

ATMOSPHERIC MERCURY DEPOSITION FLUXES IN EUROPE: CONTRIBUTIONS FROM EUROPEAN ANTHROPOGENIC EMISSIONS AND FROM GLOBAL BACKGROUND CONCENTRATIONS

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ABSTRACT

Estimates of the relative importance of European anthropogenic emissions versus global background concentrations for the atmospheric mercury input to the Baltic Sea have been inferred from a numerical simulation model. This model uses the Eulerian reference frame of the Acid Deposition and Oxidant Model (ADOM) and incorporates detailed physical and chemical processes of mercury in the atmosphere. The model predicted annual mercury input rate to the entire Baltic Sea for 1997 and 1998 is in the range of 2.5-3.8 tons per year with a global background contribution of about 50% of the total input and a tendency to go to higher background contribution percentages from the central to the northern parts of the Baltic Sea. These results reveal a substantial influence of global background concentrations on the mercury input to the Baltic Sea and are hence an important issue for European marine environmental protection agencies in developing effective control strategies for atmospheric mercury load reduction of the Baltic and its surrounding countries.

INTRODUCTION

In remote areas, atmospheric mercury consists almost exclusively of gaseous elemental mercury (Hg^0) with a global atmospheric residence time of roughly 6-24 months. Thus, Hg^0 can undergo long-range transport on a global scale and global background concentrations of Hg^0 may have a significant impact on the mercury deposition fluxes to remote terrestrial and aquatic environments in Europe. On the other hand, mercury is emitted from a variety of central European combustion and industrial sources and the Hg^0 fraction of these emissions also contributes to the deposition to the above mentioned areas.

An assessment of the potential ecological and health risks associated with atmospheric mercury fluxes to the Baltic Sea requires an understanding of the relationships between European sources of emission to the atmosphere and the levels of concentrations measured in ambient air and precipitation in the Baltic Sea area. However, the complexity of the physico-chemical processes of atmospheric mercury species makes results from measurement programs difficult to interpret without a clear conceptual model of the workings of the atmosphere. Further, measurements alone cannot be used directly by policy-makers to form balanced and cost-effective strategies for dealing with this problem; an understanding of individual processes within the atmosphere does not automatically imply an understanding of the entire system. A complete picture of individual mercury processes and their interactions with the atmospheric system as a whole can only be obtained by means of numerical modeling.

THE ADOM MODEL SYSTEM

Recent progress in understanding the atmospheric mercury cycle (Schroeder and Munthe, 1998) has allowed for direct modeling of the complex non-linear mercury chemistry by fully three-dimensional Eulerian models. As a first step in this direction, the cloud mixing, scavenging, chemistry and wet deposition modules of the Acid Deposition and Oxidants Model (ADOM), originally designed for regional-scale acid precipitation and photochemical oxidants studies (Venkatram et al., 1988; Misra et al., 1988) have been restructured to accommodate recent developments in atmospheric mercury chemistry. A stand-alone version of these modules referred to as the Tropospheric Chemistry Module (TCM) was designed to simulate the meteorology and chemistry of the entire depth of the troposphere to study cloud mixing, scavenging and chemical reactions associated with precipitation systems that generate wet deposition fluxes (Petersen et al., 1998). The TCM chemistry scheme was developed by systematic simplification of the detailed Chemistry of Atmospheric Mercury (CAM) process model, which is based on current knowledge of physico-chemical forms and transformation reactions of atmospheric mercury species (Pleijel and Munthe, 1995).

After comprehensive testing under different environmental conditions the TCM has been implemented into both a North American and a European version of the full ADOM model. Within the constraints of the available computer resources and input data, these models incorporate an up-to-date understanding of the detailed physical and chemical processes in the atmosphere. In both models, the vertical grid consists of 12 unequally spaced levels between the surface and the top of the model domain at 10 km. The North American and the European version are run for a grid cell size of 127 by 127 km (fine mesh Canadian Meteorological Center (CMC) grid) over a 33 by 33 grid domain and of 55 by 55 km (High Resolution Limited Area Model (HIRLAM) grid) over a 76 by 76 domain, respectively.

