A global transport model of lead in the atmosphere


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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...
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 (e.g. Allen et al., 2001). The dry deposition velocity depends on the size of the particle, 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 pre-existing 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:

$$\frac{\partial c}{\partial t} = -\nabla \cdot F_{\text{trans}} + F_{\text{em}} - F_{\text{dep}},$$

where $c$ is the concentration, $F_{\text{trans}}$ is the three-dimensional transport flux, and $F_{\text{em}}$ and $F_{\text{dep}}$ 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 10 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 (Kurilhara 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, Japan, 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 1988. 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
emissions for Japan and Korea are generally less than those for industrialized countries in Europe during the 2000s, as shown in Table S3. Two datasets of lead emission from Japan and Korea are used for the model. One is made directly from the inventories of the PRTR and TRI. The other is emission data optimized by applying correction factors to the Japanese and Korean components of the CGEIC emission data on the assumption that the distribution inside each nation has not changed from that described by the CGEIC data (Fig. 3).

2.3.2. Lead from sea salt

Lead emission from sea salt is derived using an empirical function of the surface wind speed, following the works of Gong (2003) and Ilyin et al. (2007b). The total emission from sea salt is 28 000 t yr\(^{-1}\) for 1990. The formulation is described in Section 2.1.7 of the Supplementary material. The emission factor of 4.0 mg kg\(^{-1}\), which has been estimated for the North Atlantic, is used for lead emission from sea water (Ilyin et al., 2007b). Ilyin et al. (2007b) noted that the contribution from the re-suspension of anthropogenic lead historically accumulated in the ocean is not negligible, although it can be overestimated in remote regions.

2.4. Observation datasets

To evaluate the model performance, comparisons with observations are made. In Europe, an atmospheric monitoring network for transboundary air pollutant was established in the late 1980s by the EMEP (Ilyin et al., 2007a). All data from the monitoring stations are binned according to the model resolution into 40 grid boxes for atmospheric lead concentrations and 75 grid boxes for precipitation lead concentrations (Fig. S1). The observed concentrations are averaged inside each box.

In Japan, aerosol particles were collected and analyzed mainly in urban areas by the National Air Surveillance Network from 1974 to 1996 (Var et al., 2000). The measurements have been continued by several local governments. In Korea, a number of heavy-metal monitoring stations in 13 cities throughout the Korean peninsula have been operated by the administrative demand of the Korean Ministry of Environment since 1991 (Kim, 2007). In China, unfortunately, no monitoring network has yet been developed. Therefore, calculated concentrations for China are compared with published observation results listed in Table S9. Observations affected by particular emission sources or meteorological events are excluded as much as possible. As for Europe, the data from the monitoring stations are binned into 23 boxes in Japan, 7 boxes in
Korea, and 32 boxes in China (Fig. S2). The average concentrations in Japan are listed in Tables S10–S12.

3. Results

3.1. Model validation using data for the European environment

The calculated concentrations both in air and in precipitation are compared with observations. The discrepancy between the model and observation is indicated by the fractional difference $f$ (Kasibhatla et al., 1997) averaged for all compared grid points:

$$f = \frac{V_{\text{mdl}} - V_{\text{obs}}}{V_{\text{mdl}} + V_{\text{obs}}}$$

where $V_{\text{mdl}}$ and $V_{\text{obs}}$ are the model value and observation value respectively. $|f| < 0.33$ indicates an error factor of less than 2.

Calculated air concentrations generally agree well with measurements for the early 1990s as indicated by $|f|$ values less than 0.33 (Fig. 4). There is slight underestimation in the late 1990s, and this increases in the 2000s. There is a similar tendency for the calculated lead concentrations in rainwater (Fig. 5). Ilyin et al. (2007a) estimated re-suspension lead flux from soil and suggested that it contributed more than 50% of the total emission for Europe in the 2000s, when anthropogenic emission had decreased significantly. The underestimation of atmospheric lead for the 2000s in our simulation is in agreement with the source estimation conducted by Ilyin et al. (2007a). This suggests the existence of other unknown sources of lead. The re-suspension flux can is a strong candidate for an unknown source, as suggested by Ilyin et al. (2007a).

