Evaluation of Long-Range Transport Models for Acidic Deposition in East Asia

MAHESH J. PHADNIS AND GREGORY R. CARMICHAEL

Center for Global and Regional Environmental Research, Department of Chemical and Biochemical Engineering, University of Iowa, Iowa City, Iowa

YOICHI ICHIKAWA AND HIROSHI HAYAMI

Central Research Institute of Electric Power Industry, Komae, Tokyo, Japan

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ABSTRACT

A comparison between transport models is done to study the sulfur deposition in East Asia. A single-layer Lagrangian model with simple chemistry is compared to a multilayered 3D Eulerian model. The comparison is done for two-month-long episodes of winter (February) and summer (August) 1989. The model-predicted sulfur deposition is about 0.1 g S (m^2 month)$^{-1}$ for regions with the largest emissions. A comparison between the model-predicted and the observed values at a network of monitoring stations in Japan shows similar temporal trends. The sulfur deposition due to volcanic emissions in Japan has been shown to be about 20% of the total deposition in that country.

1. Introduction

The last decade has seen a rapid growth in the industrial production in East Asia. The subsequent increase in the energy use has caused a simultaneous increase in the emissions of air pollutants like SO_2 and NO_x in the region (Akimoto et al. 1994), thereby affecting its air quality. The pollutants are not confined to just the urban and industrial centers, but there is a growing awareness and a potential problem associated with their long-range transport and acid deposition. Studies have shown that in Japan, the transport of pollutants from Tokyo Bay to the central mountainous region takes place under proper meteorological conditions (Chang et al. 1989a,b, 1990; Sasaki et al. 1988).

To understand the complex processes governing the deposition of these acidic substances, it is necessary to develop mathematical models that can be simulated under appropriate meteorological conditions to obtain an integrated picture of the state of the atmosphere. Some of the conventional comprehensive large-scale air quality models developed in the 1980s were the Acid Deposition and Oxidant Model (Venkatram et al. 1988), Regional Acid Deposition Model (Chang et al. 1987), and the Sulfur Transport Eulerian Model (Carmichael et al. 1986). Modeling of the long-range transport of acidic substances is just beginning in East Asia (Arndt et al. 1995; Ichikawa et al. 1995; Kotamarthi et al. 1990; Kitada et al. 1992). One of the first efforts has been by the Central Research Institute of Electric Power Industry (CRIEPI) and their Lagrangian model. While their results have been very useful, it is also clear that continued model improvements are needed.

Toward this end, a study was initiated in order to test the Lagrangian and Eulerian frameworks and how added spatial and temporal resolution in mixing, dry deposition, and emission processes affect the results. In this paper, an evaluation was carried out for two long-range transport models for sulfur deposition in East Asia, and the results compared to the data from a network of observation stations in Japan. The evaluation was done between a single-layer Lagrangian model and a multilayer Eulerian model for two-month periods in the summer (August) and winter (February) of 1989. The objective of this study was to examine the similarities and differences in behavior of the two long-range transport models for the same set of meteorology, boundary conditions, and chemical mechanisms.

2. Model description

Two different kinds of transport models were used to investigate the sulfur deposition in East Asia. The models are described below.

a. The CRIEPI model

A Lagrangian backward trajectory-type model was developed by CRIEPI (Ichikawa et al. 1995). The air-

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Corresponding author address: Dr. Mahesh J. Phadnis, CGRER, 220 IATL, University of Iowa, Iowa City, IA 52242.
E-mail: mahesh-phadnis@uiowa.edu

TABLE 1. The main features of the atmospheric transport models employed in the comparison.

	CRIEPI	Eulerian
Type	Lagrangian	Eulerian
Horizontal dispersion	Gaussian distribution	Atmospheric diffusion equation
Vertical dispersion	Uniformly mixed (1000 m)	Multiple layers (400 m each)
Grid size	80 km	~100 km
Temporal interval	Trajectories every 3 h	Transport time step of 15 min

stream in the transport process is calculated on a stereoscopic projection plane. The trajectories of air masses are determined by tracing the wind field spatially. The concentration distribution is assumed normal in the horizontal direction and uniform in the vertical direction. This single-layer model has a fixed mixing height of 1000 m and no diurnal variation. The dry deposition velocities of SO_2 and sulfate are assumed constant with no variation over land and sea. More details about the model formulations can be found in Ichikawa et al. (1995).

b. The Eulerian model

The Eulerian model derived from the STEM-II model (Carmichael et al. 1986) was the other model used in the study. The details about the governing mass conservation equations and the numerical formulations are given in Carmichael et al. (1986). The simulation of the model involves sequential solutions of locally one-dimensional partial differential equations for transport in the horizontal X and Y directions and the vertical Z direction. The solution of the ordinary differential equations governing the chemistry is then solved at all grid points. A summary of the two transport models is provided in Table 1.

c. The chemical mechanism

The models use a simple chemical mechanism for the conversion of SO_2 to sulfate in air and water. The rate constant is a parameterized value of 3% h^{-1} and 1% h^{-1} during summer and winter, respectively. It assumes the existence of in-cloud processes taking place at all instants, and, hence, predicts cloud-phase sulfate all the time. The mechanism also incorporates in-cloud and below-cloud wet scavenging of sulfate and below-cloud wet scavenging of SO_2. A schematic of the chemical mechanism is given in Ichikawa et al. (1995). The values of all the parameters used in the simulation are summarized in Table 2.

For this comparison study, the Eulerian model was constructed using the same chemical and removal formulations as the CRIEPI Lagrangian model.

d. Model domain

The domain in the study was about 63° in longitude from 99° to 162°E and 36° in latitude from 19.63° to 55.54°N, covering Korea, Japan, Taiwan, and parts of China, Russia, and Mongolia. This region has a diverse topography and mixture of rural/urban centers and agricultural/rural regions. The interactions between continental and marine influences play a major role in determining the effects of pollutant production and transport in this region. The dimensions of each grid were 1.125 in the zonal and 1.122 in the meridional direction. In the vertical direction, the resolution is 400 m and the top of the model goes in the midtroposphere to a height of about 8 km. For the comparison with the Lagrangian model, the lower 2 km is taken into account.

e. Input data

1) EMISSIONS

The SO_2 emissions used in this study are from Fujita et al. (1991). Emissions from both natural and anthropogenic sources are included. The area emissions include industrial activity, domestic activity, and trans-

TABLE 2. Parameter values used in the Eulerian simulations.

Parameter	Unit	Value
Conversion rate coefficient of SO_2 sulfate	1 h^{-1}	0.01 for winter 0.03 for summer
Wet deposition rate coefficient of SO_2	1 h^{-1}	0.1*I
Dry deposition rate coefficient of SO_2	1 s^{-1}	3.5 in winter $\times 10^{-6}$ 4.5 in summer $\times 10^{-6}$
Rainout removal rate coefficient of SO_2	1 h^{-1}	0.02
Wet deposition rate coefficient of sulfate	1 h^{-1}	0.1*I
Dry deposition rate coefficient of sulfate	1 s^{-1}	1.0 in winter $\times 10^{-6}$ 1.05 in summer $\times 10^{-6}$
Rainout removal rate coefficient of sulfate	1 h^{-1}	0.02
Intake rate coefficient of cloud sulfate in rain	1 h^{-1}	0.1*I