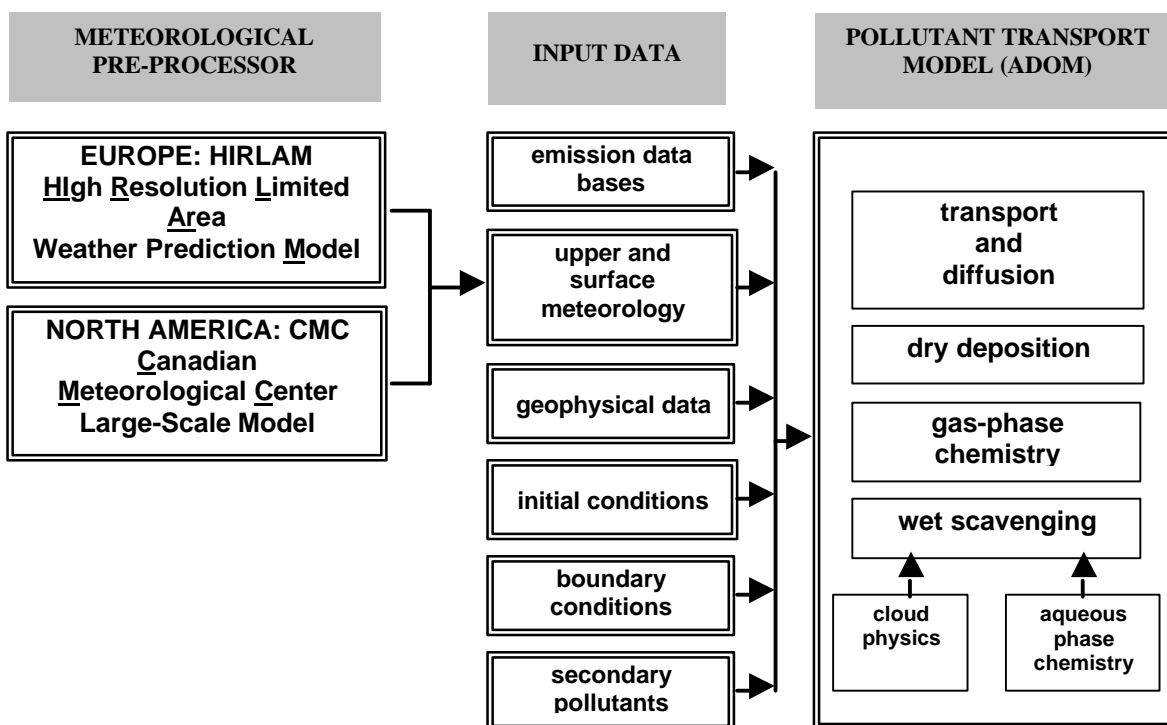


Fig. 1. Relationship between ADOM modules and input parameters.

The major modules making up the mercury version of ADOM together with the model input data sets are schematically depicted in Fig. 1. The details of each module comprising the original ADOM version for acid rain studies are given in ERT (1984). The development and testing of the mercury wet scavenging module consisting of cloud physics and mercury gas and aqueous phase chemistry sub-modules is described in detail in Petersen et al.(1998).

RESULTS AND DISCUSSION

The model has been applied within the EU MAST III Baltic Sea System Study (BASYS) subproject 'Atmospheric Load' during two episodes, i.e. a BASYS summer network study from June to August 1997 and a winter network study from February to March 1998. The general objective of the model simulations was to quantify the atmospheric long-range transport of mercury species over Europe. According to the specific objective of the 'Atmospheric Load' subproject the Baltic Sea was the area for which the relationship between European mercury sources and the mercury input via the water surface had to be established. Table 1 summarizes Baltic Sea total and global background induced daily, monthly and annual deposition fluxes and input rates during the two network studies mentioned above. The total fluxes and input rates have been obtained by employing a data base for anthropogenic mercury emissions in Europe (Umweltbundesamt, 1994) and by assuming Hg^0 concentrations of 1.5 ng m^{-3} constant in space and time at the inflow boundaries of the model domain. The contributions from global background have been quantified by keeping the boundary concentrations but cutting off the anthropogenic mercury emissions in the model simulations. As can be seen from Table 1, deposition fluxes and input rates are higher during winter due to more intense transport from the main emissions areas and inflow boundaries, lower mixing layer heights and more precipitation. In both cases, wet deposition is the main contributor to the total input (about 70% in summer and 90% in winter). The reason for the relative low dry deposition flux is twofold: Firstly, dry deposition of Hg^0 over water surfaces is assumed to be zero in the model and secondly, the average air concentrations and hence the dry deposition of the other two species, namely HgCl_2 and $\text{Hg}(\text{part.})$ is minor, because they are readily dry deposited over land close to the land-based sources and the inflow boundaries, respectively.

A comparison of the annual total and background induced mercury input rates to the Baltic Sea in Table 1 reveals a substantial background contribution (approximately 40% in winter and 50% in summer). These results indicate that due to its relative long atmospheric residence time gaseous elemental mercury is particular case for environmental policy makers such as the BALTIC MARINE ENVIRONMENT COMMISSION -HELSINKI COMMISSION-(HELCOM) in developing effective control strategies for the input reduction of mercury species to the Baltic Sea: There is strong evidence from this modeling exercise that it is not sufficient to identify the transport of mercury species from European land based anthropogenic sources only but that an assessment of long-range transport induced atmospheric deposition fluxes requires the implementation of global mercury background concentrations. For further improvements of model results an interfaced set of numerical models on Baltic, European and hemispheric scales should be introduced to take time and space dependent Hg^0 concentration profiles into account which may have a significant effect on the deposition pattern in the model domain.

Table 1. Model estimates for atmospheric mercury deposition fluxes and input rates to the Baltic Sea. (Area = 415 000 km²).

	summer 1997				winter 1998			
	total		contribution from global background		total		contribution from global background	
	deposition flux ($\mu\text{g m}^{-2}$)	input rate (kg)	deposition flux ($\mu\text{g m}^{-2}$)	input rate (kg)	deposition flux ($\mu\text{g m}^{-2}$)	input rate (kg)	deposition flux ($\mu\text{g m}^{-2}$)	input rate (kg)
daily dry	0.00476	2.0	0.00110	0.5	0.00285	1.2	0.00126	0.5
daily wet	0.01152	4.8	0.00690	2.9	0.02285	9.5	0.00858	3.6
daily total	0.01628	6.8	0.00800	3.4	0.02570	10.7	0.00984	4.1
monthly dry	0.15000	60.9	0.03000	12.5	0.09000	35.9	0.04000	15.9
monthly wet	0.35000	146.0	0.21000	87.2	0.70000	288.4	0.26000	108.1
monthly total	0.50000	206.9	0.24000	99.7	0.79000	324.3	0.30000	124.0
annual dry	1.74000	730.0	0.40000	166.0	1.04000	431.0	0.46000	191.0
annual wet	4.20000	1752.0	2.52000	1045.0	8.34000	3461.0	3.13000	1298.0
annual total	5.94000	2482.0	2.92000	1211.0	9.38000	3892.0	3.59000	1489.0

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