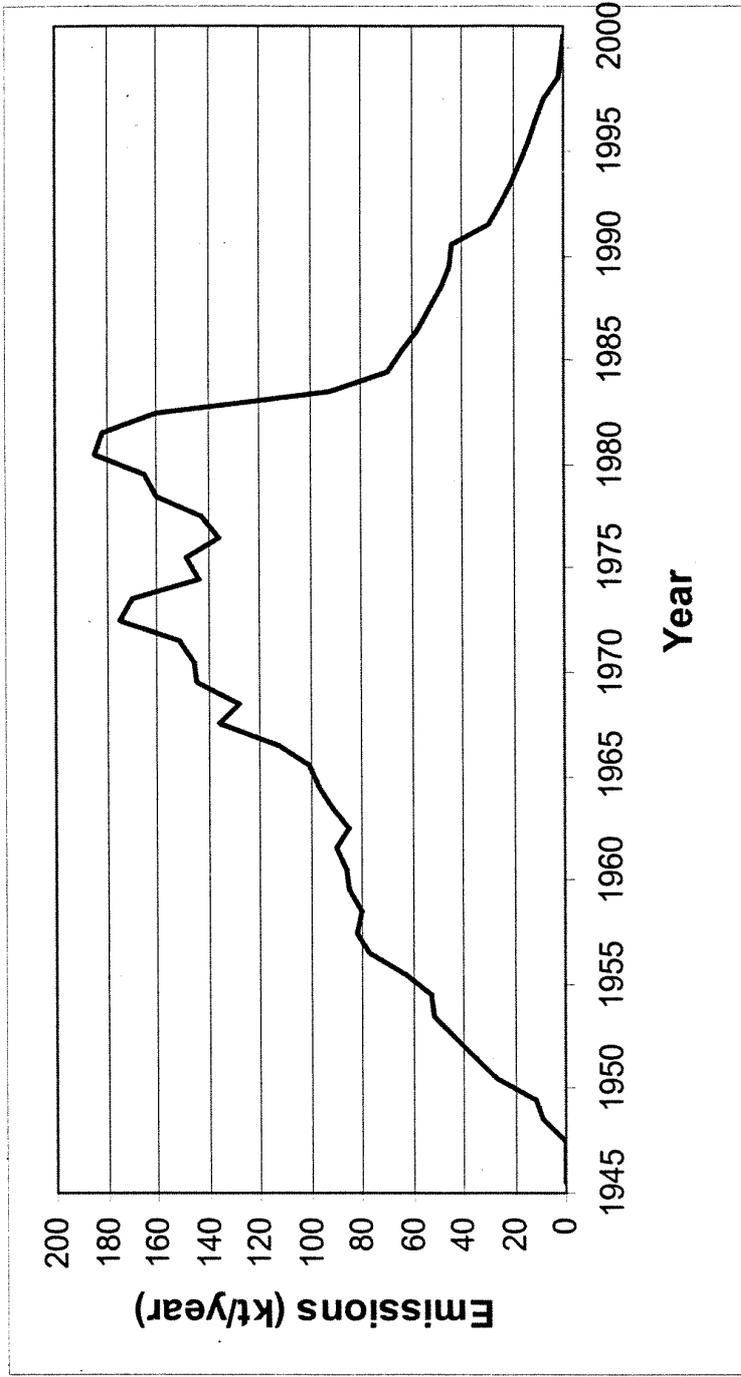


Figure 1. Annual global emissions of  $\alpha$ -HCH between 1945 and 2000

## **Introduction**

**$\alpha$ - and  $\beta$ -HCH in Technical HCH**

**$\gamma$ -HCH in Tech HCH and Lindane**

**$\alpha$ - and  $\gamma$ -HCH isomers in high Arctic due to LRT**

**$\beta$ -HCH conc. in Arctic atm is very low**

**$\beta$ -HCH conc. in Arctic surface water is high ( $\sim 240$  pg/L) and comparable to that of  $\gamma$ -HCH**

## Objectives

1. **To create global gridded  $\beta$ -HCH emission inventories;**
2. **To study the spatial and temporal trends of  $\beta$ -HCH emission.**
3. **To study the sources, pathways of  $\beta$ -HCH in the Arctic.**

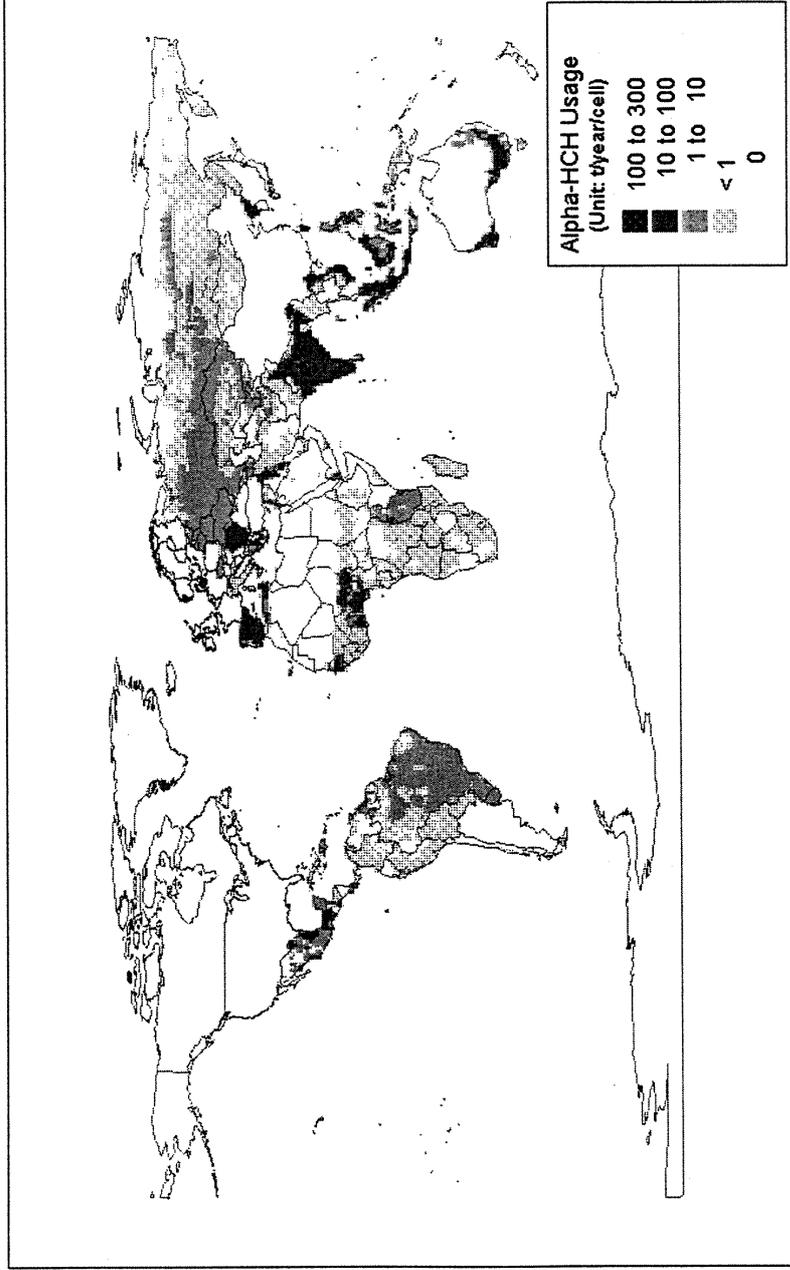


Figure 2. Gridded global technical  $\alpha$ -HCH usage with 1° by 1° longitude/latitude resolution in 1990 (Li, 1999; Li et al., 2001).

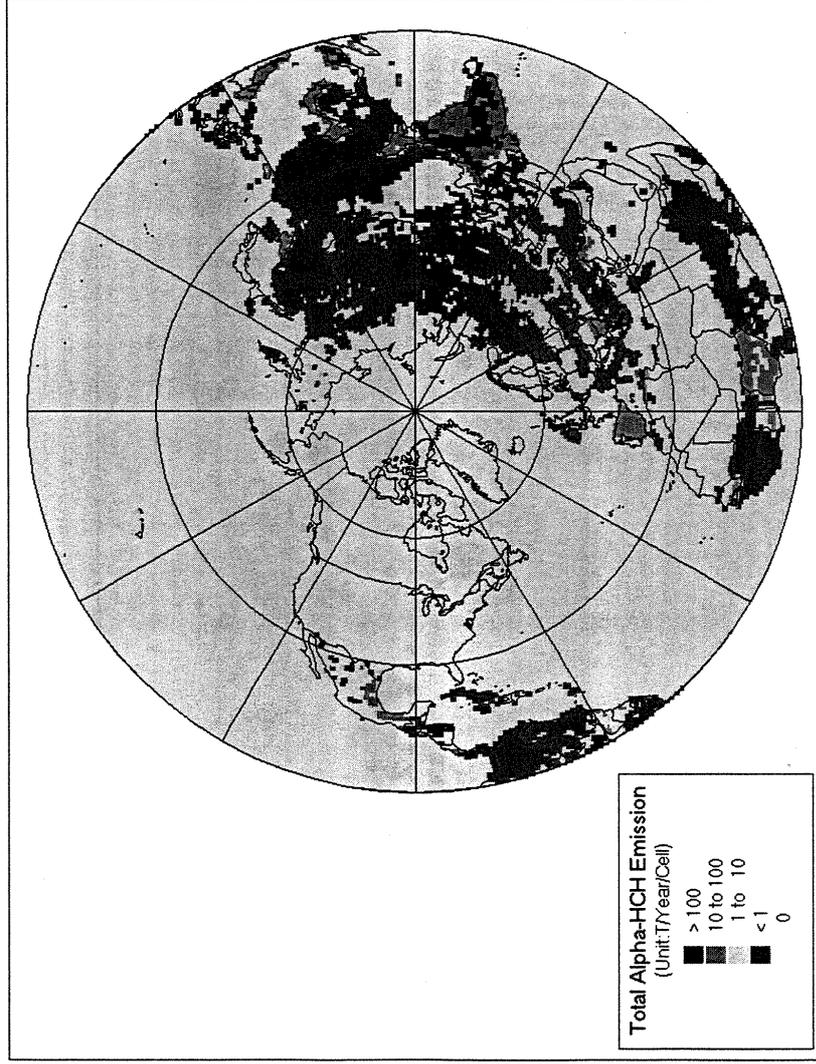
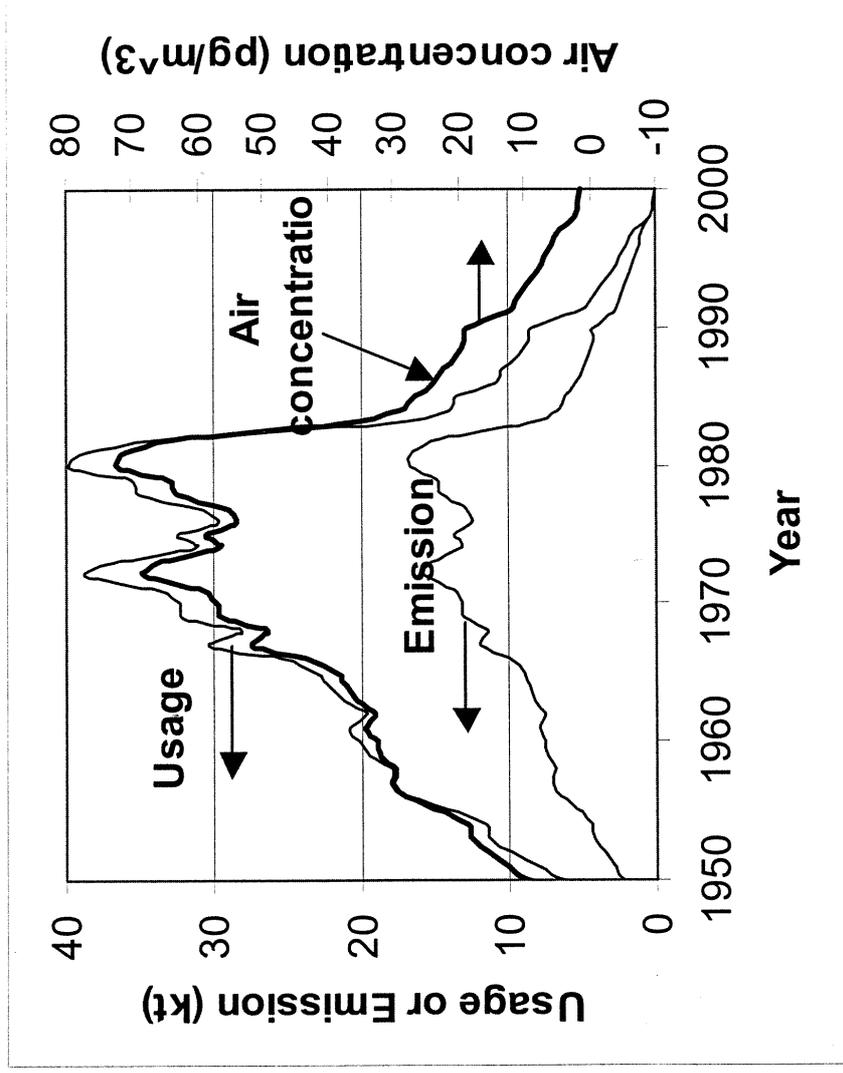


Figure 3. Gridded global  $\alpha$ -HCH emission with  $1^\circ$  by  $1^\circ$  longitude/latitude resolution in 1990 (Li et al. 2000).



