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Numerical Analysis of Wintertime Air Pollution in East Asia Region Using Long-Range Transport Model

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In order to understand the wintertime intermittent characteristics of the trans-boundary air pollutant transport observed in East Asia, a numerical simulation of the long-range transport of pollutants was applied using an atmospheric transport model(STEM-II). The numerical simulation was carried out for the entire month of January 1997 and specific atmospheric aerosol (including sulfate, nitrate, and other ion compounds) observation data were compared from four observation sites(Cheju Island, Kanghwa Island, Dazaifu, and Fukue Island). The observation data revealed that concentration peaks were intermittently observed at 3 to 4-day intervals plus the four observation sites exhibited a very similar spatial variation. The horizontal and spatial scale of the heavily polluted air masses was analyzed based on numerical results. The mechanism of the intermittent transport of air pollutants was clearly explained by a comparison of the observed data with the numerical output. It was found that the wind pattern variations associated with the synoptic scale pressure system changes play an extremely important role in the transport of pollutants in this region.

Key words : long-range transport model, sulfate, nitrate, synoptic scale pressure system.

1. Introduction

Trans-boundary air pollution has recently been highlighted as a major problem due to the considerable level of air pollutants emitted due to industrial and anthropogenic activities in the East Asia region. In addition, East Asian countries have now begun to expand their monitoring activities due to the serious threat of regional and global atmospheric pollution including acid deposition. As a result, a number of numerical modeling studies have been performed on the long-range transport of air pollutants in East Asia. The first study in East Asia was reported by Kotamarthi and Carmichael[4]. They applied a two-dimensional version of STEM(Sulfate Transport Eulerian Model). More recently, Uno *et al.*[11] performed a three-dimensional model simulation to understand the long-range transport phenomena associated with the synoptic scale pressure system in East Asia. Park and Cho[6] proposed an air pollutant transport index to identify the influence of the source region on the receptor regions in Korea and East China.

This paper investigates the characteristics of trans-boundary air pollutant(mainly sulfate) transport in the wintertime over East Asia using a long-range transport model which includes detailed chemical reactions, STEM-II(Carmichael *et al.*[3]), during the entire month of January 1997. The simulated sulfate concentrations are compared with data from four observation sites(Cheju Island, Kanghwa Island, Dazaifu, and Fukue Island). In addition, the association of the transport mechanism with the synoptic pressure system during the occurrence of intermittent peaks in the sulfate concentration is discussed along with the characteristics of the horizontal and spatial scales of trans-boundary air pollution over East Asia in wintertime.

2. Numerical Model(STEM-II) and Model Domain

To understand the observed characteristics related to the long-range transport of pollutants, an atmospheric transport model including detailed chemical reactions, STEM-II(Carmichael *et al.*[3]), was applied. STEM-II is a three-dimensional Eulerian numerical model which accounts for the transport, chemical conversion, and deposition of atmospheric pollutants. The major features of STEM-II include the emission of pollutants from point and area sources, and transport by advection, convection, and turbulent diffusion.

The gas phase reaction mechanism proposed by Lurmann *et al.*[5] was also used. This scheme contains 53 species and 112 reactions. The present version of STEM-II also includes additional biogenic volatile organic carbon reactions. The model includes a detailed NH_3 - NO_3 - NH_4NO_3 thermo-dynamical equilibrium mechanism (Stelson and Seinfeld[9]; Chang *et al.*) plus the heterogeneous reaction from SO_2(gas) to aerosol sulfate(SO_4^{2-}). The model chemistry also assumes that the formation of$(NH_4)_2SO_4$ occurs simultaneously after the generation of aerosol sulfate. The experiments were conducted using a heterogeneous reaction rate ($SO_2 \longrightarrow SO_4^{2-}$) of 1.0%/h.

STEM-II uses chemical, dynamic, and thermodynamic parameterization, which allows for a wide variety of applications such as meso-, regional-, and global scale air pollution studies. The code is modular in structure and can be driven by observed or modeled meteorological data. The dry deposition model proposed by Wesely[12] was also used in the current study.

A detailed description of STEM-II can be found in Carmichael *et al.*[3] and its recent application to East Asia can be found in Uno *et al.* [10),11] and Park and Cho[6]. In the present study, no precipitation or cloud processes were considered.

The East Asia model domain uses a 1° latitude x 1° longitude grid system. The model domain covers from 110° E to 150° E and 20° N to 50° N. This horizontal domain is then divided into a 41 x 31 mesh. The vertical model domain consists of 10 layers from ground level to 10,000 m. The vertical grid levels are 200, 400, 700, 1000, 2000,3000, 4000, 5000, 7000, and 10000 m.