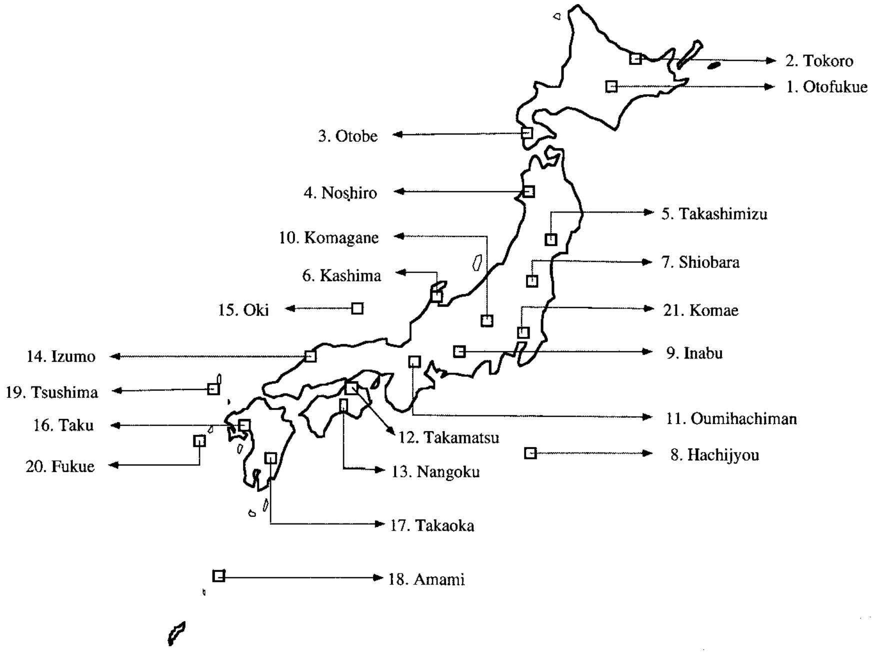


FIG. 1. Location of the observation stations in Japan.

portation. The elevated sources of emissions are mainly large point sources.

In the Eulerian model, the area emissions are allocated at the surface, while the elevated emissions are emitted at 400 and 800 m. Another part of the elevated emissions is the use of the volcanic emissions in Japan (Fujita 1992). The SO_2 emissions from the volcanoes in Japan are estimated to be equal to those from the anthropogenic activities in Japan. These SO_2 emissions are emitted at 1200 and 1600 m in the model. For the simulations with far east Russian emissions, the emission flux for that part of East Asia was interpolated from those given by Galperin et al. (1994).

In the single-layer CRIEPI Lagrangian model, both the elevated and the volcanic emissions are allocated at the surface.

2) METEOROLOGICAL CONDITIONS

The meteorological field inputs for the model were obtained from the Japan Meteorological Agency (JMA). High-altitude wind data was obtained from the Aerological Data of Japan and weather charts. Precipitation data for each day were obtained from JMA weather station data, world data, and the surface Automatic Meteorological Data Acquisition System data. Both models were simulated with the same meteorological data.

For the base-case study, vertical-eddy diffusivities used for the simulation did not vary diurnally and the vertical distribution was for a mixing height of 1000 m. The dry deposition velocities also had no diurnal variation and were constant spatially. In the sensitivity analysis, the simulations were done with diurnal variation in eddy diffusivities and spatial variation in dry deposition velocities.

3. Results and discussion

A nationwide atmospheric monitoring network to survey acidic deposition has been constructed and operated by CRIEPI. The two types of stations in this network were climatologically representative (type A) and background (type B) stations. The selection of type A stations was based on land use, annual precipitation, distance from large sources, local criteria, and maintenance of samplers. The type B stations are located on islands around Japan. Figure 1 shows the locations of the stations in Japan.

The models were first compared using the same parameterization for chemical and removal mechanisms. Then sensitivity studies were done with the Eulerian model exploring how the spatial land temporal resolution of the processes affected the results.

a. Base case simulations

The total sulfur deposited in East Asia, as predicted by the Eulerian model, is presented in Fig. 2. It clearly depicts the influence of SO_2 emissions on the amount of sulfur deposited in that region. Elevated values in excess of 0.1 g S m^{-2} are seen over eastern parts of China, most of South Korea, southern Japan, and some parts of eastern Japan. All these regions are places of high-anthropogenic emissions of SO_2 with the exception of southern Japan, where the volcanic emissions are comparable to the anthropogenic emissions and, hence,

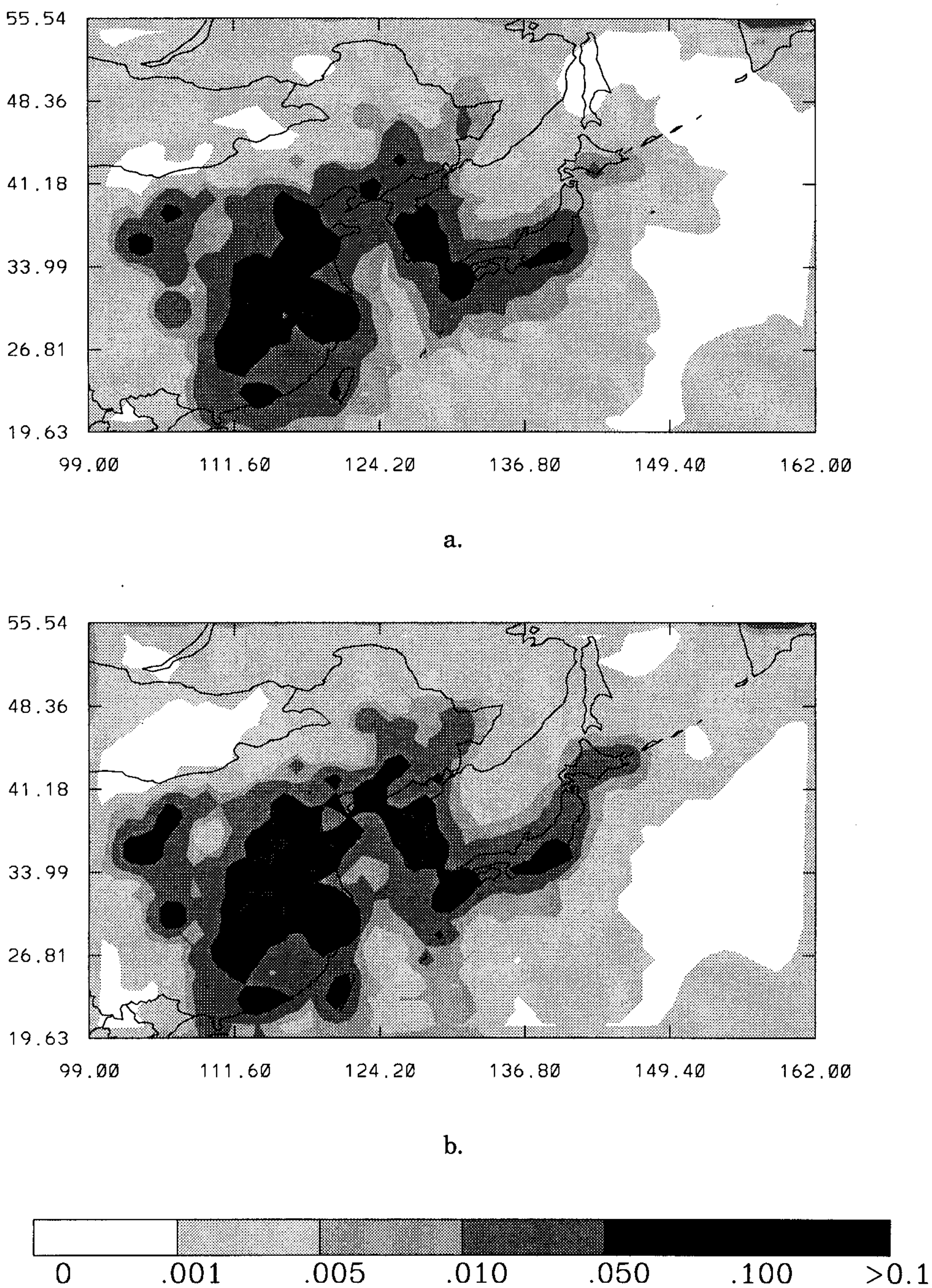


FIG. 2. Total sulfur deposition (g S m^{-2}) predicted by the Eulerian model: (a) February and (b) August 1989.

contribute significantly (which will be discussed later) to the total sulfur deposited there. The figure also shows more deposition in summer (August) than in winter (February) due to the increased production rate of sulfate and more precipitation in summer, thereby increasing the wet scavenging.

