Atmospheric Environment 44 (2010) 1806–1814

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

A global transport model of lead in the atmosphere

T. Niisoe [a], E. Nakamura [a], K. Harada [a], H. Ishikawa [b], T. Hitomi [a], T. Watanabe [c], Z. Wang [d], A. Koizumi [a,*]

[a] Department of Health and Environmental Sciences, Graduate School of Medicine, Kyoto University, Kyoto 606-8501, Japan
[b] Research Division of Atmospheric and Hydrospheric Disasters, Disaster Prevention Research Institute, Kyoto University, Uji 611-0011, Japan
[c] Miyagi University of Education, Miyagi 980-0845, Japan
[d] Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

ARTICLE INFO

Article history:
Received 25 March 2009
Received in revised form
6 January 2010
Accepted 7 January 2010

Keywords:
Transport model
Atmospheric lead
Heavy metal
Particulate matter

ABSTRACT

A global atmospheric transport model is used to calculate lead concentrations in the atmosphere. The model performance is evaluated through comparisons with observations in Europe. The model results of lead concentrations in surface air were compared with measurements in East Asia. The detailed comparisons showed generally good agreement for recent decades, although systematic underestimation was found in China. Anthropogenic lead emissions in China are estimated from economic statistics to be 56 000 t yr^{-1}, which is not small considering the economic scale of China. The underestimations suggest a hidden source of lead emissions. The emissions in Japan and Korea are derived from optimization by the model. The magnitude is about 2000 t yr^{-1}, which is much greater than that reported by the Pollutant Release and Transfer Register in Japan and Toxics Release Inventory in Korea.

1. Introduction

Lead is one of the most abundant hazardous heavy metals in the atmosphere. It exists in particulate matter in the atmosphere and is transported to a large extent by air flow. Lead enters the human body through inhalation and the consumption of food and water, and this could have serious adverse effects on human health. Leaded gasoline is recognized as being the largest source of atmospheric lead followed by nonferrous metal production and fossil fuel combustion.

In many developed countries, anthropogenic lead emission has been reduced remarkably in recent years because of the phasing out of leaded gasoline and industrial emission controls (e.g. Ilyin et al., 2007a). On the other hand, efforts to reduce emissions are insufficient in many Asian countries, several of which are in rapid economic progress. Although it is urgent to establish a system to assess transboundary air pollutants in Asia, a monitoring network and infrastructure for sharing information have not yet been established and are not likely to be established in the near future. Therefore, the development of a numerical modeling framework as a common and cost-conscious tool is required to assess atmospheric pollutants in Asia.

In this work, a global atmospheric transport model is presented to estimate long-term air concentrations and depositions of lead from the 1980s to 2000s. This project is a first step in establishing an assessment system for transboundary air pollutants in East Asia. The transport model used in this study is not unique, but we formalized all necessary processes. The model has advantage for numerical cost using an existing meteorological field. We also collected information and parameters for atmospheric lead assessment in Asian countries. We conducted a detailed and comprehensive comparison between results of a long-term model simulation and observations in East Asia of the atmospheric concentration.

2. Materials and methods

The numerical models and datasets used for model validation in the present study are described in this section. Tables and figures with numbering preceded by S are given in Section 3 of the Supplementary material.

2.1. Experiment methodology

An atmospheric transport model of lead was developed, and numerical simulations were carried out from 1979 to 2007. To evaluate the model performance, the model predictions were compared with observations in Europe, where information on lead emissions has high reliability and long-term observations are available. Lead concentrations in the atmosphere predicted by the model were then compared with observations in East Asia. The observed concentrations in Japan and Korea were compared with the model results. The model was run for two different emission

* Corresponding author. Tel.: +81 75 753 4456; fax: +81 75 753 4458.
E-mail address: Akio.Koizumi@z06.mbox.media.kyoto-u.ac.jp (A. Koizumi).

1352-2310/$ – see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2010.01.001

data sources in Japan and Korea; one dataset was derived from the national inventories and the other from an optimization of a pre-existing grid data.

2.2. Atmospheric transport model

Atmospheric lead is emitted in the form of fine particle mainly directly from leaded gasoline use, nonferrous metal production and fossil fuel combustion. Lead has no significant chemical process in the atmosphere. The removal of atmospheric lead is mediated by dry and wet deposition. It is broadly accepted that the observed mass size distribution of atmospheric lead is typically centered in the 'accumulation mode', corresponding to a diameter of 0.1–1.0 μm (e.g. Allen et al., 2001). The dry deposition velocity depends on the particle size in general, but size dependency is almost negligible for particles with diameters ranging between 0.05 and 2.0 μm (e.g. Slinn et al., 1978). Wet deposition depends on the precipitation rate rather than the particle size. Therefore, we can discard the effects of the size distribution of particles in both dry and wet deposition as is done in preexisting models of atmospheric lead (Ilyin et al., 2007b). We fix the particle size at 1.0 μm.