**Figure 4.** Global usage and emissions of  $\beta$ -HCH from 1950 to 2000. The estimated air concentrations of  $\beta$ -HCH in the Arctic calculated by Equation (3) are also shown.

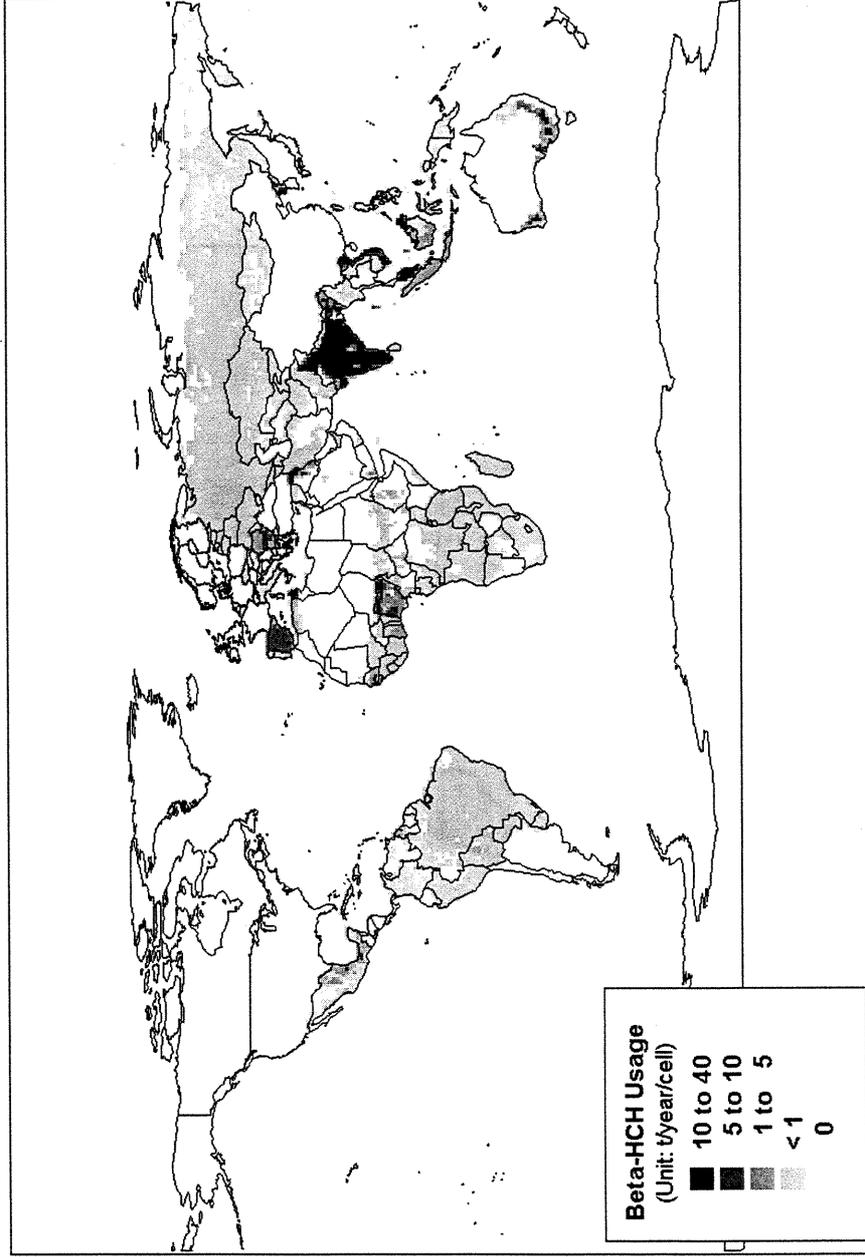


Figure 5. Global gridded  $\beta$ -HCH usage data on a  $1^\circ \times 1^\circ$  longitude and latitude grid system for 1990. The total usage in this year was 8.4 kt.

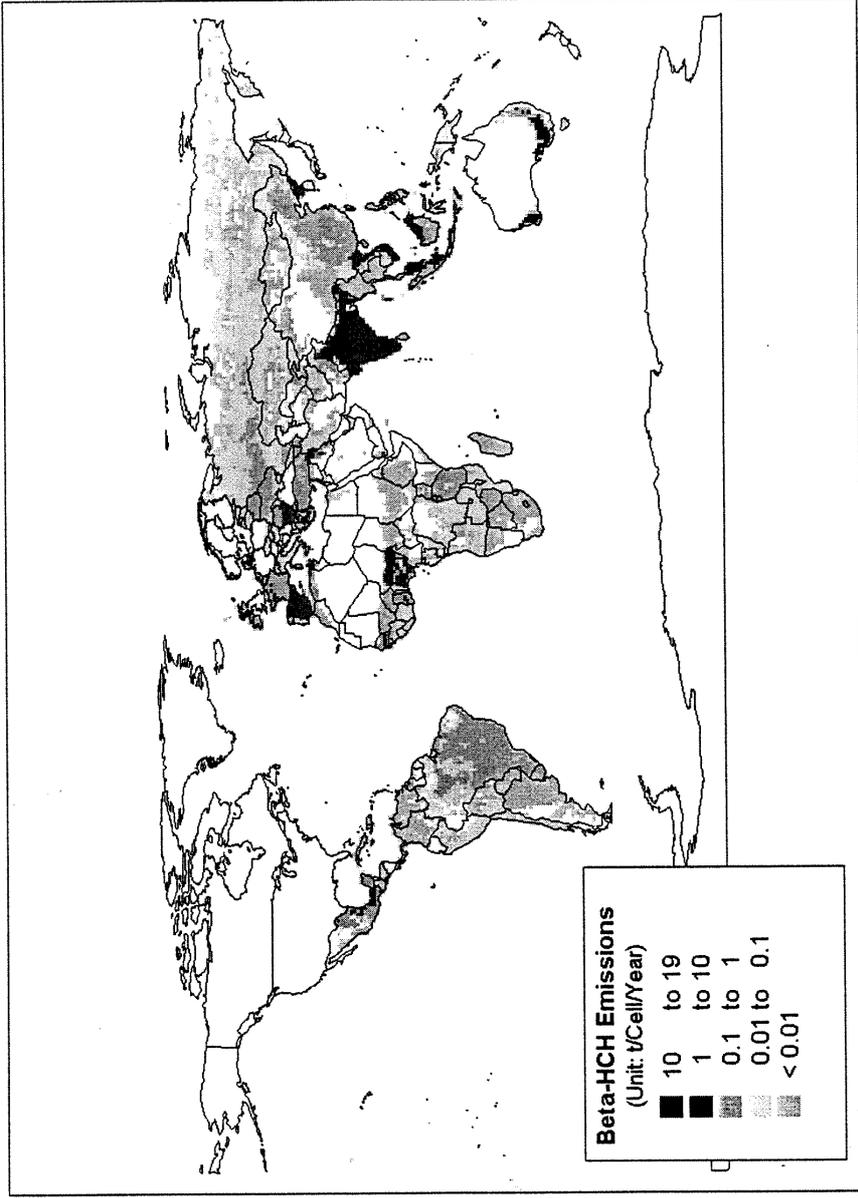
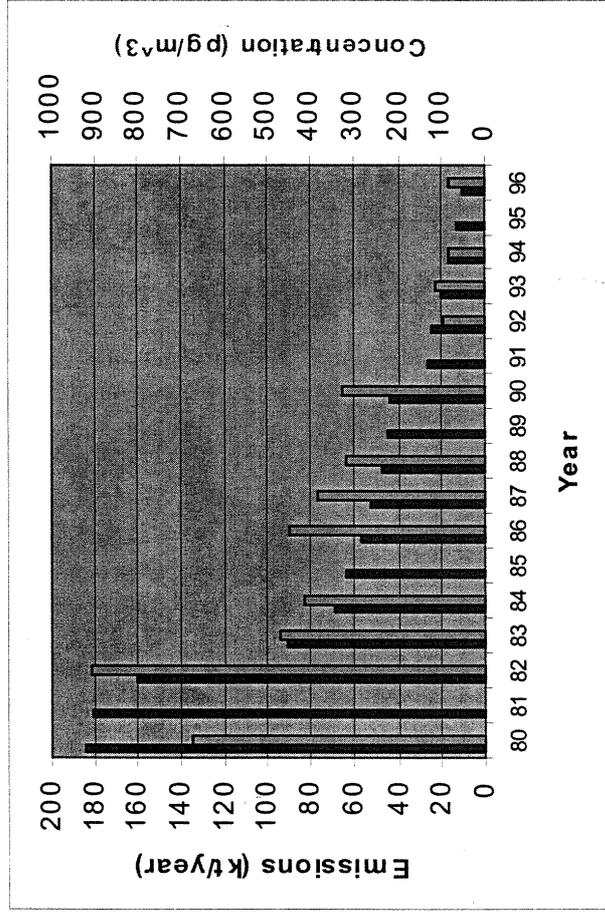
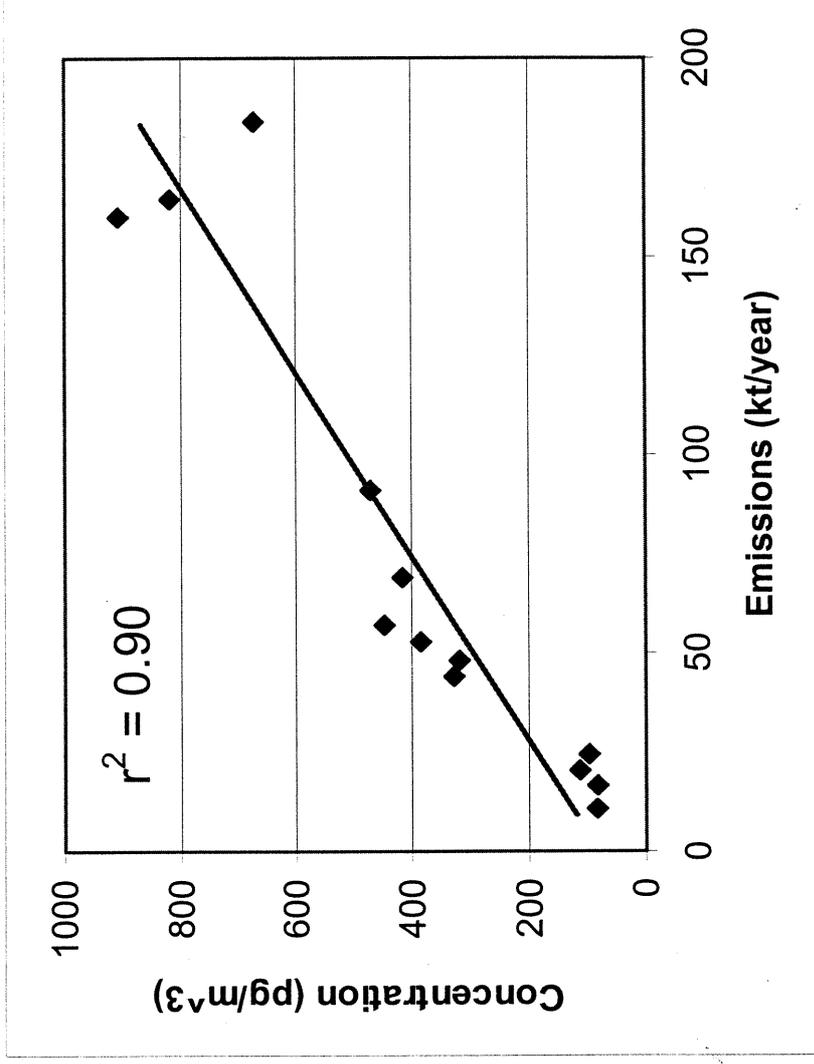