The performance of the Eulerian model for the two months was compared to that of the CRIEPI model at the 21 monitoring stations in Japan. The results for SO_2 and sulfate in air and sulfate in water are presented in Fig. 3 for February and August 1989. Both models show the same general trends in temporal and spatial variability. This is a good indication that both modeling approaches using the same meteorological data capture the general transport features. The concentration of SO_2 in air for February for almost all the stations is higher than

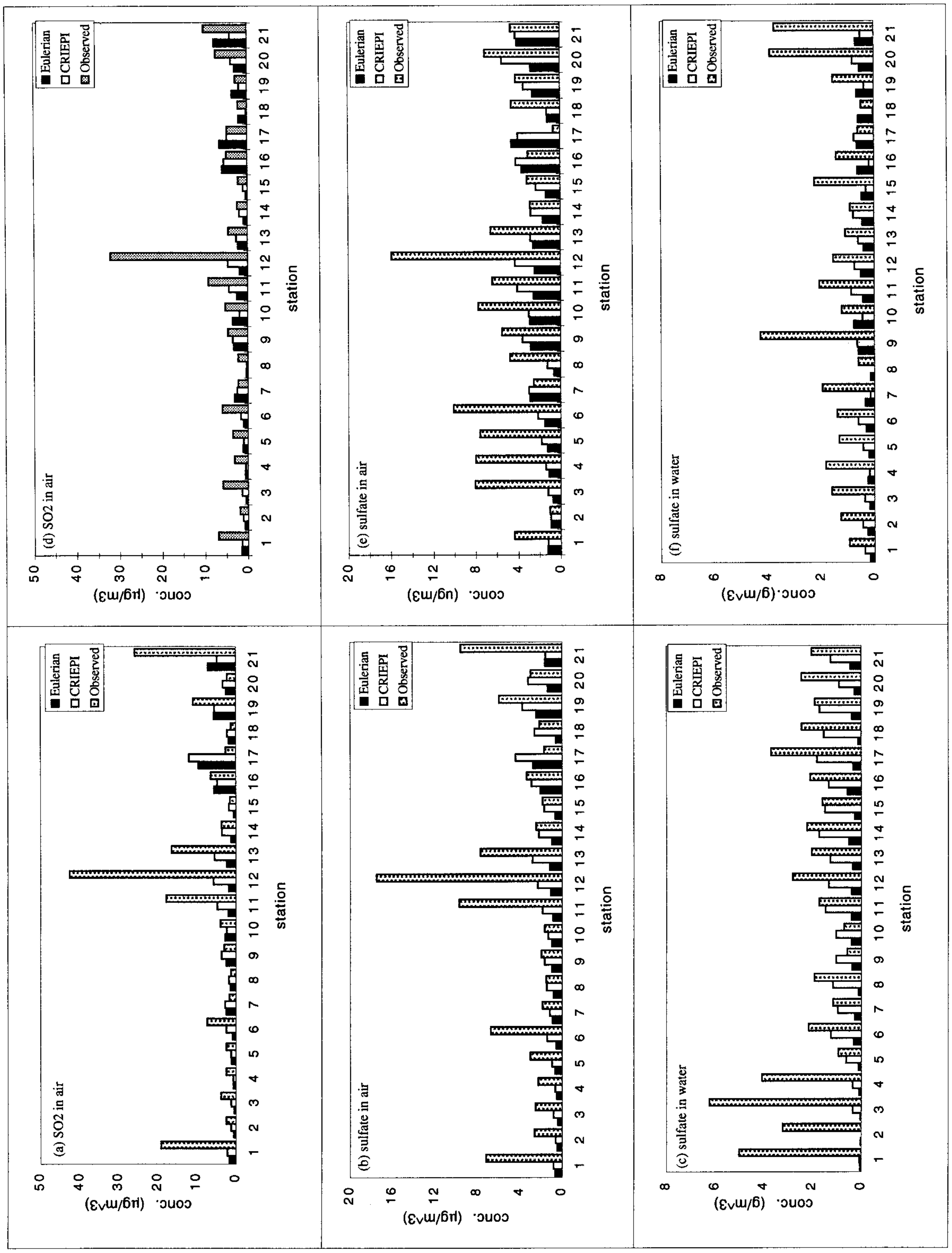


FIG. 3. Comparison among Eulerian, CRIEPI-predicted, and observed surface concentrations: (a), (b), (c) for February 1989 and (d), (e), (f) for August 1989, respectively.

that in summer because of the lower solar intensity and, hence, lower photochemical oxidation rates in winter. The converse is true for the concentrations of sulfate in air. The values predicted by the Eulerian model are, in general, lower than those predicted by the CRIEPI model.

The underprediction of the Eulerian model with respect to the CRIEPI model can be explained on the basis of the presence of multiple layers in the Eulerian model as against a single layer in the CRIEPI model. The pollutants emitted from the surface diffuse vertically through a number of layers in the Eulerian model. In each of these layers, there is uniform mixing of the pollutants. But due to a predominant vertical diffusion, the probability of the pollutants going back to the previous layer is lowered. As against this, in the one-layer CRIEPI model, the pollutants get ‘‘trapped’’ due to uniform mixing in this layer, subsequently increasing their surface concentration.

Both the Eulerian and the Lagrangian models underpredict the observed concentrations of SO_2, sulfate in air, and sulfate in water. The exclusion of an explicit treatment of both the gas and aqueous phases sulfur chemistry and the assumption of constant overall conversion rates might be some factors contributing to the lack of agreement between the predicted and the observed values. Other reasons for the underprediction can be due to the fact that the background concentrations of the species might be too low or that the sulfur emissions inventory is slightly off. Also, the stations might be influenced too much by local sources. For instance, for the island station Oki, the predictions are in fair agreement with the observations. This is not true for some rural stations in northern Japan.

The sulfate in water predicted by the Eulerian model is well below that predicted by the CRIEPI model. The chemistry used for this comparison study, which involves the production of cloud-phase sulfate at all instants is not suited for the Eulerian model. In the chemical mechanism used in the modeling, there is a constant conversion of sulfate in air to cloud-phase sulfate. Hence, at any grid point there exists SO_2, sulfate in air, and sulfate in cloud, which are transported to the next grid by the governing conservation laws. In this framework, a species is removed in water only if there is precipitation over a particular grid point. Consequently, even though there is always a production of sulfate in air and cloud, the wet removal is dominated by the precipitation over a grid. In the simulations with the Eulerian model, the precipitation was localized over a small region of East Asia (and, hence, only a few grid points). Whereas in the Lagrangian model, all of the three sulfur species are transported by the air trajectory so that more wet removal takes place when this trajectory encounters precipitation.

The model predictions are compared to observed values at selected stations in Fig. 4. The observed values are obtained by sampling over a 10-day period. A time-series analysis for SO_2 and sulfate in air and sulfate in water is shown in Fig. 4a for Takamatsu. Both models perform well with respect to one another, but fall short of the observed data. This station being near an industrial center shows very high observed concentrations of SO_2 and sulfate in air, which is not predicted either by the CRIEPI or the Eulerian model. The possible reason for this might be as mentioned previously, the proximity of the observation station to an industrial center.

Figure 4b shows a time-series analysis for the same species as above for the station Taku. The influence of continental outflow in winter, the southwesterly flow in summer, and the possible influence of nearby volcanic emissions characterize Taku as a good station to understand the comparison of model predictions. From the figure, it can be seen that both models predict the transport features of the species in a similar manner. Moreover, the model-predicted values are in reasonable agreement with the observed values.