The numerical model employed in this work is a Eulerian atmospheric transport model. The time evolution of the lead concentration in each grid box is calculated by solving the continuity equation:

$$\frac{\partial c}{\partial t} = -\nabla \cdot \boldsymbol{F}_{\mathrm{trans}} + F_{\mathrm{em}} - F_{\mathrm{dep}},$$

where c is the concentration, F_{trans} is the three-dimensional transport flux, and F_{em} and F_{dep} are the local emission and deposition fluxes. The computation region is the globe and the horizontal resolution is 1.25°. The vertical structure consists of 12 layers with sigma coordinates of 0.99, 0.98, 0.95, 0.92, 0.83, 0.66, 0.55, 0.44, 0.33, 0.22, 0.11 and 0 stacked from the surface to 100 hPa. The typical depth of the lowest layer is about 150 m. The transport and deposition fluxes are assessed using 6-hourly meteorological fields determined from the JRA-25 reanalysis datasets provided by the Japan Meteorological Agency and Central Research Institute of Electric Power Industry (Onogi et al., 2007). The variables used in the model are listed in Table S1. The vertical air velocity in sigma coordinates is converted from the original isopressure coordinates. The change in the concentration is assessed by operator splitting with a time step of 1 h.

Mass-conservative transport flux within the Euler forward time step of 10 min is calculated using a parabolic-spline method (Emde, 1992) for advection and a box method (Kurihara and Holloway, 1967) for diffusion. The horizontal diffusion coefficient is proportional to the magnitude of the derivative of the horizontal wind velocity component parallel to the interface between two adjacent boxes (Mahlman and Moxim, 1978; Levy et al., 1982), and the vertical coefficient depends on the height, shear, and stability (Louis, 1979). The boundary layer height is derived by bulk formulation (Vogelezang and Holtslag, 1996) and the concentration is assumed to be vertically uniform in the boundary layer.

To validate the performance of the transport model, a cone-shaped distribution was advected in rotational wind fields (Williamson and Rasch, 1989). It was confirmed that the model is positive definite, shape preservative and mass conservative with high accuracy even for cross polar advection, and that numerical diffusion is sufficiently small.

Wet deposition modeling includes in-cloud and subcloud scavenging. In-cloud scavenging can occur only in the fraction of a grid box occupied by liquid cloud with precipitation, and subcloud scavenging only in the fraction covered by cloud with precipitation. Vertical transport flux in deep convective clouds is also parameterized by the water–vapor mass balance (Feichter and Crutzen, 1990).

Dry deposition in the lowest layer is estimated using a conventional framework for the dry deposition velocity. The velocity is a function of aerodynamic resistance depending on atmospheric stability and surface roughness, quasi-laminar sublayer resistance depending on the Brownian diffusivity of the particles, and the particle settling velocity given by Stokes' Law (Seinfeld and Pandis, 1998).

The model was run from 1979 to 2007. The initial lead concentration was determined by a preparatory one year run for 1979 starting with zero concentration. The detailed model formulations are described in Section 2.1 of the Supplementary material.

2.3. Lead emission

In this study, two lead sources, one anthropogenic and the other being the sea surface, are considered. Lead is emitted to the lowest layer and is assumed to be uniform inside each grid box. The anthropogenic emissions are assumed to have no seasonal variability.

2.3.1. Anthropogenic emission

The global emission of anthropogenic lead is considered. Different emission datasets are used for Europe, China, Japan and Korea, and the rest of the world.

Global anthropogenic emissions except those for Europe, Japan, Korea, and China are taken from version 1.0 of the global inventory of the Canadian Global Emission Interpretation Centre (CGEIC) (Pacyna et al., 1995). Data are presented in a 1-degree grid system for the reference year 1989. The inventory has a minimum scenario and maximum scenario. The average of the two scenarios is adopted in this work.

For Europe, an anthropogenic lead emission inventory is provided by the European Monitoring and Evaluation Programme (EMEP, http://www.emep.int/). The dataset includes a horizontal distribution of lead emission with 0.5-degree resolution for 1990. For the following years (1991–2007), the dataset gives the annual emission for each country but not the distribution (Tables S2 and S3). Therefore, we assume that the horizontal distributions for 1991 and 2007 are the same as that in 1990 and allocate the annual country emission to the corresponding horizontal grids. There are also missing data, which are replaced by linear interpolation (or extrapolation) in time. The emissions for countries reported only in 1990, which are 6% of the total emission for Europe in 2005, are assumed to be constant. The spatial distribution of lead emissions in 1990 and the temporal variability are shown in Fig. 1. Emissions in Germany and northern Europe are more than 75% less by 1995, and the same is seen for most of Europe by 2005. Emissions before 1990 in Europe are fixed at the European component of the CGEIC inventory.

For China, lead emission is derived from economic statistics for 2001. Annual emission from each province is estimated as fuel consumption and industrial production (Table S4) multiplied by corresponding emission factors (Table S5). Table S6 presents the estimated emission for each province. The total emission for China is 56 000 t yr^{-1}. The total lead emission is horizontally distributed on the basis of three assumptions (Fig. 2): (1) 60% of the total lead emission in each province is allocated to the city with the largest population and cities with populations of more than two million as listed in Table S7, (2) the ratio of emissions from the cities is proportional to the ratio of populations of the cities, and (3) the spatial distribution of the remaining (40%) total emission within each province is the same as that described by the CGEIC emission data.

In Japan and Korea, direct emission inventories are available. The Pollutant Release and Transfer Register (PRTR, http://www.env.go.jp/en/chemi/prtr/prtr.html) has data beginning in 2001 for Japan and the Toxics Release Inventory (TRI, http://tri.nier.go.kr/) has data beginning in 2002 for Korea. Table S8 presents national emissions for Japan and Korea reported by the inventories. The reported