Figure 6. Global gridded  $\beta$ -HCH emissions on a  $1^\circ \times 1^\circ$  longitude and latitude grid system for 1990

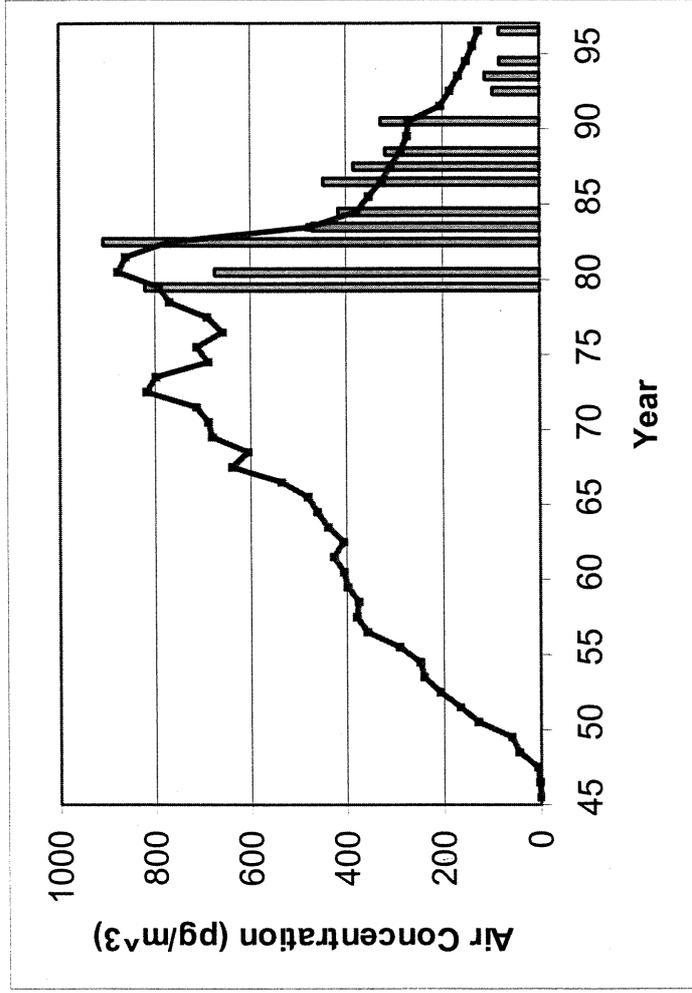
## Global $\alpha$ -HCH emission and mean air concentrations of $\alpha$ -HCH in the arctic from 1980 to 1996



Li, 2000, Correlation between global emissions of  $\alpha$ -hexachlorocyclohexane and its Concentrations in the Arctic Air, *Geophysical Research Letters*, Submitted.



**Figure 8.** Correlation between global emissions of  $\alpha$ -HCH and its concentrations in Arctic air with  $r^2 = 0.90$ . The straight least-squares line is given by Equation (1).



**Figure 9.** The concentrations of  $\alpha$ -HCH in Arctic air calculated from the annual global emissions using Equation (1) for the years between 1980 and 1996 and Equation (4) for the years between 1945 and 1980 (curve). Measurement data (bars) in the Arctic atmosphere are also presented for comparison.

# Pathways for $\beta$ -HCH

Data for  $\beta$ -HCH sparse

Can produce  $\beta$ -HCH using similar methods as  $\alpha$ -HCH

**Non-detectability of  $\beta$ -HCH in arctic air, indicates LRT probably not a significant factor**

**Hence estimated emission probably not correct**

**Hypothesis:  $\beta$ -HCH in arctic air and seawater has been close to equilibrium since early 1950s**

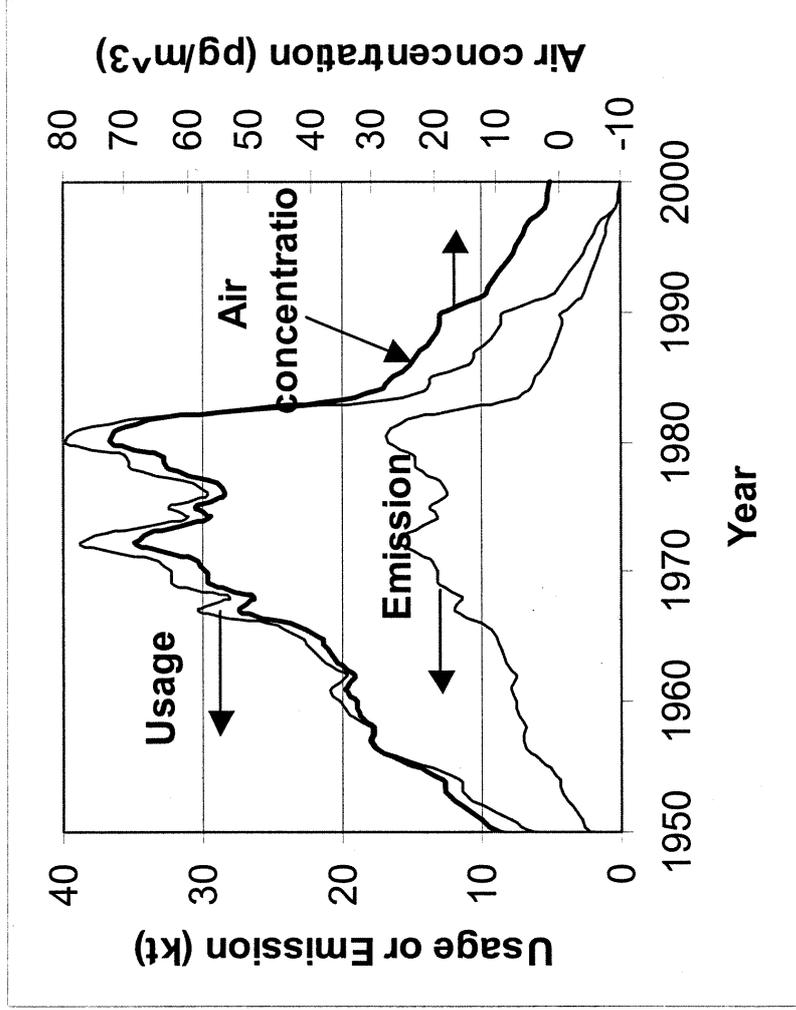
**$\beta$ -HCH conc in Arctic seawater 0.25 ng/l  
(Jantunen and Bidleman, 1995, 1998)**

**$\beta$ -HCH conc in Arctic air 0.57 pg/m<sup>3</sup>  
(Halsall et al., 1998)**

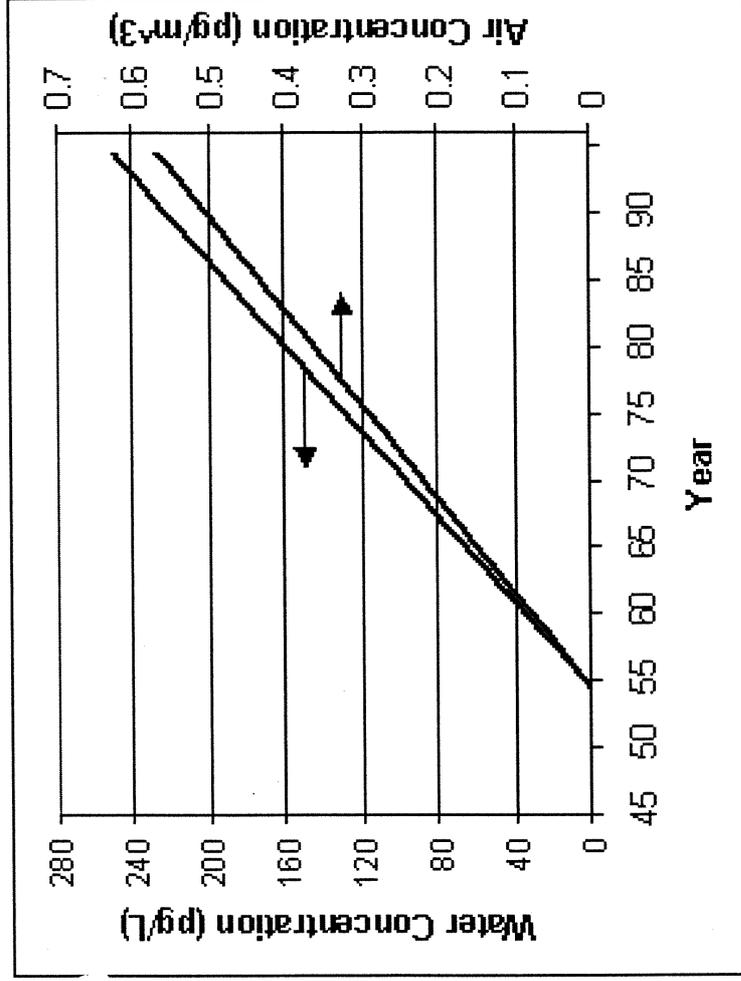
**Very small Henry's law constant**

**Favours partitioning into water and soil**

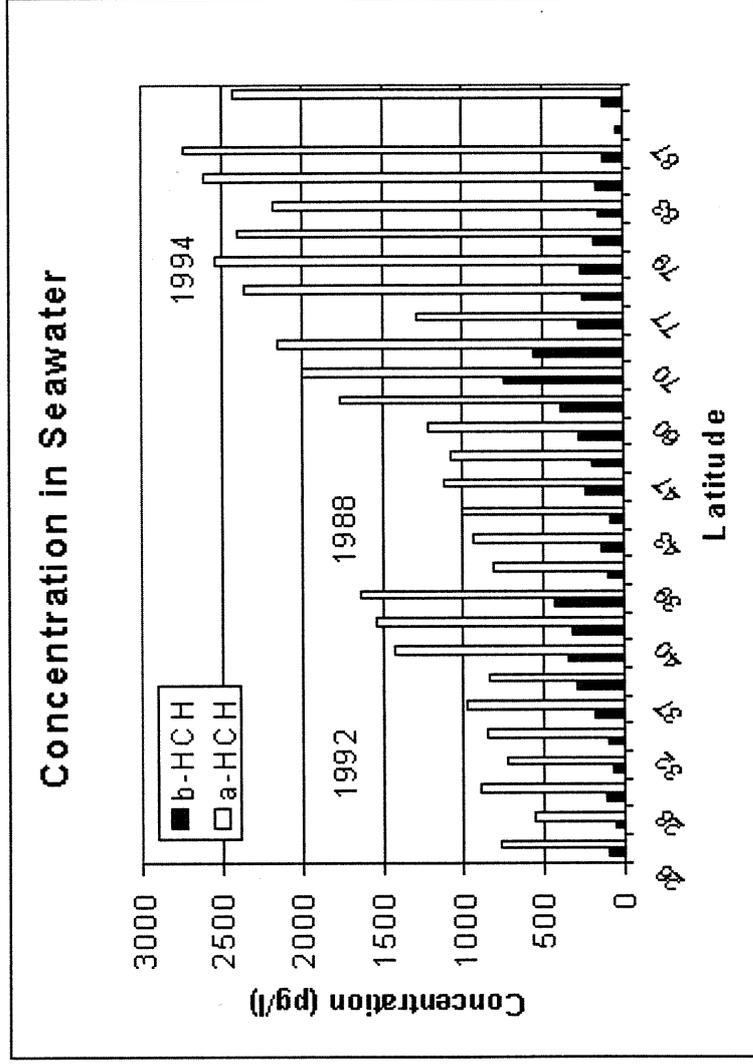
**Ocean current becomes main pathway**



**Figure 10.** Global usage and emissions of  $\beta$ -HCH from 1950 to 2000. The estimated air concentrations of  $\beta$ -HCH in the Arctic calculated by Equation (3) are also shown.