A time-series analysis at Amami in Fig. 4c, shows that the Eulerian model predicts the surface concentrations better than the CRIEPI model, which tends to overpredict them on some days. This might be true for a remote island like Amami, which is not influenced by anthropogenic or volcanic sources. Therefore, the single-layer model tends to predict more continental outflow than the multiple-layer model.

The discrepancy in the concentrations of sulfate in water can be expected by the reasons discussed earlier about the cloud-phase treatment of sulfate in the Eulerian model. Other reasons could be an incomplete treatment of cloud chemistry for sulfur and also the involvement of interpolation errors in the estimation of the precipitation fields. In the model simulations, the precipitation at the observation stations was interpolated from a sparse areal field of data. The resultant values were quite different from the observed values at those stations. This could account for larger discrepancy with the observations for the liquid concentrations compared to the gas phase.

The Eulerian model was also simulated with just the volcanic emissions from Japan, emitted at altitudes of 1200 and 1600 m. The fraction of the sulfur deposited by volcanoes is shown in Fig. 5. The figure clearly shows the higher sulfur deposition in August due to the reasons discussed before. It can also be seen that Japan experiences about 30%–40% of its total sulfur deposition due to the volcanic activity occurring on its islands. The spatial pattern of the sulfur deposition also shows the influence of winds in depositing more sulfur to the northwest of Kyushu Island in summer and vice versa. In case of the CRIEPI Lagrangian model, the volcanic contribution predicted is about 20%.

A time-series analysis of the surface concentration due to anthropogenic and volcanic emissions can be best compared at a station near the volcanic source. Figure 6 shows such a comparison for Fukue. The figure shows SO_2 and sulfate in air predicted by the Eulerian and the CRIEPI models. At this station, the Eulerian model predicts al-

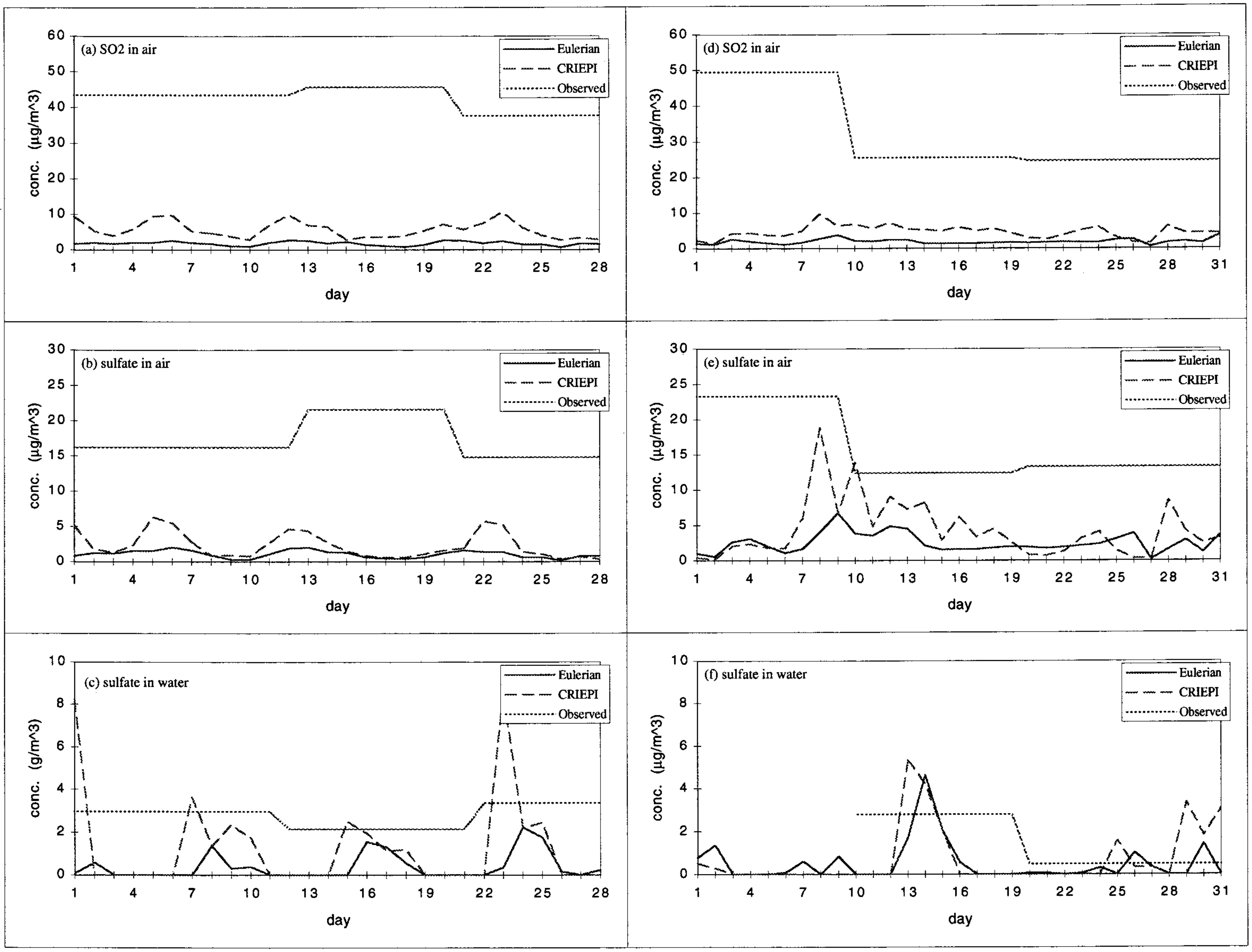


FIG. 4a. Time-series analysis with Eulerian, CRIEPI-predicted, and observed surface concentrations at Takamatsu: (a), (b), (c) for February 1989 and (d), (e), (f) for August 1989, respectively.