3.2. Lead levels in East Asia

Fig. 6 compares the calculations and observations for China from the 1980s to 2000s. Most of the observed concentrations exceed 100 ng m$^{-2}$ yr$^{-1}$, which is much greater than concentrations observed in Europe. There is no remarkable temporal variability throughout the periods. The model roughly reproduces observed concentrations with an error factor of less than two, although there is general underestimation especially for the 1980s and 1990s.

The surface concentrations observed for Japan and Korea in 2004 are compared with concentrations calculated by the model using the inventories of PRTR for Japan and TRI for Korea. To assess the transboundary contribution, an additional simulation with only the national emission was conducted, and the result is also plotted in Fig. 7. The model results excluding remote sources are an order of magnitude less than observation values. Taking the remote sources into account, there remains an underestimation by a factor of more than two, especially in eastern Japan. The underestimation of the model is attributed not only to the inflow but also to the national emission of lead, although the effect of the inflow is significant. Therefore, it is concluded that both the PRTR and TRI underestimate lead emission, which implies the necessity to use the optimized emissions determined for Japan and Korea instead of the inventories. The results presented hereafter are only those of the model using the optimized emissions.

Fig. 8 shows national lead emissions for Japan, Korea and China in the model from 1979 to 2007. The emissions for Japan and Korea are optimized using the correction factors listed in Table S13, and the emission for China is estimated using economic statistics as described in Section 2.3.1. Japan and Korea have nearly the same
magnitude of lead emission, which decreases in the early 1980s and
the late 1990s in Japan, and in the early 1990s in Korea. Both Japan
and Korea have lead emissions of about 2000 t yr\(^{-1}\) in the
mid-2000s, which is about 14\% of that for 1980. The emission of
56 000 t yr\(^{-1}\) for China is much greater than emissions for Japan
and Korea. The emissions for Japan, Korea and China are 87\% of the
total emission for East Asia (100°–150E, 0°–60N) in 2005.

Comparisons of the calculations and observations of surface lead
concentrations for Japan from the 1980s to 2000s show fairly good
agreement as indicated by \(f\) values less than 0.1 for most of the
years.

![Fig. 4. Scatter plot of annually averaged lead concentrations (ng m\(^{-3}\)) at the surface over Europe for the model results and observations in 1990, 1998, and 2005. Dots on the solid line represent a perfect fit between calculation results and observations, and dots between the two dashed lines represent error factors of less than 2. The \(f\) value is the fractional difference averaged for all pairs of the model results and observations. A time series of the \(f\) value for each year from 1990 to 2005 is also shown. Generally good agreement in the early 1990s and underestimation in 2000s are demonstrated by descending trend of the \(f\) value.](image)

![Fig. 5. Scatter plot of the precipitation-weighted concentration in rainwater (\(\mu g L^{-1}\)) over Europe for the model results and observations in 1990, 1998, and 2005. A time series of the \(f\) value for each year from 1990 to 2005 is also shown. Generally good agreement in the early 1990s and underestimation in 2000s are demonstrated by descending trend of the \(f\) value. The tendency is similar to that of the surface air.](image)
period (Fig. 9). The highest concentrations exceed 100 ng m\(^{-3}\) in the 1980s, and the concentration decreases to several tens of nanograms per cubic meter in the 2000s. The decline shows the result of emission control efforts, but the present concentration level of surface air lead is somewhat greater than that for Europe.

Calculated concentrations for Korea generally agree with observations from 1991 to 2004 (Fig. 10). Most observed concentrations exceed 100 ng m\(^{-3}\) in the early 1990s and decrease significantly later, although they are still about 70 ng m\(^{-3}\) in the 2000s and are greater than concentrations observed in Europe and Japan.

3.3. Global budget

The annually averaged global budget (Table 1) shows that most deposition is due to precipitation, reflecting ineffective dry deposition for fine particles with a diameter between 0.1 and 1.0 \(\mu\)m. The residence time in the atmosphere is controlled mainly by wet deposition. The uncertainty of the in-cloud scavenging rate is a factor of 3 for rain and a factor of 6 for snow (Scott, 1982). That of the subcloud scavenging rate is controlled by the collection efficiency between 10\(^{-3}\) and 0.7 (Scott, 1978). The range of the residence time is from 1.8 days for the fast deposition case to 4.7 days for the slow deposition case.

Atmospheric lead is emitted at the surface and removed mainly by precipitation. There is no removal process above cloud layers. The residence time averaged over the troposphere is 2.9 days, while it is longer in the upper troposphere (6.3 days above 400 hPa). Larger wind speed makes the spatial scale of transport greater in the upper troposphere. Global-scale transport plays a more important role in the global distribution of deposition flux than in the global distribution of the surface air concentration.

