E3S Web of Conferences **1**, 17006 (2013)
DOI: 10.1051/e3sconf/20130117006

Mercury simulations within GMOS: Analysis of short-term observational episodes

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Abstract. A number of contemporary chemical transport models for mercury are applied within the framework of the EU GMOS project to study principal processes of mercury transport and transformations in the atmosphere. Each model is involved in simulation of short-term episodes corresponding to particular Hg measurement campaigns in Europe and other regions. In order to evaluate different physical and chemical mechanisms the models perform sensitivity runs with various parameterizations and/or combinations of considered processes. The modeling results are compared to detailed measurements of Hg species (Hg^0/TGM, RGM, HgP) with high temporal resolution (hours) aiming at reproduction of short-term temporal variability of Hg air concentration.

Key words: Mercury simulation, chemical transport models, short-term episodes, GMOS project

Introduction

Mercury (Hg) is widely recognised as a toxic chemical capable of long-range transport, bioaccumulation in the environment and adverse impact on human health and the environment. Due to its long residence time in the free troposphere it can be easily transported between continents and affect vulnerable ecosystems of remote regions (Arctic, Antarctica, Open Ocean, etc.) Therefore evaluation of present-day Hg pollution levels as well as forecasts of future changes should take into account the global character of Hg atmospheric transport and dynamics of Hg emission changes in different continents. In this aspect, the application of global chemical transport models can supplement direct measurements of Hg concentration and deposition levels giving more comprehensive and detailed information on Hg pollution.

Applications of mercury atmospheric models on hemispheric and global scales performed by different scientific groups demonstrated satisfactory ability to evaluate background levels of Hg and its intercontinental transport as well as reasonable agreement with available measurements (Christensen et al., 2004; Seigneur et al., 2004; Travnikov, 2005; Selin et al., 2007; Dastoor et al., 2009; Jung et al., 2009; Holmes, 2010). However, current knowledge on Hg behaviour in the atmosphere and its potential to cycling between different environmental media is still incomplete. There are significant gaps in the understanding of chemical processes affecting Hg atmospheric transport and deposition, characteristics of the air-surface exchange and processes responsible for re-emission of Hg to the atmosphere.

Development and application of chemical transport models are an essential part of the EU GMOS project (Global Mercury Observation System) funded by the European Commission 7th Framework Programme (DG Research). The project is aimed at (i) establishment of a global monitoring system for mercury including

land-based, over-water and aircraft observations; (ii) improvement and validation of regional and global scale atmospheric mercury models; (iii) model application to evaluate source-receptor relationships, temporal trends and scenarios; (iv) development of an interoperable system for dissemination of the project output data.

The program of model activities within GMOS includes a number of Tasks on improvement and further development of global and regional scale models and their application for the assessment of present and future concentration and deposition levels, hindcasting historical trends, and evaluation of source-receptor relationships. The first phase includes simulation of short-term observational episodes involving detailed measurements of Hg species at ground monitoring sites, ship-based and aircraft platforms. Global and regional scale models both of the GMOS model consortium (GLEMOS, ECHMERIT, WRF-Chem, CMAQ-Hg) and of external partners of the project are involved in the study.

Multi-model simulations and analysis

A number of contemporary chemical transport models for mercury are applied to study principal processes of Hg transport and transformations in the atmosphere. Each model is involved in simulation of short-term episodes corresponding to particular Hg measurement campaigns in Europe and other regions. The observational episodes are selected to characterize Hg behavior in different atmospheric environments:

- Continental boundary layer
  (EMEP measurements at site Langenbrügge, Germany, 2009)
- Marine boundary layer
  (RV Urania cruise, Mediterranean Sea, June 2009)
- Free troposphere and UT/LS
  (CARIBIC project, Europe/global, 2009; INTEX-B campaign, North America, 2006).

In order to evaluate different physical and chemical mechanisms the models perform simulations of the observational episodes with various parameterizations and/or combinations of considered processes. The modeling results are compared to measurements of Hg species (Hg^0/TGM, RGM, HgP) with high temporal resolution (hours) aiming at reproduction of short-term temporal variability of Hg air concentration. The sensitivity analysis is focused on the following questions:

- What are relative contributions of redox chemistry and direct anthropogenic emissions to air concentration of oxidized Hg forms (RGM, HgP) in the CBL and deposition to the ground?
- What is the speciation of Hg anthropogenic emissions?
- What are the major mechanisms of Hg oxidation in the CBL and MBL?
- What is the physical state of the oxidation products (gaseous or particulate)?
- What is the role of the air-surface exchange in Hg cycling in the MBL?
- What are the prevailing mechanisms of Hg oxidation and removal in the UT/LS?

An example of the model analysis of Hg^0 concentrations in the CBL measured at the German monitoring site Langenbrügge (DE02) of the EMEP monitoring network is given in Fig. 1.

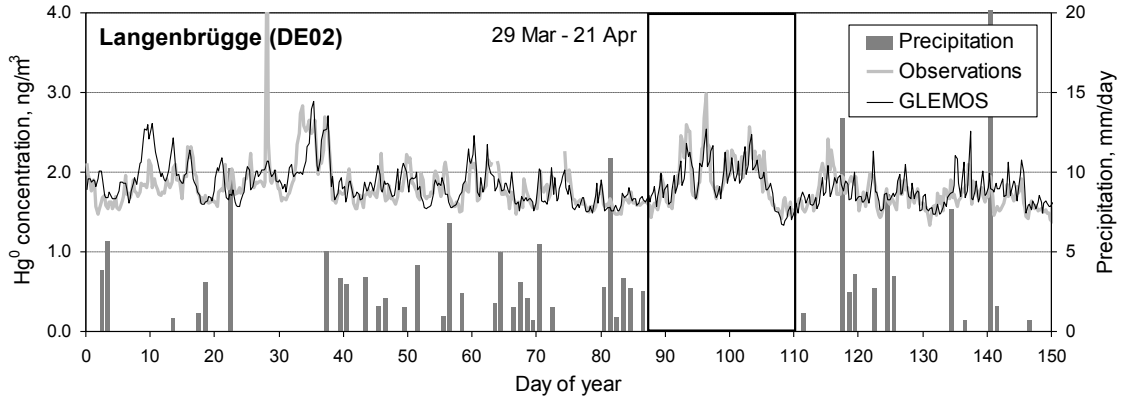

Fig. 1. Comparison of simulated and observed air concentrations of Hg^0 at site Langenbrügge, Germany (DE02) in the first half of 2009. The rectangle shows a short-term episode with no precipitation chosen for the analysis of the gas-particle partitioning in the.

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