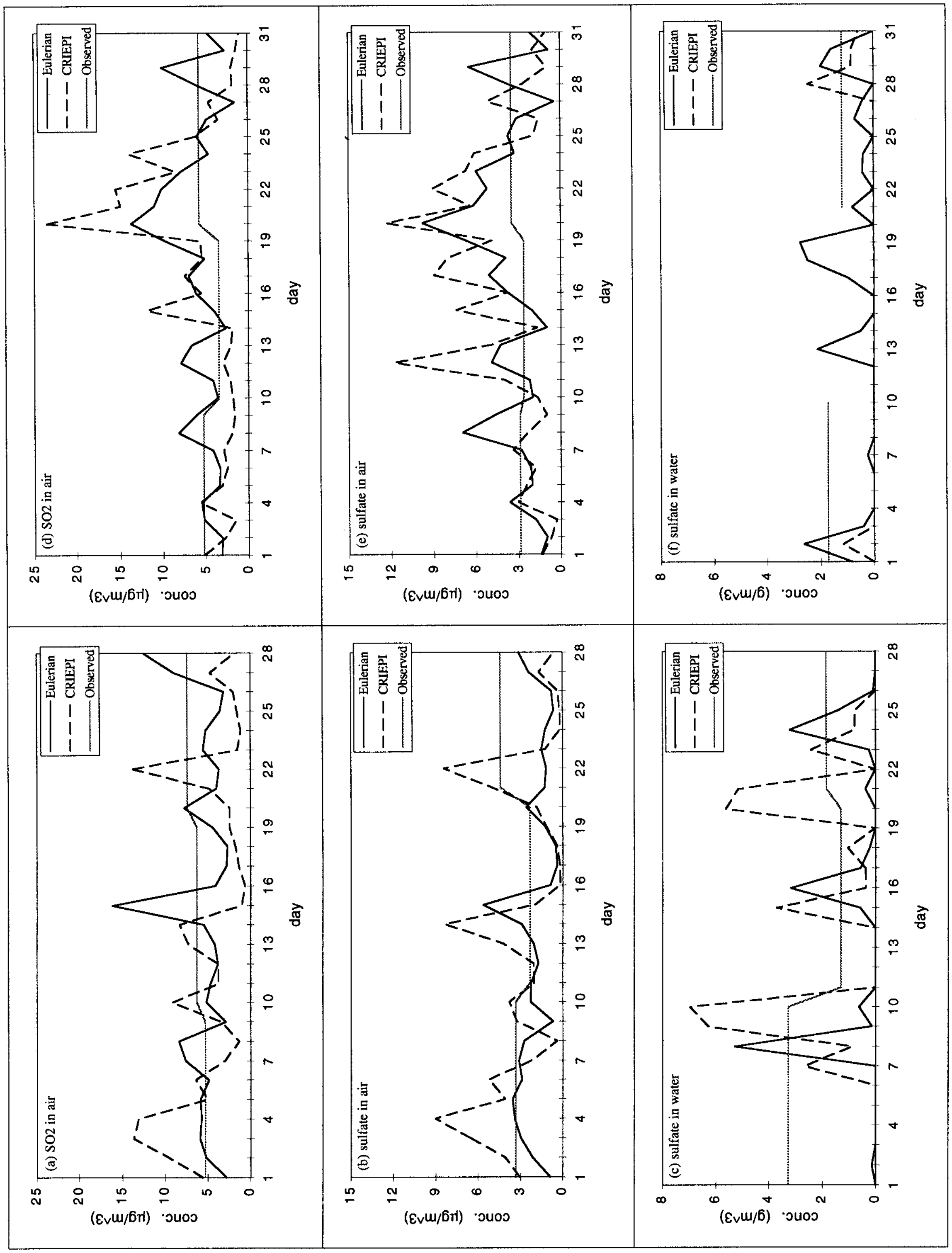


FIG. 4b. Time-series analysis with Eulerian, CRIEPI-predicted, and observed surface concentrations at Taku: (a), (b), (c) for February 1989 and (d), (e), (f) for August 1989, respectively.

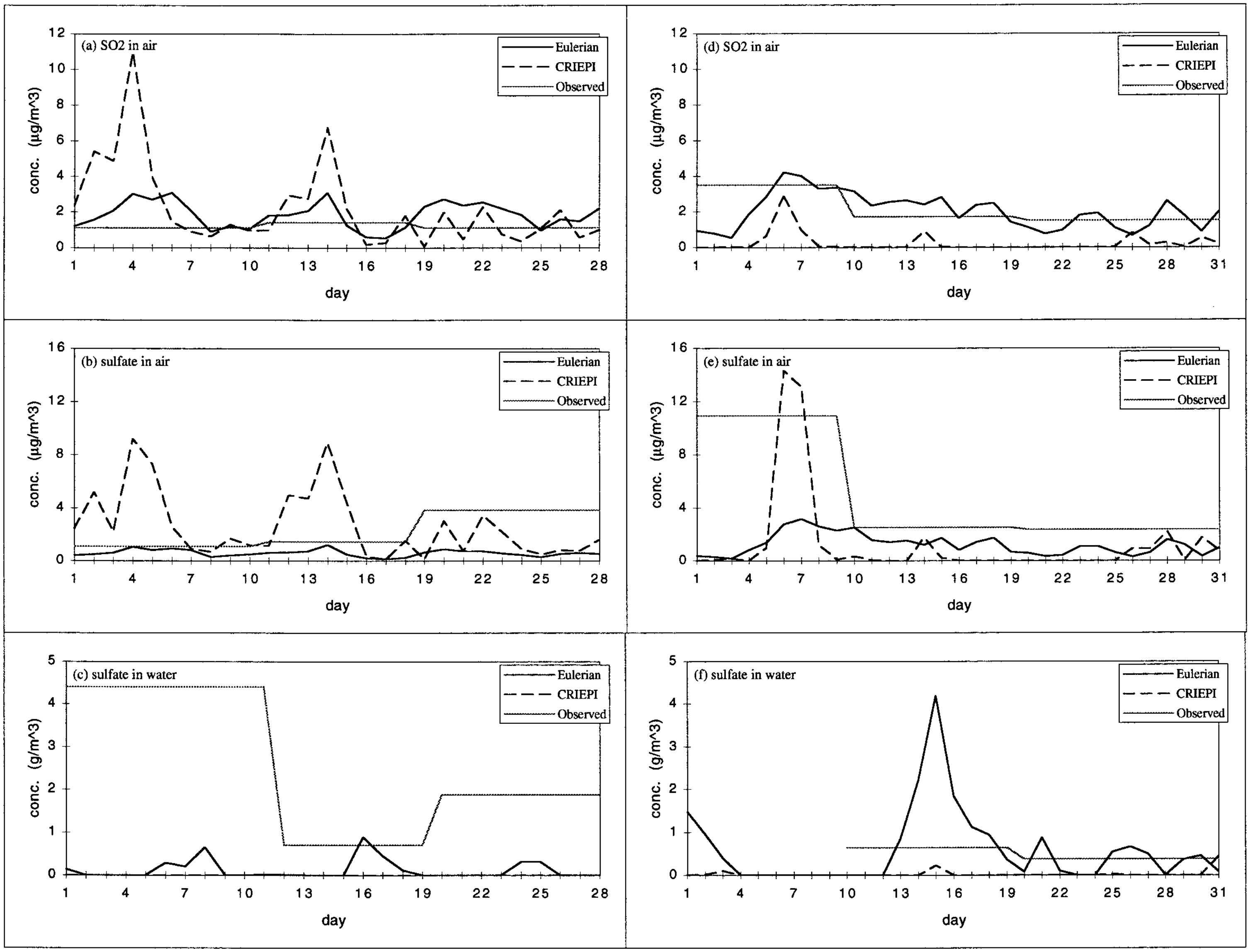
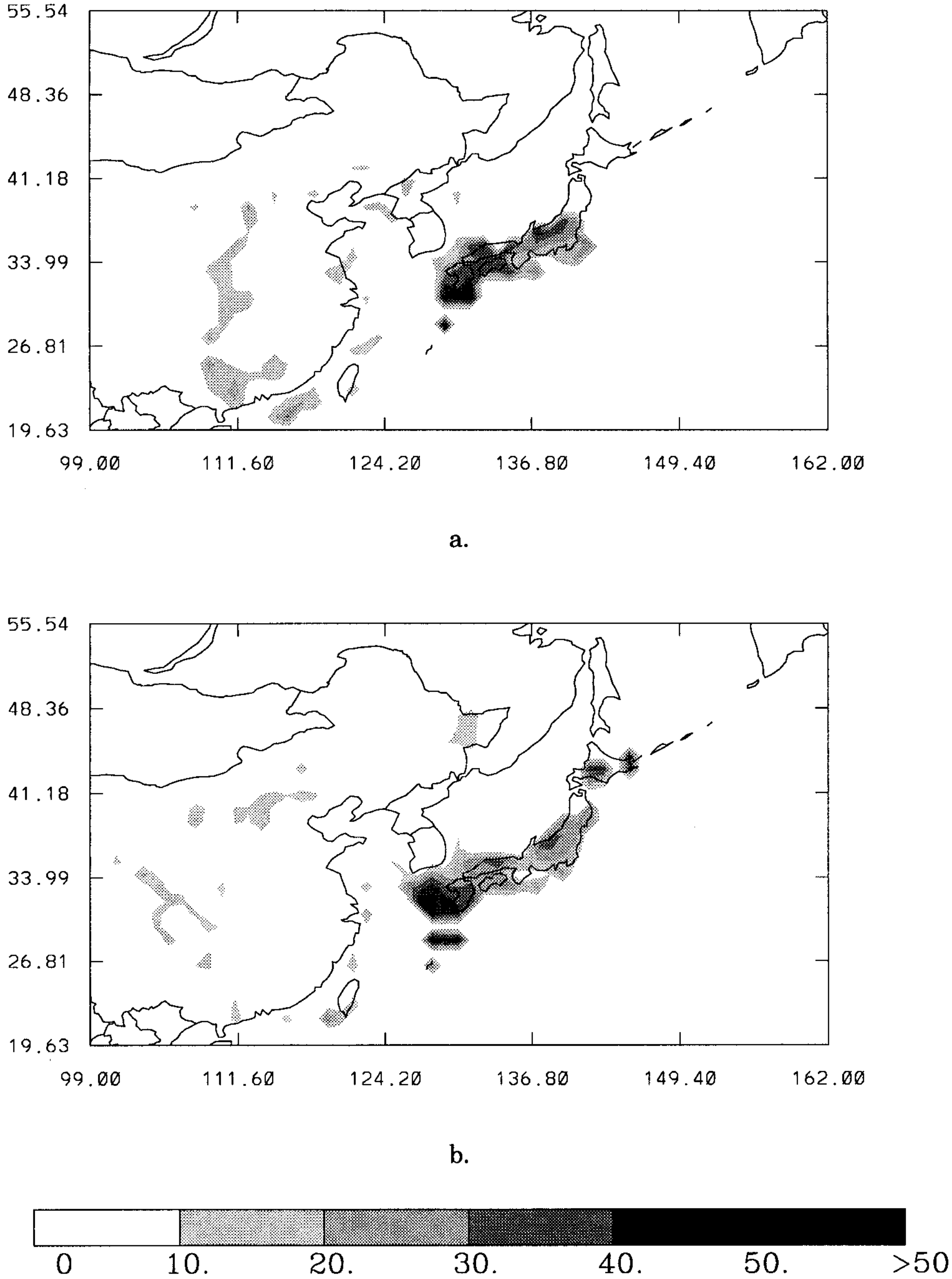