![Fig. 6. Scatter plot of lead concentrations (ng m\(^{-3}\)) at the surface over China. The plot is for model results and observations from the literature for the 1980s, 1990s, 2000s, and all periods. Calculated concentrations are averaged for the period corresponding to that of the observations. There is a slight underestimation.](image)

![Fig. 7. Scatter plot of lead concentrations (ng m\(^{-3}\)) at the surface over Japan and Korea. The plot is for the model results determined using PRTR and TRI and the observations in 2004. (●) and (○) represent calculations including only the national source and those including global sources respectively. A significant underestimation remains even if the transboundary inflow is taken into account.](image)
4. Discussion

4.1. Sensitivity tests

Sensitivity tests are carried out for the year of 1990. Table 2 shows the variances in the lead burden below a height of about 1 km over the Northern Hemisphere for 50% changes in the meteorological precipitation rate, emission, and major model parameters relating to deposition or diffusion. The sensitivities are for 50% perturbation in one parameter only.

The burden is found to be most sensitive to emission. As lead does not have any chemical production or removal process in the atmosphere, the burden is linear to the emission.

The dominant removal process of atmospheric lead is wet deposition by precipitation as shown in Table 1. The process of wet deposition in the model depends on the meteorological precipitation rate. It has been reported that the precipitation rate of the JRA-25 datasets has a general overestimation of about 10% through comparison with an observation-based estimation of precipitation (Onogi et al., 2007). The sensitivity tests show that a 50% decrease in the large-scale precipitation rate causes only an 11.5% increase in the lead burden.

The lower planetary boundary layer prevents lead ventilation to the free troposphere and consequently increases the burden in the lower layers. A 50% decrease in the height of the boundary layer results in an increase of about 14% in the lead burden of lower layers. The sensitivity to the parameterization of the boundary layer is much lower than that of emission, although it is fairly high. The burden has little sensitivity to other model parameters.

4.2. Lead emission in East Asia

To simulate the distribution of atmospheric lead correctly, both a numerical model with good performance and a reliable dataset of emission are necessary. The performance of the transport model in this work was demonstrated by comparisons with observations in Europe in Section 3.1. Comparison with other models were made in Section 1 of the Supplemental material. The model was...
found to make predictions that are somewhat similar to those made by other models.

The sensitivity tests in Section 4.1 show that the model results are most sensitive to emission. Unfortunately, no reliable emission inventory is available for Asia. Therefore, the emissions for the three major countries—China, Korea, and Japan—were estimated as described in Section 2.3.1. Since there is no particular trend in the observed concentrations in China, the emission in China is assumed to have no trend and the emission is derived from economic statistics for a single year, 2001. The calculated surface concentrations in China roughly agree with observations, although there is systematic underestimation as described in Section 3.2. Such underestimation is most likely due to underestimated emission in China. However, the magnitude of the anthropogenic lead emission in China in this work, which is estimated from the economic statistics for 2001 to be 56 000 t yr\(^{-1}\) or nearly 4 times the Chinese component in the CGEIC data for 1989, is not small considering the economic scale in China. The discrepancy suggests an unknown source of atmospheric lead in China.

Obviously descending trends are found in observed concentrations in Japan and Korea. Considering that the territories of Japan and Korea are relatively small, the spatial distribution of emission inside each country is fixed at that of the existing emission inventory of CGEIC data described in Section 2.3.1. The magnitude of the annual emission in each year is optimized by multiplying the national annual emission by a correction factor. The correction factors are chosen to minimize the absolute value of the averaged fractional difference, \(f\). It should be noted that this optimization is based on the assumption that the discrepancy between model results and observations is attributed principally to the magnitude of the national emission.

