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Intercomparison study of atmospheric mercury models: 2. Modelling results vs. long-term observations and comparison of country deposition budgets

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Abstract

Five regional scale models with a horizontal domain covering the European continent and its surrounding seas, two hemispheric and one global scale model participated in the atmospheric Hg modelling intercomparison study. The models were compared between each other and with available measurements from 11 monitoring stations of the EMEP measurement network. Because only a very limited number of long-term measurement records of Hg were available, significant attention was given to the intercomparison of modelling results. Monthly and annually averaged values of Hg concentrations and depositions as well as items of the Hg deposition budgets for individual European countries were compared. The models demonstrated good agreement (within ±20%) between annual modelled and observed values of gaseous elemental Hg. Modelled values of Hg wet deposition in Western and Central Europe agreed with the observations within ±45%. The probability to predict wet depositions within a factor of 2 with regard to measurements was 50–70% for all the models. The scattering of modelling results for dry depositions of Hg was more significant (up to ±50% at the annual scale and even higher for monthly data). Contribution of dry deposition to the total Hg deposition was estimated at 20–30% with elevated dry deposition fluxes during summer time. The participating models agree in their predictions of transboundary pollution for individual countries within ±60% at the monthly scale and within ±30% at the annual scale. For the cases investigated, all the models predict that the major part of national anthropogenic Hg emissions is transported outside the country territory.

Keywords: Atmospheric mercury; Numerical modelling; Model intercomparison; Transport and deposition; Transboundary pollution; Uncertainty

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1. Introduction

Human activity can significantly disturb the natural environmental Hg cycle and cause enhanced accumulation of Hg in soil, water, and vegetation. Subsequent bio-magnification in food webs may lead to elevated concentrations of Hg in marine fish, mammals and birds, and finally may result in significant human exposure to Hg through the diet. The atmosphere plays an important role in the delivery to and cycling of Hg in ecosystems; understanding and quantifying this role is of critical importance. Modern methods of monitoring are not able to provide spatially comprehensive information on total (both wet and dry) Hg deposition. In addition, by itself, monitoring is not able to provide comprehensive information on source attribution for atmospheric deposition and the extent of transboundary Hg pollution. Numerical chemical transport models must be used to provide the above information. Comparison of modelling results obtained by different chemical transport models as well as comparison of the modelled values with available measurements provides information about the uncertainty in such models.

To this end, an intercomparison project for evaluation of numerical models of Hg long-range atmospheric transport and deposition on the European scale has been carried out under the auspices of EMEP. Results of the first phase of the intercomparison – a study of physical and chemical schemes of Hg transformations applied in the models – were published earlier (Ryaboshapko et al., 2002). Results of the second phase – an intercomparison of modelling results for relatively short-term detailed observations in Europe – are presented in a companion paper (Ryaboshapko et al., 2007). The final phase of the project – described here – included a comparison of modelling results with long-term observations of Hg deposition fluxes and concentrations in air and precipitation as well as a comparison of model predicted atmospheric budgets of Hg species in the entire EMEP domain and for selected European countries. The major emphasis of the final phase was the evaluation of the model performance in policy-oriented applications and the development of additional information about the uncertainty in modelling results. A summary of the results of the final phase is presented here; additional details are available in report form (Ryaboshapko et al., 2005).

2. Program and participating models

The main task of this study was to characterize the ability of contemporary chemical transport models to predict Hg levels in the atmosphere and depositions on long-term time scales (months to years). The models are a potentially valuable tool in assessing Hg accumulation in the environment and long-term adverse effects on human health, and so it is important to evaluate their accuracy and model-to-model differences. Three types of model results were analyzed via comparison with available measurement data and by intercomparison of the results of the different participating models: Hg concentration in ambient air, wet and dry deposition fluxes, and balances of atmospheric deposition for selected European countries.

In contrast to the short-term (second) phase of the project, only gaseous elemental mercury (GEM) was considered in the atmospheric concentration aspects of the analysis, because there were insufficient long-term data records for other atmospheric Hg forms — reactive gaseous mercury (RGM) and total particulate mercury (TPM). GEM is characterized by a long residence time in the free troposphere (months to a year) and is therefore particularly important in long-range atmospheric transport. Comparison of model predictions with measured GEM concentrations provides important information about how well a given model is able to describe the regional and global transport of Hg in the atmosphere, including an accounting of major sources and sinks of atmospheric Hg. Evaluation of wet and dry deposition processes provides information about the ability of models to estimate Hg atmospheric loads to ecosystems. Finally, intercomparison of model results for country-specific deposition budgets is useful for assessing the current level of model performance in answering policy-relevant source-attribution questions.

Since there is very limited long-term measurement data for atmospheric Hg in Europe (there are only about a dozen sites measuring Hg in air and/or precipitation on a regular basis), it is impossible to perform a comprehensive evaluation of a model performance by comparison of modelling results with measurements. Nevertheless, comparison with the limited available data allows at least an initial characterization of the overall accuracy of the model simulations. In addition, for processes that cannot be routinely measured, e.g., dry deposition and source attribution, comparison of simulation results obtained by different models yields information about the uncertainty in the estimates.

Eight models were involved in this phase of the intercomparison:

- ADOM, GKSS-Forschungszentrum, Germany (Petersen et al., 2001);
- CMAQ, EPA, USA, (Bullock and Brehme, 2002);