FIG. 4c. Time-series analysis with Eulerian, CRIEPI-predicted, and observed surface concentrations at Amami: (a), (b), (c) for February 1989 and (d), (e), (f) for August 1989, respectively.



a.

b.

FIG. 5. Percentage fraction of sulfur deposited by volcanoes as predicted by the Eulerian model: (a) February and (b) August 1989.

most 25% of the surface concentration for both SO_2 and sulfate due to the volcanic emissions. In February, there is no difference in the predicted values by the CRIEPI model. This might be due to the introduction of both the anthropogenic and the volcanic emissions in the single surface layer and the northwesterly flow in winter that drives the SO_2 and sulfate in air away from the island. In summer there is a reversal in the wind direction that brings the pollutants from the volcano to Fukue. The Eulerian model always predicts volcanic contributions because of the upper-level emissions being spread in all directions and brought down to the surface.

b. *Sensitivity analysis*

The Eulerian model results presented above were for those obtained with a constant eddy-diffusivity profile. An advantage of Eulerian models is that parameters like

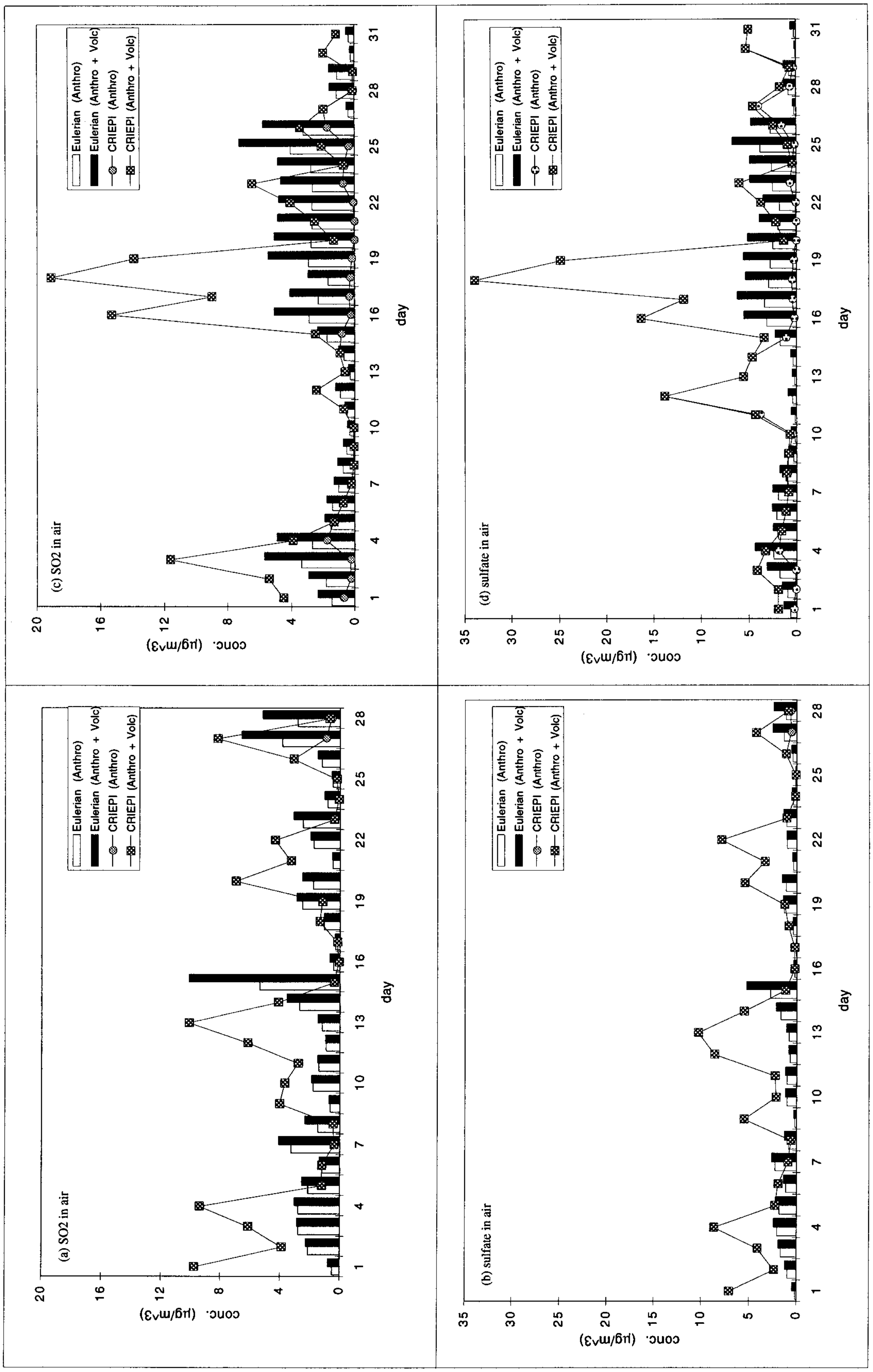
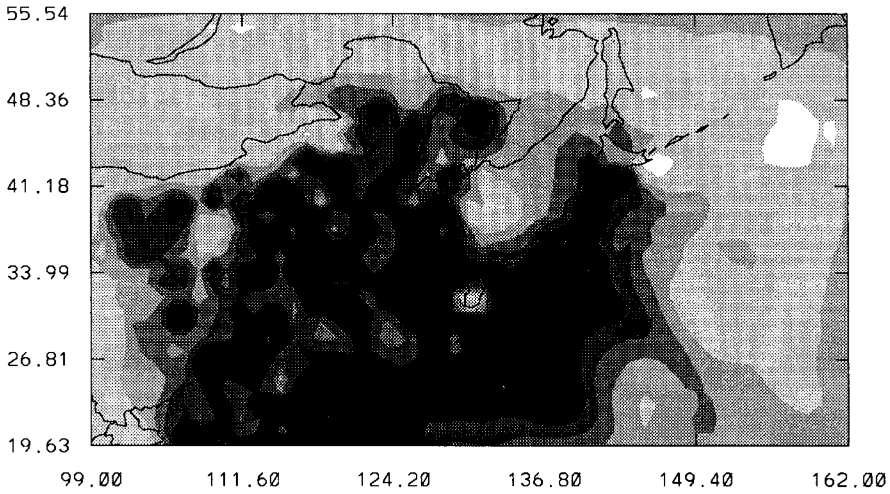
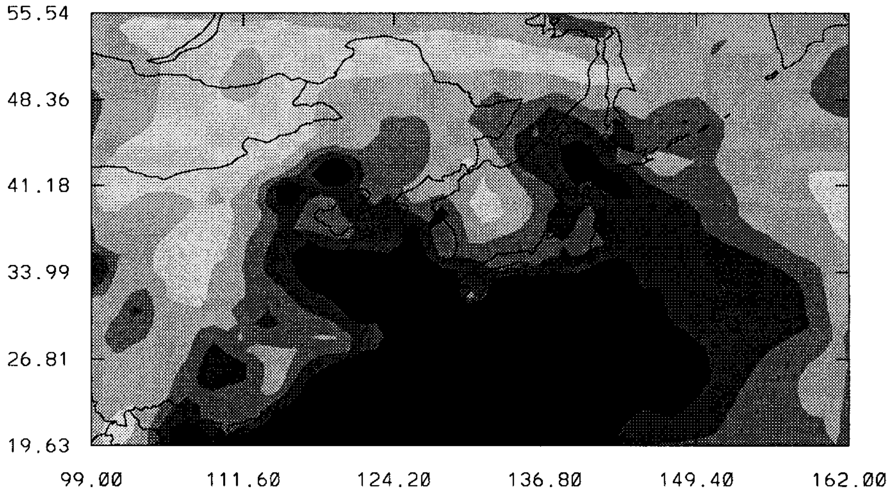