**Figure 11.** Calculated concentration of  $\beta$ -HCH in air and seawater in the Arctic.



**Figure 12.** Concentrations of  $\beta$ -HCH and  $\alpha$ -HCH in seawater as functions of latitude [Data for 1988 and 1992, from Chernyak et al. 1995; Data for 1994 from Jantunen et al. 1999, Li et al., 2001].

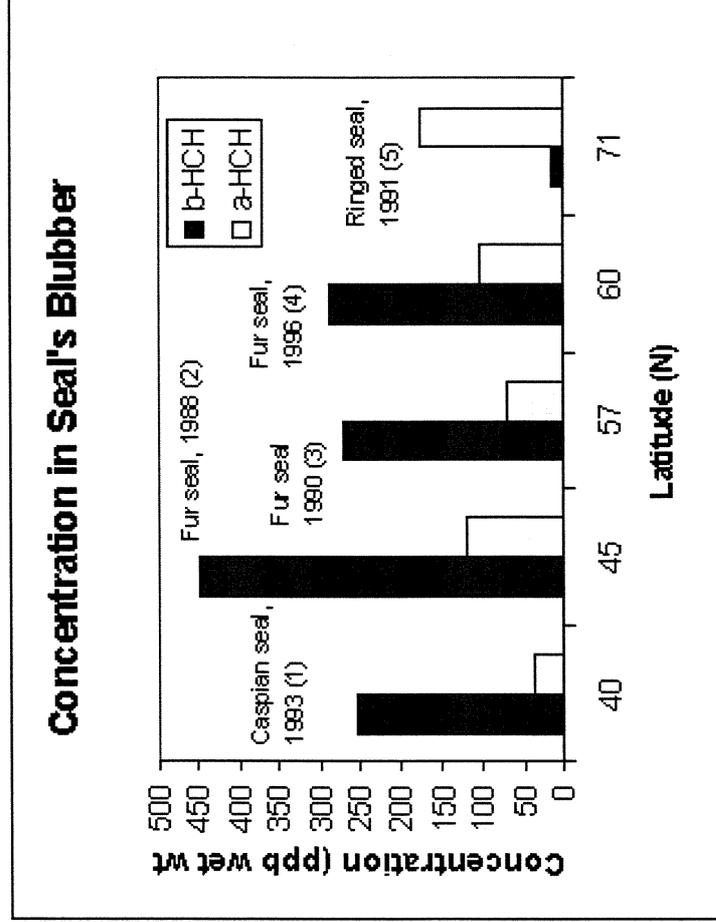
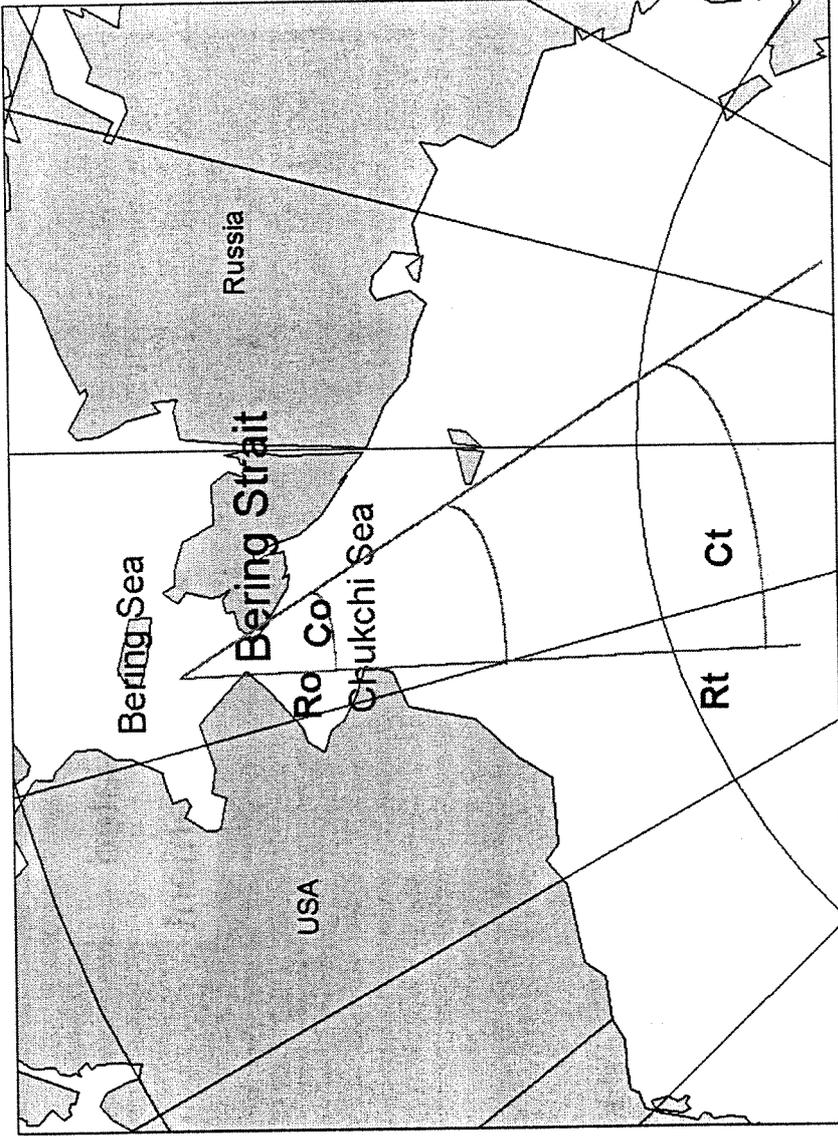


Figure 13. Concentrations of  $\alpha$ - and  $\beta$ -HCH in blubbers of different seals in the Pacific Ocean and Arctic Ocean. References: (1) *Vetter et al.*, 1995; (2) *Tanabe et al.*, 1994; (3) *Mossner et al.*, 1992; (4) *Mossner and Ballschmiter*, 1997; (5) *Addison*, unpublished data.



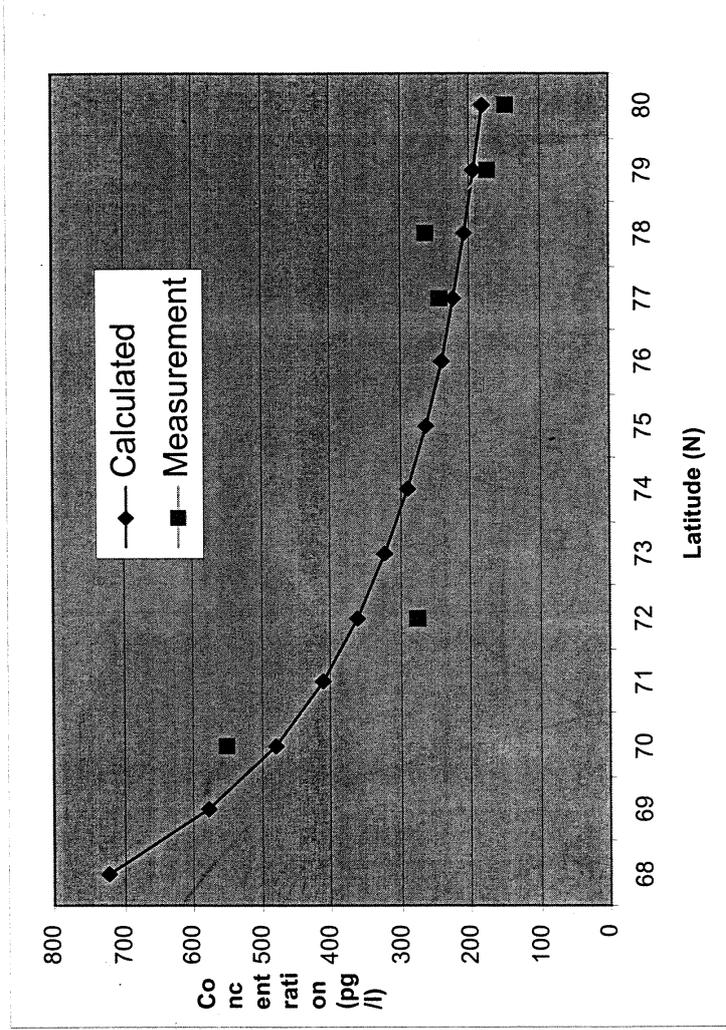


Figure 15. Comparison of the constant flux model (Eq. 5) calculation and the  $\beta$ -HCH concentration measurements in seawater north of the Bering Strait (Jantunen *et al.*, 1999).

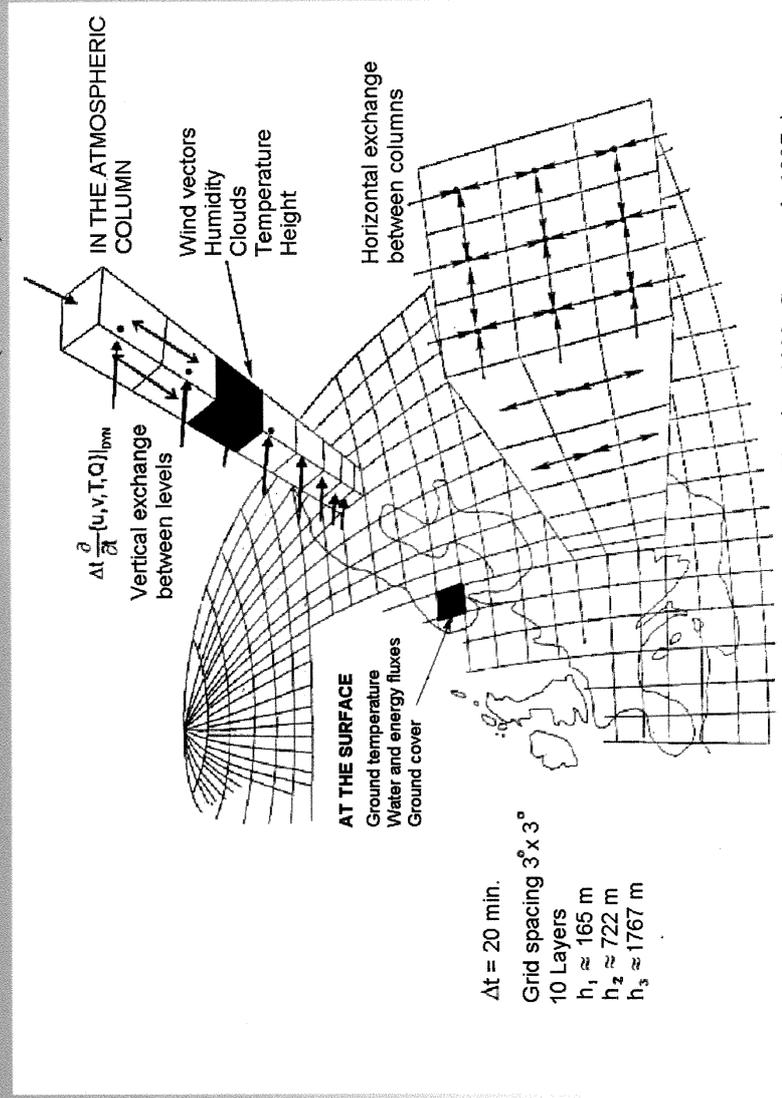
## Summary and Conclusions

1. Strong correlation between  $\alpha$ -HCH conc in Arctic air and global emissions
2.  $\alpha$ -HCH conc in Arctic air mainly due to atmospheric LRT
3.  $\beta$ -HCH hardly detected in Arctic air even during 1980 when technical HCH was used in large amounts
4.  $\beta$ -HCH conc in Arctic did no respond to global  $\beta$ -HCH emissions leading to hypothesis " $\beta$ -HCH in arctic air and seawater has been close to equilibrium since early 1950s"
5. Ocean current is main pathway for  $\beta$ -HCH to enter the Arctic