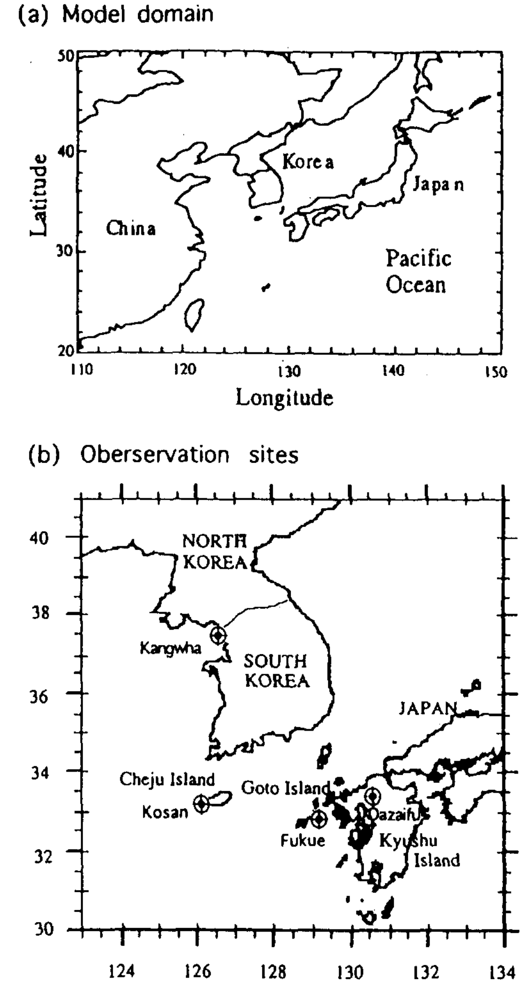

Fig. 1. Model domain and field measurement sites.

The model uses a terrain-following coordinate system. Figure 1 shows the model domain and field measurement sites. The 1° latitude x 1° longitude gridded NOx and SO_2 emission inventory, as reported by Akimoto and Narita[1], was used. Piccot *et al.*[7] also estimated global VOC (Volatile Organic Carbon) emissions due to anthropogenic activities on a grid scale of 10° longitude x 10° latitude. Their data was used to prepare a regional scale VOC emissions inventory for the 1° x 1° grid domain shown in Figure 1. These 1° x 1° estimates were obtained by breaking down the larger scale emissions using population density(on a 1/3 degree scale) as the weighting factor. The VOC category classes were reclassified into the chemical reaction mechanism scheme of

Lurmann *et al.*[5]. NOx and VOC were emitted into the 1st and 2nd vertical levels, while SOx was emitted into the 1st thru 3rd levels.

All meteorological data sets were obtained from the ECMWF(European Center for Medium-Range Weather Forecasts). The ECMWF data consisted of 12-hour intervals of 2.5° x 2.5° spatially grided meteorological data points(wind speed and direction, temperature, relative humidity, geopotential height) at specified pressure levels. The 12-hour interval ECMWF data was interpolated linearly in time and space to generate an hourly interval meteorological data set. The PBL(Planetary Boundary Layer) over the ocean was diagnosed from SST data and ECMWF data.

3. Summary of Field Observation

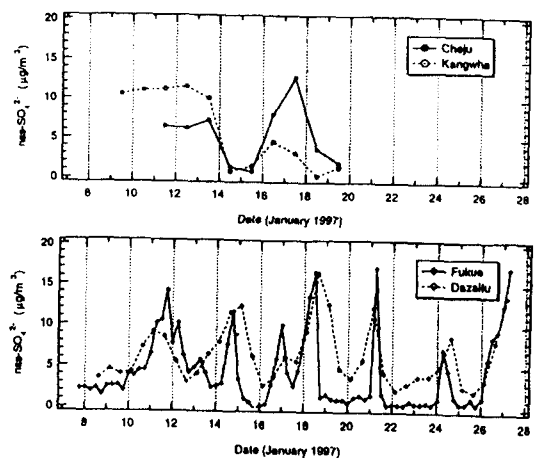

Fig. 2. Diurnal variations of observed nss-SO_4^{2-} concentration at four observation sites.

The field observation details and results of a basic analysis of the observed aerosol data have already been reported by Shimohara *et al*[8], therefore, only a brief outline is given here. The special atmospheric aerosol(including sulfate, nitrate, and other ion compounds) was collected at four locations in Japan and Korea; two Japanese sites(Dazaifu in Fukuoka prefecture, 32.6°N and 128.65°E, and Fukue Island in Nagasaki prefecture, 33.5°N and 130.50°E) and two Korean sites(Cheju Island, 33.3°N and 126.5°E and Kanghwa Island, 37.6°N and 126.4°E). The observation sites are shown in Figure 1. The horizontal distance between Dazaifu and Fukue Island is about 200 km, whereas that between Cheju Island and Fukue Island is about 250km. Fukue Island has no serious anthropogenic emission sources. Dazaifu is located just south of Fukuoka city and has relatively clean air. The two observation sites of Cheju Island and Kangwha Island in Korea are located at places with relatively clear conditions. Figure 2 shows the diurnal variations in the nss-SO_4^{2-} concentrations observed at the four observation sites during the winter monsoon season. The observations were taken from January 8 to 27, 1997 at Dazaifu and Fukue Island (6-12 hour averaged concentrations were monitored at both sites), while the daily average aerosol concentrations at the Korean sites were observed from January 9 to 19, 1997. The observed data from both sites in Japan revealed that concentration peaks were intermittently observed at 3- to 4-day intervals and both observation sites exhibited a very similar spatial variation. These patterns were also observed on the same days at both Korean sites. The characteristics of the observed intermittent sulfate concentration peaks will be discussed in the next section along with the results of the numerical simulation.