The national lead emissions in Japan and Korea reported by the PRTR and TRI are less than those in major European countries reported by the EMEP in the 2000s (Tables S3 and S8). On the other hand, observed air concentrations of lead in Japan and Korea are generally higher than those in Europe in the 2000s. As shown by Figs. 4, 9, and 10, representative concentrations are less than 10 ng m\(^{-3}\) in Europe, about 20 ng m\(^{-3}\) in Japan, and about 60 ng m\(^{-3}\) in Korea. The surface air concentrations calculated by the model using the PRTR and TRI data are much less than observations in Japan and Korea as shown in Section 3.2. Although the effect of inflow from windward is significant, the discrepancies are also attributable to underestimations in the inventories. In the PRTR, emissions from the use of materials with the amount of constituent chemical being less than 1% are excused from registration. For example, fossil fuel

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Annually averaged global budget of lead in the atmosphere for 1990.</th>
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<tbody>
<tr>
<td>Burden</td>
<td>Residence time</td>
</tr>
<tr>
<td>1.9 Gg</td>
<td>2.9 days</td>
</tr>
</tbody>
</table>

Fig. 10. Scatter plot of annually averaged lead concentrations (ng m\(^{-3}\)) at the surface over Korea. The plot is for the model results determined using the optimized emission and observations in 1991, 1997, and 2004. A time series of the \(f\) value is shown from 1991 to 2004. Generally good agreement is shown in Korea.

<table>
<thead>
<tr>
<th>Table 2</th>
<th>Sensitivity of the lead burden in the lower layer ((-1) km) over the Northern Hemisphere to 50% changes in the meteorological precipitation rate, emission, and model parameters for 1990 (%).</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Pr_l): large-scale precipitation rate.</td>
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<tr>
<td>(Pr_c): convective precipitation rate.</td>
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<tr>
<td>EM: emission.</td>
<td></td>
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<tr>
<td>(V_{dry}): dry deposition velocity.</td>
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<tr>
<td>(R_i): in-cloud scavenging rate.</td>
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<tr>
<td>(R_s): subcloud scavenging rate.</td>
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<tr>
<td>(D_h): horizontal diffusivity.</td>
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<tr>
<td>(D_v): vertical diffusivity.</td>
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<tr>
<td>(H_{pbl}): planetary boundary layer height.</td>
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</tbody>
</table>

\[ \begin{array}{cccccccc}
\text{\% change} & -50\% & -2.5 & +50.0 & -1.1 & -4.1 & -6.3 & -0.2 & -2.5 & -26.7 \\
\text{\% change} & +50\% & +11.5 & +3.6 & -50.0 & +0.7 & +6.1 & +9.5 & -0.2 & +2.3 & +13.8
\end{array} \]
combustion is excluded in the inventory. Coal consumption for industrial use in Japan is $9 \times 10^7$ t yr$^{-1}$ in 2000 (SBJ, 2009), corresponding to lead emission of 900 t yr$^{-1}$ using an emission factor of 10 g t$^{-1}$. Fossil fuel combustion is included in the emission inventory for Europe reported by the EMEP (EEA, 2009). It is necessary to inspect the frameworks of pollutant registration in Japan and Korea carefully.

It should be noted that observations of atmospheric particles in East Asia have been carried out mainly in urban regions. Although observations affected by particular sources are excluded in evaluating the model as much as possible, effects of particular sources may remain in the observation results. Therefore, observations in remote regions over East Asia are necessary for more detailed model validation. It should also be noted that the samples of atmospheric particles collected in Korea and some of those collected in China include coarse particles. Lead in coarse particles collected near sources could result in underestimation in the model results.

5. Summary

A global atmospheric transport model for lead was developed as a first step in establishing of an assessment system for trans-boundary air pollutants in East Asia. The results were compared with a large number of observations in East Asia.

First, model validation was carried out by comparison with observations of atmospheric particles and rainwater in Europe. The model results generally agreed well with observations in the 1990s, when anthropogenic emission was dominant.

The anthropogenic emission in China was estimated to be 56,000 t yr$^{-1}$ using economic statistics for 2001. The calculated lead concentrations in the surface air generally agree with observations within a factor of two, although systematic underestimation was found. The underestimation suggests a lack of knowledge about lead emission.

The results obtained using the emission inventories—the PRTR for Japan and TRI for Korea—were much less than the observed lead concentrations in surface air. Those obtained using the optimized emission agreed well with observations.

It is concluded that the transport model in this work reproduces the features of the atmospheric lead distribution in East Asia. In the present study, lead was selected as a model chemical for three reasons. First, there is an accumulated collection of environmental data. Second, the fate of lead is relatively simple since it never degrades in the environment. Third, the toxicological importance of lead is well accepted. The transport model will be applied to other chemicals in the future.

Acknowledgments

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Appendix. Supplementary information

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.atmosenv.2010.01.001.

References