6. Bering Strait is a bottleneck for  $\beta$ -HCH to enter Arctic
7.  $\beta$ -HCH conc in seawater decreases from Bering Strait to North pole while it is the opposite for  $\alpha$ -HCH - due to cold condensation effect
8. Entry of  $\beta$ -HCH into Arctic seawater is a slow process and concentrations in water and seal blubber are much lower than those for Pacific waters
9.  $\beta$ -HCH in Arctic air is through volatilization from water, and not from atmospheric LRT. Hence no direct correlation with global usage of Tech HCH
10. With very small HLC  $\beta$ -HCH in Arctic air is often below detection limits

# LCM - Local Climate Model (FIZ-C)

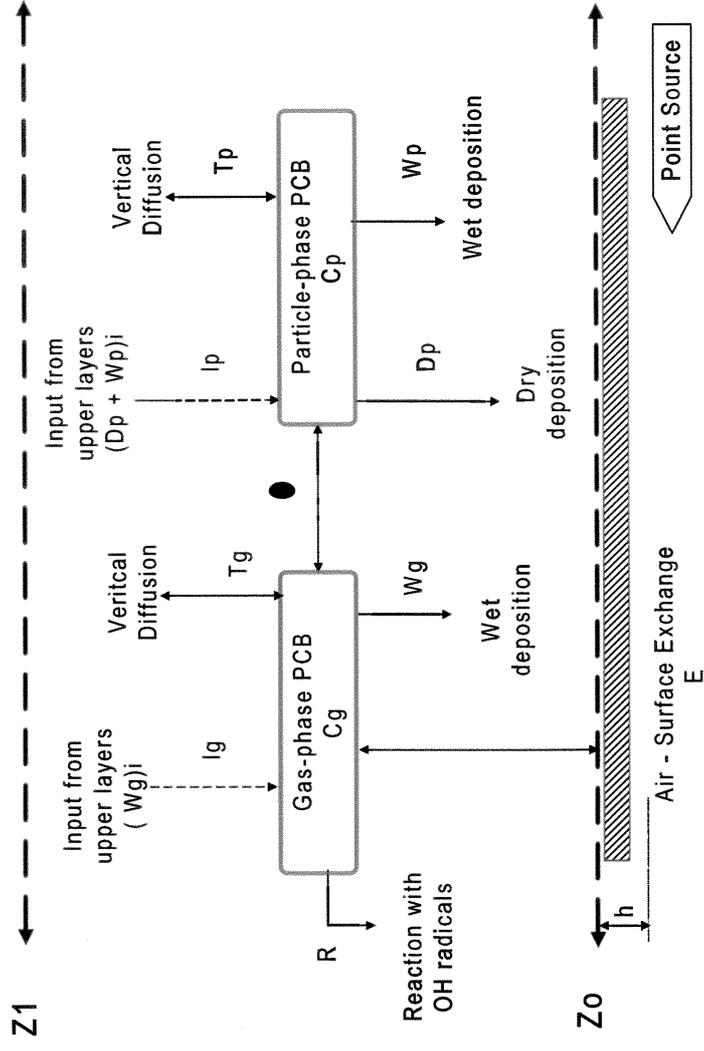
NARCM

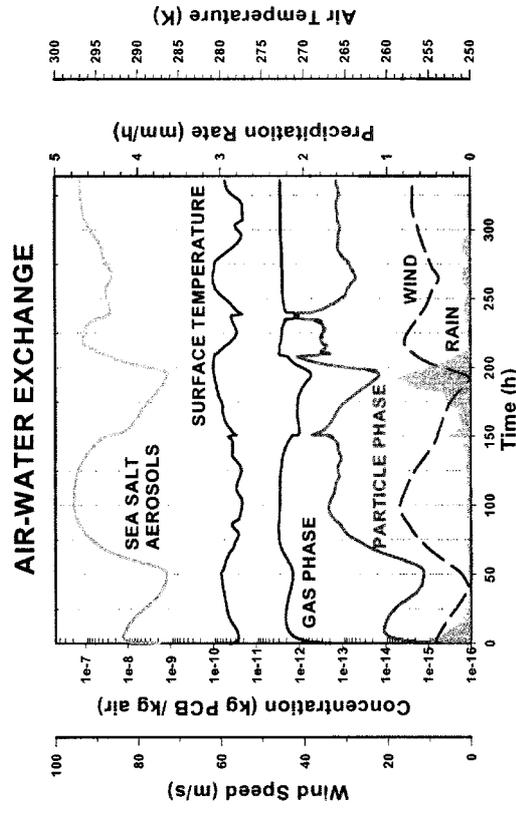


(Therrien, 1993; Gong *et al.*, 1997a)



# Sources and sinks of gas and particle phase PCB's at the surface layer in 1-Dimensional Air-Water Column





Time series of modeled parameters for particle and gas phase concentrations of PCB-18 in the surface layer (0-167m). The model runs in May for 15 days over North Atlantic Ocean, west of Ireland (Sahuvar, 1999).

### 6.2.8 Benoit Fribourg-Blanc

## EEA AND EMISSIONS TO WATER : FIRST STEPS TOWARDS EUROPEAN REPORTING AND ASSESSMENT TOOLS

AMAP WORKSHOP on Sources, Emissions and Discharges

Oslo, Norway

23 – 24 August 2001

This document presents briefly the main elements of the work undertaken by the European Topic Centre for Water of the European Environment Agency relevant to the workshop.

It presents firstly the framework that is to say the EEA, the role of the ETC and DPSIR framework. Then the main elements of the workprogram are presented that are indicators in support of policy and development of Eurowaternet. Finally the development of Eurowaternet-Emissions (pressures), and work done so far and work to be done in the future are detailed.

### The European Environment Agency

The European Environment Agency (EEA) was launched by the EU in 1993 to implement an information and measurement network in order to harmonise European initiatives. This organisation constitutes the head of the European environmental structure.

The EEA's Mission Statement is as follows:

*“The EEA aims to support sustainable development and to help achieve significant and measurable improvement in Europe's environment through the provision of timely, targeted, relevant and reliable information to policy making agents and the public.”* (From: *European Environment Agency – [On Line] – Available from Internet: <URL: <http://org.eea.eu.int>>*).

This information must enable Member States to take the requisite measures to protect their environment, to assess the result of such measures and to insure that the public is properly informed about the state of the environment.

More precisely, EEA's main objectives are to :

- Implement and co-ordinate the EIONET with EEA Member States
- Collect information on state of the environment with the use of EIONET.
- Provide Community institutions and EEA Member States with information allowing the development and implementation of sound and efficient regulations.
- Develop and regularly publish reports on state and evolution of the environment in Europe, including reports on environmental indicators.
- Ensure large diffusion and acceptability of environmental information.
- Work with EEA Member States, European institutions and other organisations to avoid any redundancy.

In terms of water, the EEA requires information on the state of Europe inland waters (availability, demands, water stress), time trends and pressures on the environment

arising from human activities, industry, agriculture. This would enable identification of what is causing the problems and an assessment of the state of action on policies.

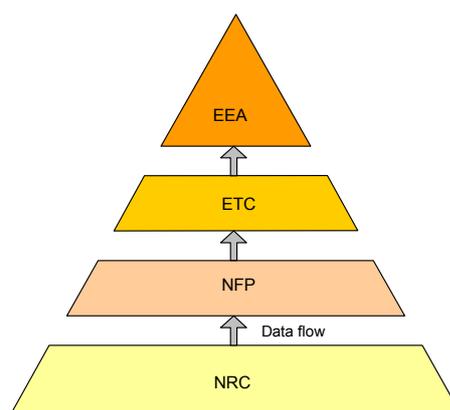
The Agency is the hub of the European Environment Information and Observation Network (EIONET), a network of some 600 environmental bodies and institutes across Europe.

EIONET helps the EEA to retrieve data to identify special issues and to produce efficient and timely information on environment in Europe.

EIONET is made up of :

- five European Topic Centres (ETCs), in charge of the collection, analysis, assessment and synthesis of information on environment in Europe. Each ETC deals with a precise subject. They cover **Waters, Terrestrial Environment, Air and Climate Change, Nature Protection and Biodiversity and Waste and Material Flows**. Each centre is a consortium of European specialist partners organisations, with one leading institution.
- National Focal Points (NFPs), typically ministries or national environment agencies, responsible for national co-ordination of activities related to EEA work programme.
- National Reference Centres (NRCs), in charge of providing the EEA with national information on different themes. Generally, NRCs are national experts on a precise subject (e.g. Cemagref or INRA in France).
- Main Component Elements (MCEs), collectors, interpreters and suppliers of environmental data, with expertise in environmental science, monitoring or modelling. About a quarter of these are NRCs.

Information originally produced by the numerous NRCs is collected by NFPs and aggregated by ETCs. Finally, after peer review, it is transmitted to the EEA for publication. Each country is responsible for organising the relationship between NFPs, NRCs and MCEs, the following structure being established :



*EIONET structure*

The geographical scope of EEA's work is not confined to EU Member States; membership is open to countries sharing the concerns of the EU and the objectives of the Agency.

Membership in 2001 includes all 15 EU Member States<sup>2</sup>, as well as Iceland, Liechtenstein and Norway, Bulgaria, Cyprus, Latvia, Malta, Slovenia and the Slovak Republic. The 6 other Countries of Central and Eastern Europe (CEECs) in accession to the Community<sup>3</sup> and Turkey are expected to join this year.

The Agency uses not only existing capacities in Member States, but also co-operates actively with other bodies and international organisations to build synergy and to avoid duplication of efforts.

Figure 1.1 presents EEA work area.



#### *Work area of the European Environment Agency*

(From: Intercarto – [On-Line] – [Last modified on 10/04/00] – Available from Internet: <URL: <http://www.intercarto.fr>>)

<sup>2</sup> Austria, Belgium, Denmark, Finland, France, Germany, Greece, Ireland, Italy, Luxembourg, The Netherlands, Portugal, Spain, Sweden, United Kingdom.

<sup>3</sup> Czech Republic, Estonia, Hungary, Lithuania, Poland, Romania.