FIG. 6. Eulerian and CRIEPI-predicted surface concentrations due to anthropogenic and volcanic emissions at Fukue: (a), (b), (c) for February 1989 and (d), (e), (f) for August 1989, respectively.

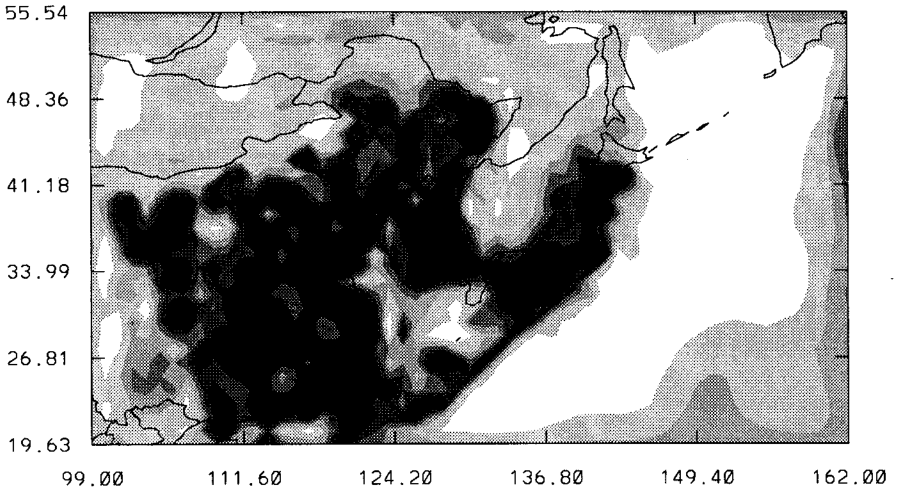


a. SO2 in air

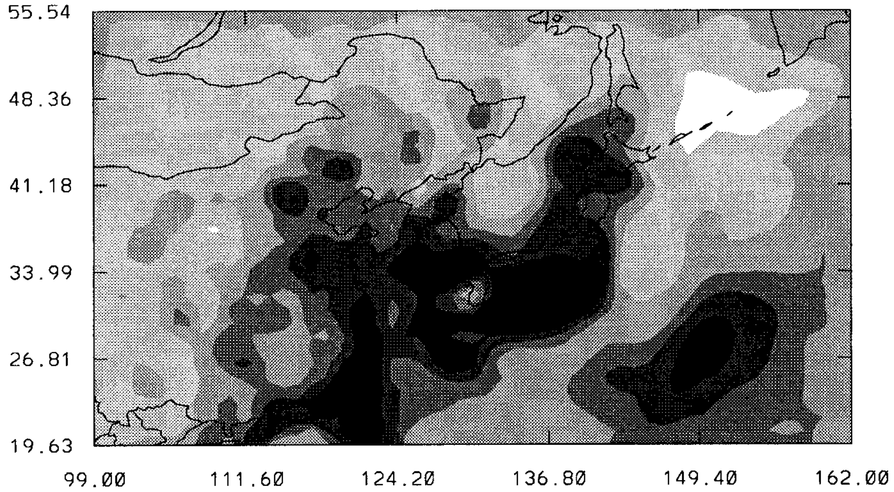


b. sulfate in air

FIG. 7. Percent change in the surface concentrations (February 1989) due to variations in the eddy diffusivities. The values shown are the changes over the base case, which in these cases is the one with no diurnal variation in the mixing heights, while the test case represents the diurnal and spatial change.



a. SO2 in air



b. sulfate in air

FIG. 8. Percent change in the surface concentrations (August 1989) due to variations in the eddy diffusivities. The values shown are the changes over the base case, which in these cases is the one with no diurnal variation in the mixing heights, while the test case represents the diurnal and spatial change.

eddy diffusivities can vary spatially and temporally. The Eulerian model was also run using diurnal variation in the eddy diffusivities.

Figures 7 and 8 show the effect of changes in the mixing height (and the eddy diffusivities in the Eulerian model) on the surface concentrations in East Asia for February and August 1989, respectively. In general, it can be seen from the figures that the change from con-

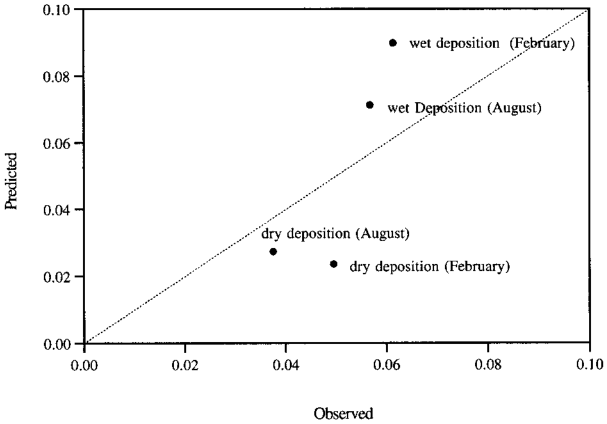


FIG. 9. Predicted and observed average sulfur deposition in Japan (g S m^{-2} per month).

stant to diurnally varying eddy diffusivities results in increased concentrations over land and decreased concentrations over sea for a primary pollutant like SO$_2$. Over vast regions in Japan, the surface concentration increased by 30%–40%. This is because of the lower average vertical diffusivities for a diurnally varying case than that over the constant case. For sulfate in air, the effects are spread over a larger region. The secondary nature of sulfate and its longer lifetime allows it to be advected over large distances. The influence of the northwesterly flow of winds in the winter is clearly depicted in Fig. 7. The increases are less pronounced in August because of the flow of cleaner maritime air onto the continent with simultaneous increase in precipitation, thereby reducing the sulfate concentration in air.

The average sulfur deposition calculated from the network of stations across Japan is shown in Fig. 9. The values shown are the ones obtained from the simulation where there were diurnal variations in the eddy diffusivities and spatial variations in the dry-deposition velocities. The observed dry deposition was calculated from the observed concentrations and the dry-deposition velocities used in the model simulation. One basic problem when comparing wet removal is the precipitation amount. The analyzed precipitation fields systematically yielded lower precipitation than that recorded at the measurement sites. The precipitation field was then reanalyzed to align the precipitation amount with the monitoring sites. When this was done, the predicted sulfate in rain was in good agreement with the observations.