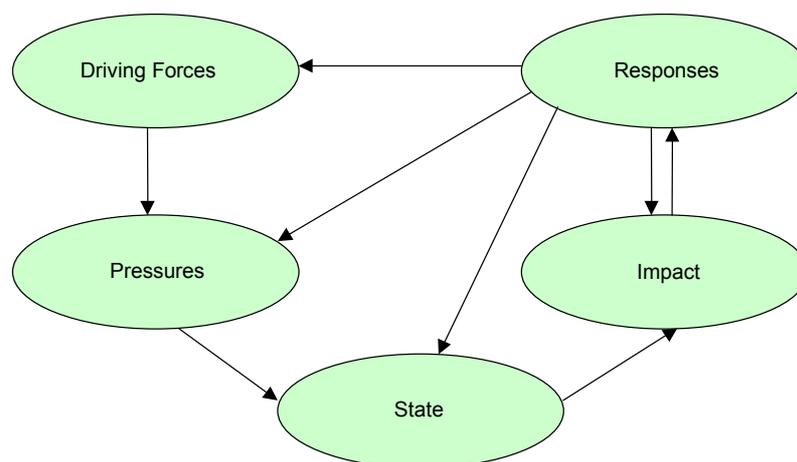
## DPSIR Framework

The EEA has developed a conceptual framework known as the DPSIR assessment framework within which each work must be included. The role of the EEA is to provide information on the DPSIR elements and their interconnections, and on the effectiveness of responses.

DPSIR stands for:

- *Driving forces*, the human or economic activities that cause pressure – e.g. population increase, increased urbanisation.
- *Pressures*, expression of the driving forces – e.g. emissions of harmful substances.
- *State of the environment*, which describes the quality and quantity of natural resources – e.g. nitrogen and phosphorus levels in inland and coastal waters.
- *Impacts*, effects or loss of use experienced in the environment – e.g. loss of amenity in a waterway, decrease in biodiversity.
- *Responses*, various corrective actions undertaken that may affect any of the inputs of the conceptual framework – e.g. EU Directives, taxes, incentives.

Relationships between the components are described in the following Figure:



*The DPSIR framework*

Emissions to water belong to the Pressure category. The State category corresponds, for example, to substance concentrations in a water body, and Driving Forces may be related to agricultural activity for example.

The DPSIR framework is useful in describing the relationships between the origins and consequences of environmental problems, but in order to understand their dynamics it is also useful to focus on the links between DPSIR elements. For instance, the relationship between Driving Forces and Pressures by economic activities is a function of the eco-efficiency of the technology and related system in use.

### The European Topic Centre on Waters (ETC/WTR)

The present European Topic Centre on Waters (ETC/WTR) is the result of the merging of former Topic Centres on Inland Waters and on Marine and Coastal Environment. It's one of the five technical centres of the EEA

The Water Research Centre (WRc) in the United Kingdom is the lead organisation of the ETC/WTR. This consists of a consortium of several European organisations, which each have a representative on a Management Committee which agrees the allocation of tasks and budget following the advice and recommendations of a smaller, Technical Committee.

The Management Committee is chaired by Dr Tim Lack of WRc who also provide the services of a Technical Coordinator (Steve Nixon) who chairs the Technical Committee.

The organisations represented on the Topic Centre are:

- **Water Research Centre (WRc PLC)**
- **Centre for Ecology and Hydrology (CEH) (Formerly the Institute of Hydrology) (In association with WRc)**
- **Vituki Consult Rt.**
- Austrian Working Group on Water (AWW - Austria)
- Centro de Estudios y Experimentacion de Obras Publicas (CEDEX - Spain)
- International Office for Water (IOW - France)
- **IFREMER (In association with IOW)**
- National Environmental Research Institute (NERI - Denmark)
- Geological Survey of Denmark and Greenland (GEUS) **(In association with NERI)**
- Norwegian Institute for Water Research (NIVA - Norway)
- **National Centre for Marine Research (NCMR)**
- **Ente per le Nuove Tecnologie, l'Energia e l'Ambiente (ENEA)**
- **Institute of Meteorology and Water Management (IMGW)**

There are a number of supporting organisations and national experts throughout Europe who also contribute to the Topic Centre's work programme carried on behalf of the Agency.

The Topic Centre work is based on a three years contract and workplan and on an annual workprogramme with many different tasks.

For 2001 the main tasks are :

- to assess existing tools, methodologies and models for use in the integrated assessment of the water environment in relation to policy,
- to develop a core set of indicators for all types of water body, and to produce an indicator based report on water,
- to progressively improve data flows on water at a European level by the implementation of EUROWATERNET and by cooperation with international data holders such as Marine Conventions and Eurostat.

## Indicators in support of policy

Indicators are just a way of packaging information in a simple and straightforward way that helps to deliver clear and meaningful messages to target audiences. However, the development of a multi-purpose core set of indicators is a major challenge the EEA and ETCs face.

A first core set was developed in the transport sector and ongoing exercises are led with respect to energy, agriculture, waste and material flows, and air pollution and climate change.

A similar exercise is ongoing for water (including hazardous substances): the ETC/WTR is currently developing a core set of indicators for all types of water bodies, the ultimate aim being to produce an indicator based report on water, regularly updated and improved. This core set of indicators should answer seven policy questions that are :

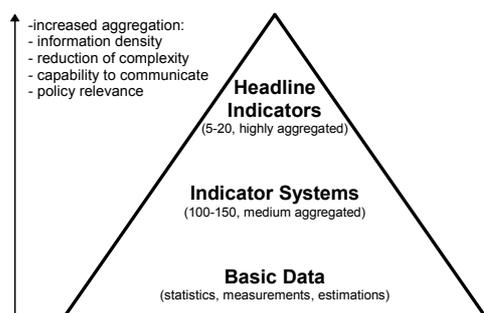
- Are policies reducing pressures on the water environment?
- Is the state of the water environment improving as a result?
- Are ecological and human health standards for water being met?
- Are policies reducing impacts on aquatic ecosystems?
- Is water being used sustainably by economic sectors?
- Is climate change having an impact on water resources?
- Are we paying the full market price for water?
- Do the benefits of water management policies outweigh the costs?

A database in support of the other reports of the EEA has also to be produced.

These indicators should address the multi-purpose needs of EEA, the desire to have a relatively small and well-balanced core set of indicators, and the data availability and quality.

Due to the lack of homogenous or available data at a European scale, they will be developed in a step-by-step process from the present core set based on data already available to the future core set based on data available in the future.

The following figure shows the overall organisation of these indicators :



## Development of Eurowaternet

In order to respond to the statutory questions it has to answer in the water domain, EEA launched as soon as 1994 a process targeted to harmonising information production. For continental running waters, this process was called EUROWATERNET, (founding document EEA Topic report n° 10, 1996) the aim being to allow producing useful and comparable statistics from the existing monitoring networks.

It is defined as following : "EUROWATERNET is the process by which the EEA obtains the information on water resources (quality and quantity) it needs to answer questions raised by its customers."

The **goal** of EuroWaternet is to obtain representative, policy relevant, reliable and timely information on rivers, lakes, reservoirs, groundwater bodies and since 2001 should be extended to transitional and coastal waters. This is done through the progressive implementation of:

- A basic network (selection in existing networks),
- An impact network (specific pressure, substance, policy),
- A "fully representative network",

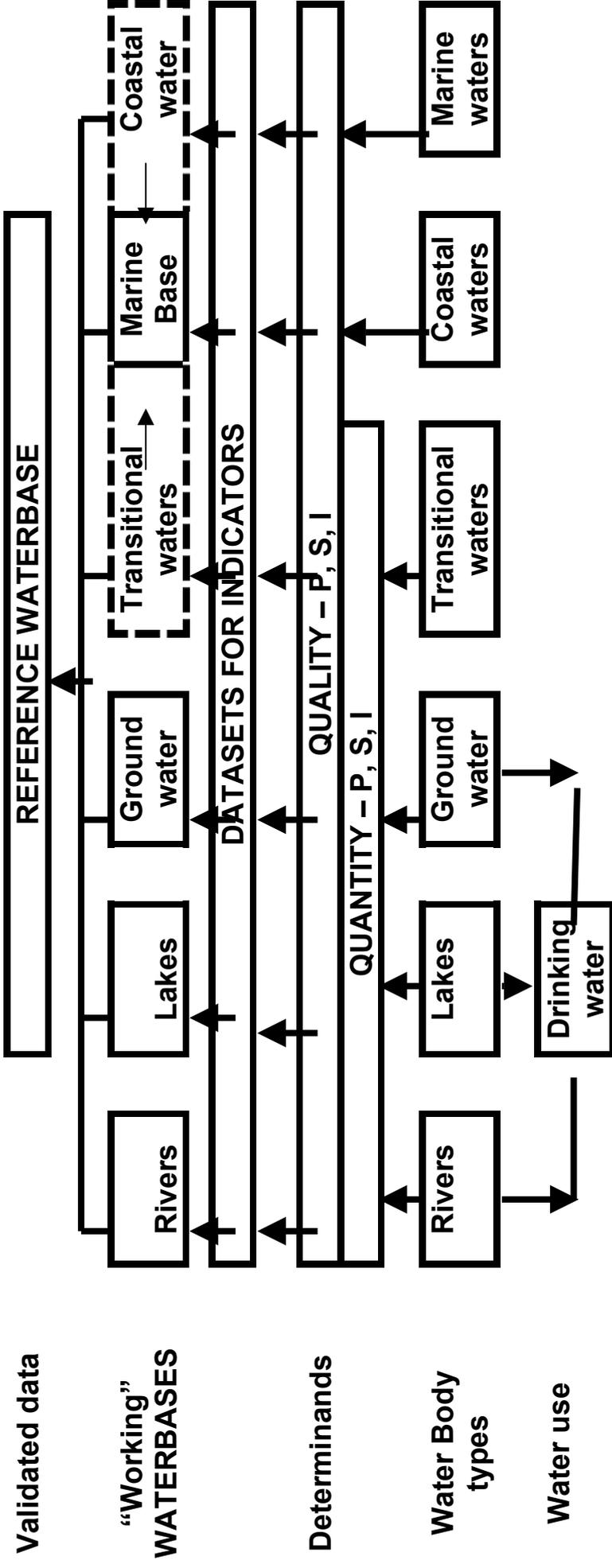
These networks are tailored to respond to both EEA needs and Water Framework Directive demands and to assess effectiveness of EU legislation. To cope with the disparity of existing monitoring systems, the methodology that was designed is based on stratified survey of the existing networks that imply selecting monitoring points from the bulk of the existing points. This builds a European network of samples stations for each water body type.

The data flows associated with EUROWATERNET representative networks are straightforward: member states select representative monitoring points, transfer to EEA (through its ETC) raw data attached to the points. These data are stored in a database called WATERBASE, that is planned to be accessible via the Web in the future. A wide quantity of data with long time series for most of the EEA-Members, and for the basic network, especially for rivers, is already stored. The information collected is used to produce assessment reports for the EEA (e.g. Groundwater Quality and Quantity in Europe, Excessive Nutrients in Europe's Ecosystems, Sustainable Water Use in Europe and State and Pressures of the Marine and Coastal Mediterranean Environment). They also contributed to the relevant chapters of Europe's Environment: Second Assessment and the State of the European Union at the Turn of the Century reports.

Statistical assessments were undertaken in pilot studies in different Member States (Austria, Denmark, Finland, France, Germany Spain and UK)

Flexible and adaptable to different and changing need for information, EUROWATERNET is designed to be implemented and developed progressively, and aimed to improve information quality and quantity as soon as possible.

EUROWATERNET and the subsequent WATERBASE is subdivided as presented in the following figure, each station being geographically referenced and the associated data for several determinands and as many years as possible (available) are collected and stored :



### **Development of Eurowaternet "Emissions"**

The task "emissions" of Eurowaternet process was started more recently (1997) and is still in a conception phase.

EEA's original request to ETC/IW (1997) was *"To develop and start implementation of IEI (Integrated Emissions Inventory) to air and water."*

The need for an emissions inventory at European level has stemmed from the implementation of EU Directives, and the work programme of the EEA, and of international organisations dedicated to the protection of Rivers and Seas. There are a number of requirements for information on emissions at European level. However, the first step in developing an assessment method revealed the fundamental differences between emissions to water and to air, as regards to the possible nomenclatures and the pollutant pathways from their point of emission to their discharge into the environment. Indeed, unlike emissions to air, emissions to water include complex pathways (e.g. leaching or runoff) and purification processes within the medium. Consequently, it was decided to develop independently an inventory of emissions to water, called European Emissions Inventory (EEI).

Indeed, emission registers and inventories are important tools for the formulation and monitoring of pollution control policies at national and international levels. They support pollution control policies in the following points:

- Development of technical and regulatory measures to reduce emissions. Emissions inventories result in the identification of most important pollutant sources and thus allow focusing pollution reduction measures on these sources.
- Monitoring of measures already in force. Comparison of emissions inventories carried out at different period makes it possible to assess the evolution of emission figures and relate it to implemented measures.
- Analysis of the relationships between economic factors and emissions and between emissions and state of the environment, allowing the development of regulatory measures and of assessment of existing measures.
- Providing information to the general public.

In order to fulfil these possible functions, emissions inventories may contain very detailed data. However, for many purposes these data must be aggregated, perhaps in several different ways. Aggregation may deal either with the type of emissions (e.g. according to the substance, to the source or to the process involved) or with the presentation of the results (temporal or spatial aggregation).

In the framework of EUROWATERNET, an inventory of emissions to water is needed, with access to "pressure" information. EUROWATERNET uses at the moment "proxy" indicators of the pressures in the catchments above lakes and rivers, and in the recharge area of groundwater bodies (e.g. land-use categories, catchment population density). The emissions data will be stored in WATERBASE and used to formulate appropriate indicators for the issues that the EEA is required to assess.

## **1. From 1997 to 2000**

The first step undertaken was to develop the framework methodology and design of the Integrated Emissions inventory for all sources of pollutant, point and diffuse.

It gave rise to a report published : « *Technical report no 8. A European Inventory of Emissions to Inland waters. A first proposal.* » and another report to be published, and to a data collection guidebook

The Main conclusions highlighted were:

- The Lack of homogeneous data, and multiplicity of current methods require a step by step approach,- The complexity of pollutant's pathways lead to propose a reductible procedure, based on the emissions to water
- The need to highlight the nomenclature, propose a common GIS, show transfer processes, identify reporting needs, first test with a base set of determinants
- Classification based on 7 groups of sources
- Basics of the system: DPSIR, calculation system based on 4 pollutant's pathways, use of registered and modelling data, use of technical coefficients, step-by-step implementation
- An « upward » approach : Individual *input* data giving aggregated *output* results, for different geographical coverage

## **2. The proposed system**

What a European Emissions Inventory is ?

It's a complete system that encompasses :

- all the quantities of polluting substances
- emitted from all the sources
- by all the pathways

The methodology is intended to address all possible sources of emission. The basic methodology must be flexible and simple, yet take into account the complexity of the various steps in a pollutant's pathways. It must not be restricted to emissions submitted to legal reporting requirement since it covers only part of possible sources: legal requirements are restricted to specific source groups or pollutant groups and minimum emission level. The EEI intends to provide emission data at various geographical levels (local, regional, national, European) and with different groupings (e.g. urban and domestic/industrial sources). Thus, emission assessments carried out by different Member States must provide comparable data.

The long-term aim of the emission task is to provide Member States with a single template related to emissions data. Member States would be required to fill this template and transmit it to the European Commission, which would be in charge of managing these data to extract the information needed for Directives assessment. This single template would also integrate needs of some international

organisations so that Member States will have to report emissions information once only.

It should be emphasised that Member States are not subject to any mandatory request to apply the emissions inventory methodology proposed by the EEA. However, as far as any State is part of the European Community, it has to respect European Directive, and thus has to report on emissions when required. The proposed methodology constitutes a tool available to Member States in order to facilitate emissions reporting and mainly to avoid duplication. One Member State may refuse to apply it and report on emissions using its own methodology. However, in this case, results comparability would not be reliable, and their use at European level may be questioned.

The proposed system is based on the EEA's reference, the DPSIR approach where Pressures (sources of emission) depend on Driving Forces (activities creating the emissions). It combines both the "register" and "model" approaches. In both cases, data can originate either from measurements or from estimations based on models. The whole calculation system remains the responsibility of National Authorities.

The methodology consider the information unit, called group of sources, which comprises different elements that are :

- a source category : seven categories that are Urban, Industrial, Agricultural, Exploitation of forests, transport, wastes, natural and semi-natural contributions
- a substance : a list based on Directives priority lists
- a spatial aggregation : the catchment level of about 5 to 10 000 km<sup>2</sup>, the NUTS 2 or 3 level
- a temporal aggregation : the annual load

this information unit and the pathways: four steps from the emission to the receiving media, will be provided to Eurowaternet through two types of tables, progressively implemented for each group of sources and each substance. These tables are :

|        | Raw pollution | Final pollution | Purified pollution |
|--------|---------------|-----------------|--------------------|
| Source |               |                 |                    |

where :

- the raw pollution is the real pollution emitted by the source,
- the purified pollution is the one destroyed and that never gains waters (including that stored in sludge)
- and the final pollution is the sum of all the pollution that gain waters
- and raw pollution = final + purified pollution

and :

|               | Entry | Immediate discharge | Internal purification | Transfer to next step |
|---------------|-------|---------------------|-----------------------|-----------------------|
| Raw pollution |       |                     |                       |                       |
| Collection    |       |                     |                       |                       |
| Purification  |       |                     |                       |                       |
| Final         |       |                     |                       |                       |

where the pathway is divided in four steps:

- the raw pollution is the real pollution emitted by the source,
- the collection groups the systems through which the substance flows,
- the purification groups the systems through which the substance is purified,
- the final groups the systems through which the residual substance is discharge in water.

and for each step :

- the entry is the quantity of substance that enters this step,
- the immediate discharge is the quantity of substance that directly gains waters in the step,
- the internal purification is the process by which the substance is destroyed and never gains waters and that occurs in the step,
- the transfer to next step is the quantity of substance that will be the entry of the next step.

In this framework :

(entry – immediate discharge – internal purification) = transfer to next step, and  
(sum of all the immediate discharge of each step + sum of all internal purification of each step) = the raw pollution entry.

This last table is the minimum level that meets all the needs.

The information can then be used to assess the suitability of the political tools, such as Directives, in each sector and for each step such as collection or purification, and hence give a powerful tool to improve the situation to the benefit of the waters. The power of this system is however much greater than the simple answer to the Directives and will constitute a real improvement in the assessment and implementation of the water policy.

#### **4. Main findings**

The emission evaluation systems proposed by both the Netherlands and France among others use the same basic system for emission calculations so that data restitution are easy to obtain.

The general methodology presented in this report was applied to the Loire-Bretagne Water agency area, the first runs being limited to total nitrogen, for which agricultural surplus data had been computed separately.

The proposed methodology, although very ambitious, remains simple. Using the informations asked by the different existing Directives, it doesn't need much more collection efforts. Although this information was proved to be incomplete, especially for those trace-substances, it can already be used for the major substances.

The already existing information systems at the national level appear to be enough implemented to allow the use of this method to build an inventory of emissions at the EEA level.

The development of a new model specifically for the EEA needs is not necessary. Different international working groups (OECD, UNEP...) and national ones are actively involved in the development of nomenclature for sources of emission or in register framework. An harmonisation of all this work is however needed, and could be in a first step done through the elaboration of a database of a core set of definitions, coefficients, and models that could be used for the calculation of the tables required. Improvement with input data and a pilot study at the EU level on a limited number of parameters are also needed.

### **5. 2001 Objectives :**

For 2001 a Workprogram was built with four main elements that are :

- develop the collection of data
- implement EUROWATERNET and WATERBASE emission : data model and deliverables for Waterbase, scheme for data-collection through Eurowaternet
- help the process by giving some guidelines and tools : collection of methods, models, coefficients

co-operate with other international organisations

**EEA and emissions to water :  
first steps towards european reporting and  
assessment tools**

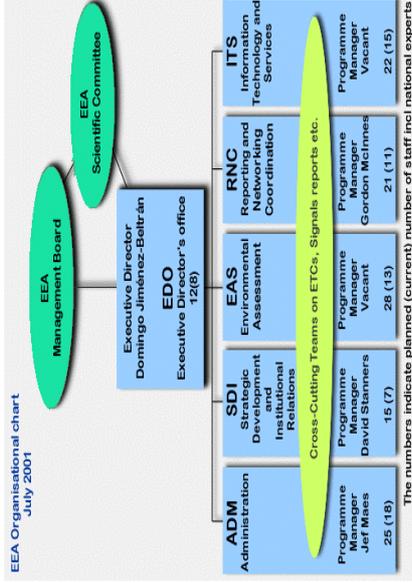
**Presented by :  
Benoît FRIBOURG-BLANC**

## The European Environment Agency

- Established by the European Union in 1990 by Council Regulation 1210/90 (subsequently amended by Council Regulation 933/1999, based in Copenhagen)

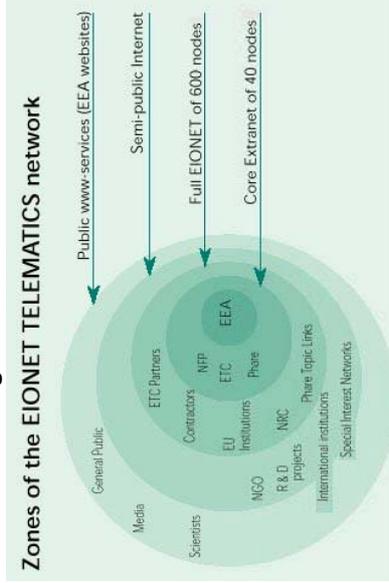


- *“the EEA aims to support sustainable development and to help achieve significant and measurable improvement in Europe’s environment through the provision of timely, targeted, relevant and reliable information to policy making agents and the public.”*
- 24 members (15 EU, 3 EFTA and 4 PHARE, Cyprus and Malta), soon 31 members



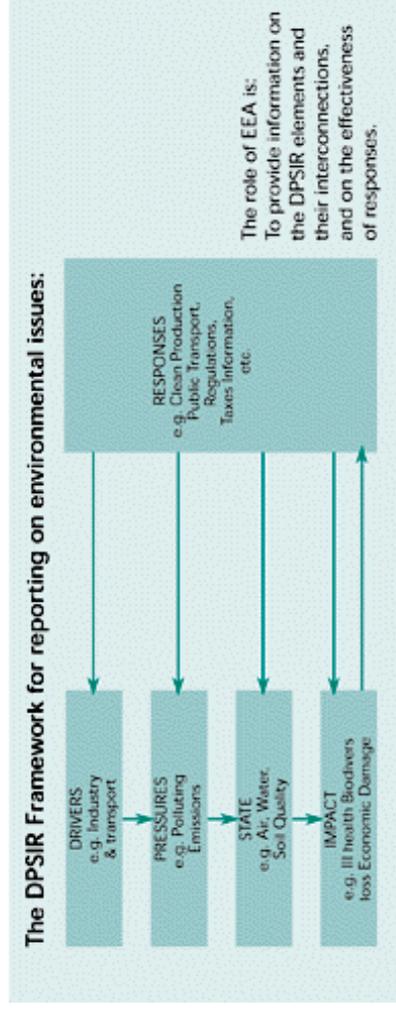
- **73 staff members**

- **5 Topic Centers : Waters, Terrestrial Environment, Air and Climate Change, Nature Protection and Biodiversity and Waste and Material Flows**
- **EIONET : a network of about 600 organisations**