From the figure, it can be seen that the predicted wet deposition of sulfur is in fair agreement with the observed values. The average values are 0.09 and 0.07 g S m^{-2} for February and August, respectively. The predicted dry deposition of sulfur is also in good agreement with the observed values. The sulfur deposition in Japan during the two months of analysis is dominated by wet deposition at most of the stations. At the northern stations like Otofuke and Tokoro where there is less precipitation, the wet deposition is very low ($\sim 3 \times 10^{-4}$ g S m^{-2} month^{-1}) and the major contribution to the total sulfur deposition is due to dry removal ($\sim 3 \times 10^{-2}$ g S m^{-2} month^{-1}).

To further investigate the behavior of the Eulerian model under the circumstances of diurnal variation in eddy diffusivities, the surface concentrations of SO$_2$ in air, sulfate in air, and sulfate in water are compared to the observed concentrations in Japan. The results are presented in Fig. 10. As seen from the figure, the Eulerian model in these conditions behaves well and the predicted concentrations of all the three species for both February and August are comparable to the observed concentrations. The Eulerian model tends to underpredict at Takamatsu, which might be attributed to the presence of the observation station near a pollutant source. The comparison for sulfate in water is also good at almost all the stations, but there is underprediction at the northern stations in Japan. This is because of low rainfall in northern Japan, the estimation of model-predicted precipitation and sulfate production in liquid phase and other reasons mentioned earlier in section 3b.

4. Summary

A numerical evaluation of two transport models was done to study the deposition of sulfur in East Asia. A single-layer Lagrangian model (CRIEPI model) was compared to a multiple-layer Eulerian model for two episodes of winter and summer.

From the comparison of the two models, it can be concluded that the use of multiple layers in a model tends to decrease the surface concentrations of the pollutants. This is because of the vertical diffusion of the pollutants and their subsequent transport in the different layers of the multiple-layer model. The overall transport features of the two models were similar in nature, showing the highs and the lows on appropriate days, thereby confirming the existence of similar meteorological advective processes in the two models.

The Eulerian model underpredicted the observed data at most of the monitoring stations in Japan when a constant eddy diffusivity and the CRIEPI chemistry are used. This could be because of the horizontal scale of the Eulerian model being too large in comparison to the location of the observation station. Hence, several subgrid processes could come into play, hereby affecting the true predictions (or the true grid-averaged observed values). Another factor affecting the performance of the Eulerian model, which was discussed in section 3a, is the assumption of the existence of in-cloud sulfate even in the absence of clouds. This assumption, though practical for a Lagrangian model, can, for a Eulerian model, affect the concentration of sulfate in precipitation, thereby reducing the predicted wet deposition. In general, the underpredictions in the model-calculated concentrations can be attributed to the assumption of the fixed-rate constants for the conversion of SO$_2$ to sulfate and

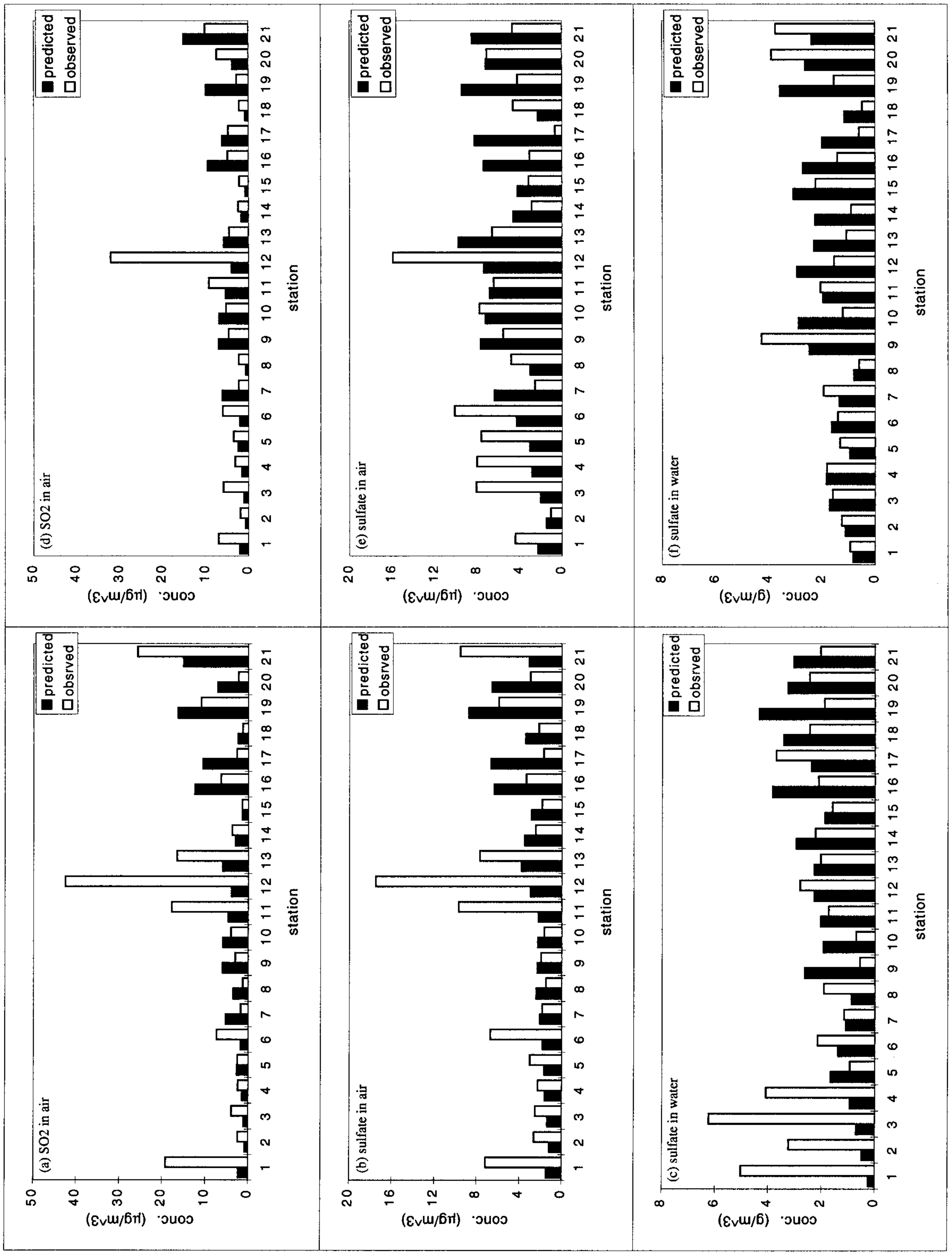


FIG. 10. Eulerian-predicted and observed surface concentrations in Japan: (a), (b), (c) for February 1989 and (d), (e), (f) for August 1989, respectively.

also to the exclusion of explicit sulfur chemistry in the model.

The predictions from the simulations with just the volcanic emissions at higher altitudes showed that volcanoes can contribute from 30%–40% of the local sulfur deposition throughout Japan. The simulations also showed that one advantage of using a multiple-layer model against a single layer is that the elevated emissions can be injected in different levels so that a more realistic profile of the vertical diffusion and horizontal transport of the upper-level emissions can be obtained.

These results indicate the need to use multiple layers in the modeling of long-range transport (LRT) in East Asia. This is required because of the presence of many elevated sources, including volcanoes, and the sensitivity of surface concentration to diurnal variation in the eddy diffusivities. This can be accomplished by use of either Eulerian or multilayer Lagrangian models or a hybrid approach (Hayami et al. 1995).

Another aspect is the estimation of wet deposition. It is important to use the observed precipitation and to modify the analyzed precipitation fields in a country like Japan where there are small-scale features of precipitation.

Finally, while the modeling of LRT of pollutants in Asia is in the early stages, it is an essential tool in the air-quality management and policy making in the region. Improved models will be needed that include better parameterization of the atmospheric processes in the region. A simultaneous increase in the network of observation stations across the region is also required both to aid in the development of the improved models and to evaluate the various models.

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