# DPSIR framework

- A conceptual framework for all the work and the reports



# The European Topic Centre for Water

- A consortium of 10 national organisations led by Water Research Centre (UK)
- Merging of former ETC on Inland Waters and on Marine and Coastal Environment
- All types of water body considered : Rivers, Lakes, reservoirs, groundwater
  - transitional, coastal and marine waters to be included
- Tasks :
  - ⇒ to develop a core set of indicators
  - ⇒ to progressively improve data flows on all types of waters
    - Quality
    - Quantity
    - Emissions and pressures

## Indicators in support of policy

- future EEA assessment and reporting activities should be indicator-based and policy relevant
- Aim : simplify complex information and crystallise key messages
- A preliminary core set of indicators for water (7 policy questions and 4 indicator-types, DPSIR framework)
- A database of the raw data used
- A step-by-step implementation : indicators to be produced in the short, medium and long term
- A European-wide coverage
- A regular update and reporting

# EUROWATERNET

*EUROWATERNET is the process by which the EEA obtains the information on water resources (quality and quantity) it needs to answer questions raised by its customers."*

- Focused on Rivers, lakes, reservoirs, groundwater bodies
- To be extended to transitional, coastal and marine waters
- Developed through progressive implementation of
  - Basic,
  - Impact
  - Fully representative networks of stations.
- Built with statistical data to assess confidence of changes

**Eurowaternet basic started implementation (EU 15 plus EFTA countries)**

|                                 | AT | BE | DE | DK | ES | FI | FR | GR | IR | IS | IT | LI | LU | NL | NO | PT | SE | UK |
|---------------------------------|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|
| <b>Rivers</b>                   | Y  | Y  | Y  | Y  | Y  | Y  | Y  | Y  | Y  | N  | Y  | N  | N  | Y  | Y  | N  | Y  | Y  |
| <b>Lakes</b>                    | Y  | Y  | Y  | Y  | Y  | Y  | Y  | Y  | Y  | N  | N  | N  | N  | Y  | Y  | Y  | Y  | Y  |
| <b>Ground water</b>             | Y  | Y  | Y  | Y  | Y  | Y  | Y  | Y  | Y  | N  | N  | N  | N  | Y  | N  | N  | Y  | Y  |
| <b>Quantity (pilot studies)</b> |    | Y  | Y  |    |    |    |    | Y  |    |    |    |    |    |    |    |    |    | Y  |

# EUROWATERNET Emissions to Water

## **The main objective :**

To develop an operational and unique framework for the assessment and inventory of emission to water, whatever the source of emission, for point sources as well as for diffuse sources of emissions.

## **Further objectives :**

- To assess emissions from reporting data according to European legislation requirements
- To assess emission emitted by sources not submitted to legal monitoring
- To assess emission regardless of the type of data available in the various member States
- To allow the use of both individual emission data from each plant and national lumped data
- To provide emission data at various geographical levels

## From 1997 to 2000

**Objective :** Development of the Framework and design of the Integrated Emissions inventory for all sources of pollutant point and diffuse.

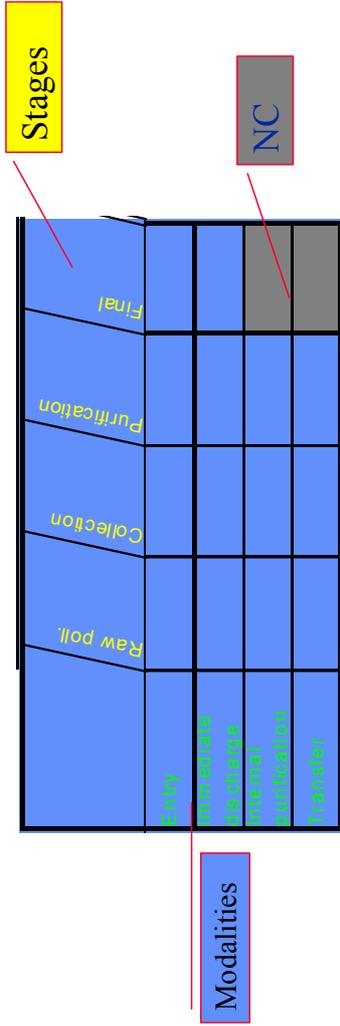
**Products :** Two reports produced and a data collection guidebook

**Main conclusions :**

- Lack of homogeneous data, multiplicity of current methods require a step by step approach
- complexity of pollutant 's pathways → proposition for a reducible procedure, based on the emissions to water
- Need to highlight the nomenclature, a common GIS, transfer processes, reporting needs, first test with a base set of determinants
- Basics of the system: DPSIR, calculation system based on 4 pollutant 's pathways, use of registered and modelling data, use of technical coefficients, step-by-step implementation
- An « upward » approach → Individual *input* data giving aggregated *output* results, for different geographical coverage

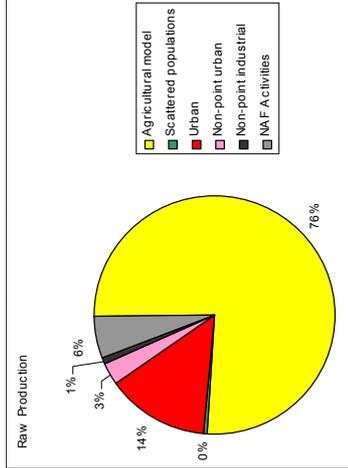
## Concepts of the emissions system

- Classification based on 7 groups of sources (branch)
- Spatial and temporal aggregation
- A wide list of substances
- All emissions processed according to a 4x4 scheme, resulting in source /vector output.
  - » 4 stages of calculation by source,
  - » 4 modalities of emission per stage

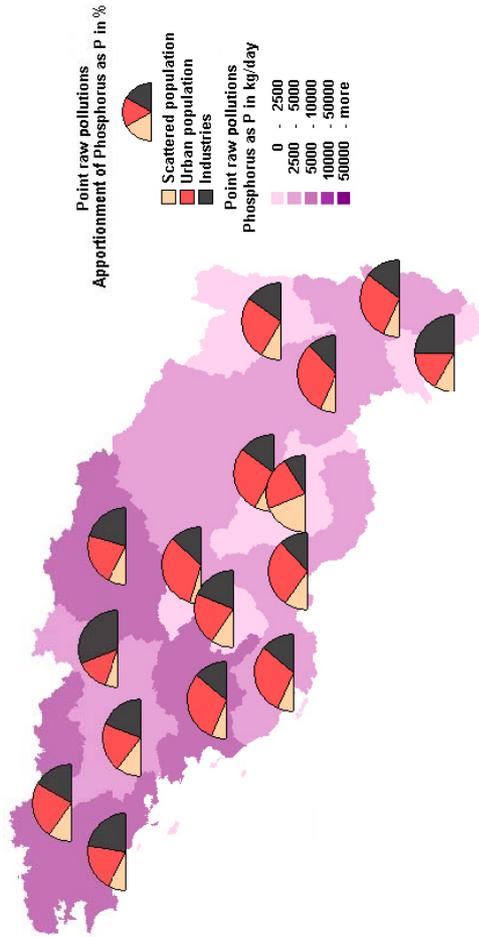


# Possible restitutions

(French pilot study)



Phosphorus raw pollution, Loire Bretagne Water Agency (1997 provisional)



## 2001

- **develop the collection of data**
- **implement EUROWATERNET and WATERBASE emission**
- **help the process by giving some guidelines and tools**
- **co-operate with other international organisations**



## **Appendix C**

### **Agenda**



## *AMAP Workshop on Sources, Emissions and Discharges*

*Norwegian Institute for Air Research (NILU), Kjeller, Norway  
23 - 24 August, 2001*

### **Program**

#### **Thursday, 23 August, 2001**

- |                   |  |
|-------------------|--|
| 09:00 - 09:20     | Registration, coffee   |
| 09:20 - 09:40     | Welcome<br>Meeting objectives and structure<br>Expected product of the meeting<br>by Jozef M. Pacyna |
| <b>Session I:</b> | <b>Emission Sources and Inventories</b><br><b>Chair: Suzanne Marcy</b><br><b>Rep.: Simon Wilson</b>  |
| 09:40 - 10:00     | Knut Breivik:<br>"Global emission of selected POPs".   |
| 10:00 - 10:20     | Jozef M. Pacyna:<br>"Global emissions of mercury, lead and cadmium".                                 |
| 10:20 - 10:40     | Janina Fudala:<br>"Emission inventories in Poland".  |
| 10:40 - 11:00     | Sergey Kakareka:<br>"Sources of heavy metals and POPs environmental<br>contamination in Belarus".    |
| 11:00 - 11:20     | Pieter van der Most:<br>"PRTR development".  |
| 11:20 - 11:40     | Slava Gordeev:<br>"Riverine transport of heavy metals to the Russian Arctic".                        |
| 11:40 - 13:00     | Lunch  |
| 13:00 - 13:20     | Lars Asplin:<br>"Modeling of pollutant transport in the marine environment".                         |

**Thursday, 23 August, 2001 (continued)**

- Session II:                   Transport of pollutants to the Arctic**  
**Chair: Peter Outridge**  
**Rep.: Knut Breivik**
- 13:20 - 13:40               Jesper H. Christensen:  
"Atmospheric modeling activities inside the Danish AMAP program".
- 13:40 - 14:00               Alexey Gusev  
"Modeling of HM & POP transport with air masses".
- 14:00 - 14:20               Mark Cohen:  
"Source-receptor modeling in North America: Lessons learned from attempts to model the transport and deposition of atmospheric toxics to the Great Lakes".
- 14:20 - 14:40               Paul Bartlett  
"North American Dioxin Emission, Transport and Deposition to the Arctic".
- 14:40 - 15:00               Coffee
- 15:00 - 15:40               Kevin Jones:  
"POPs in the Global atmosphere: putting the Arctic in the context".
- 15:40 - 16:00               Peter Outridge:  
"Discriminating natural from anthropogenic metals in Arctic marine biota, using long-term temporal trends in calcified tissues".
- 16:00 - 16:20               JoLynn Carroll:  
"Fluxes and biogeochemical processes for contaminants in Northern Seas".
- 16:20 - 16:40               S. Venkatesh:  
"The Beta-HCH story".
- 16:40 - 17:00               Benoit Fribourg-Blanc  
"The role of the ETC-DPSIR framework - indicators in support of policy - development of Eurowaternet - development of Eurowaternet - Emissions (pressures) - work done so far - work to be done in future"  
End of Session II
- 17:00- 17:20               Jozef M. Pacyna:  
Description of Session III.
- 19:00                         Dinner at Grand Café, Karl Johansgate 31, Oslo

**Friday, 24 August, 2001**

**Session III: Improvement of Working Document**  
**Group A: Chair: J.M. Pacyna**  
**Repr.: K. Breivik**  
**Group B: Chair: M. Cohen**  
**Repr.: S. Wilson**

**Rep.: Knut Breivik**

|               |   |
|---------------|---|
| 09:00 - 11:00 | Review of the Working Document in 2 groups working in parallel  |
| 11:00 - 11:15 | Coffee  |
| 11:15 - 12:30 | Review continues  |
| 12:30 - 13:30 | Lunch   |
| 13:30 - 14:30 | Presentation of the working group discussion outcome  |
| 14:30 - 16:00 | <ul style="list-style-type: none"> <li>• Gaps in knowledge</li> <li>• Final comments, conclusions</li> <li>• Recommendations</li> </ul> |
| 16:00         | End of the Workshop   |



**Appendix D**  
**List of Participants**



**List of Participants: AMAP Workshop on Sources, Emissions and Discharges,  
23 –24 August, 2001, Oslo Norway**

23 August, 2001

| Country | Last name | First name     | Institute name | Mailing address  | Direct phone   | Direct fax              | e-mail                  | Institute phone   | Institute fax       | Comments |
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