4. Results and Discussion

4.1. Comparison Between Observation and NuMerical Results

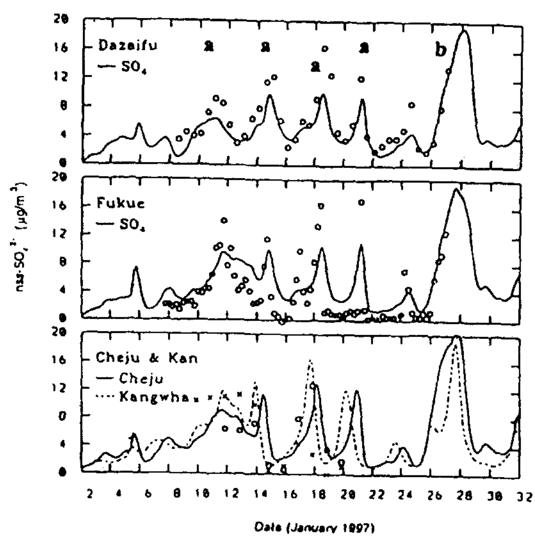

Fig. 3. Comparison of observed data and model simulation results at Kanghwa Island, Cheju Island, Fukue Island and Dazaifu.

Figure 3 shows the diurnal variations of the observed data(nss-SO_4^{2-}) along with the model simulation results at the four observation sites, the solid and dotted lines indicate the model results whereas the open circles show the observation results. The peak aerosol sulfate concentrations were observed on the 11th, 14th, 18th, 21st, and 28th of January at Fukue Island and Dazaifu in Japan. These are marked by "a"(small peak) and "b"(large peak) in figure 3.

The calculated data of both Japanese sites revealed intermittent peak concentrations at 3 to 4-day intervals and both observation sites had a very similar spatial variation. These peak concentrations were also observed on the same days at both Korean sites. Furthermore, the observed sulfate behavior corresponded very well with the calculated sulfate peaks at each site. These observational facts indicate that the horizontal scale of a high sulfate air mass must be larger than 200km(covering at least the distance between Fukue and Dazaifu), and it also shows that the intermittent transportation of a heavily polluted air mass is dependent on the wind pattern variations associated with the synoptic weather system changes during the wintertime. As a result, each observation site was covered, in turn, with the transported polluted air mass.

4.2. Synoptic-scale Weather System and Outbreak of Air Pollutants

The relation between the synoptic-scale weather system and the outbreak of pollutants is represented well in Figure 4. It was also found that wind pattern variations associated with the synoptic scale pressure system are extremely important for the transport of pollutants. The low pressure system and meso-front line movements clearly indicate the transport of pollutants from the Asian mainland to the Yellow Sea and in the direction of the western part of Japan, which occurs after the passage of a low pressure system and meso-front line. This fact indicates that a change in the synoptic-scale pressure system plays an important role in long-range transport in the East Asia region. Accordingly, the outbreak of air pollutants in the Asian region during the winter season can be explained by these two reasons. Furthermore, when a typical winter synoptic scale pressure system was formed, the pressure gradient in the north-south direction is also strong, therefore, an polluted air mass occurring on the Asian main land will seldom transport to the east horizontally.

4.3. Intermittent Characteristics of Trans-boundary Air Pollution

Simulated sulfate concentration distributions including wind fields for 3-day periods containing episode days(16th-18th, 26th-28th) and their synoptic weather conditions were described to analyze the intermittent characteristics of the trans-boundary air pollution. The long-range transport processes of air pollutants could be classified into two types, denoted as the stretching cone type (Figure 4) and the giant puff type(Figure 5).

4.3.1. Stretching Cone Type

Figure 4 shows the process by which the air pollutants were transported from the Asian mainland in a northeastward direction initially in a cone-like shape. The middle of the low-pressure system was located to the north-east side in this model region. The sulfate concentration at 200m on the ground was in excess of 16ug/m^3 in the southern main land area of the model region. The next day(17th of January), the low-pressure system developed a meso-front line moving southeastward in the Asian region. At that time, the polluted air mass was like a stretched cone shape which was driven progressively southeastward behind the moving low-pressure system. Accordingly, the horizontal distribution of the sulfate concentration in Figure 4 could be easily accounted for by the typical wind direction of the low-pressure system in the vicinity during the episode days. The last day of the episode(January 18th), the low-pressure system moved in a more southeastward direction and the meso-front line was located on the Pacific side of Japan. Therefore, the polluted air mass, which started on the main land, had travelled together with the low-pressure system, thereby explaining the peak sulfate concentrations detected at the four observation sites in figure 1. Thereafter a new episode began on January 19th with the repeat occurrence of synoptic weather conditions which is the typical winter weather pattern in the Asian region.

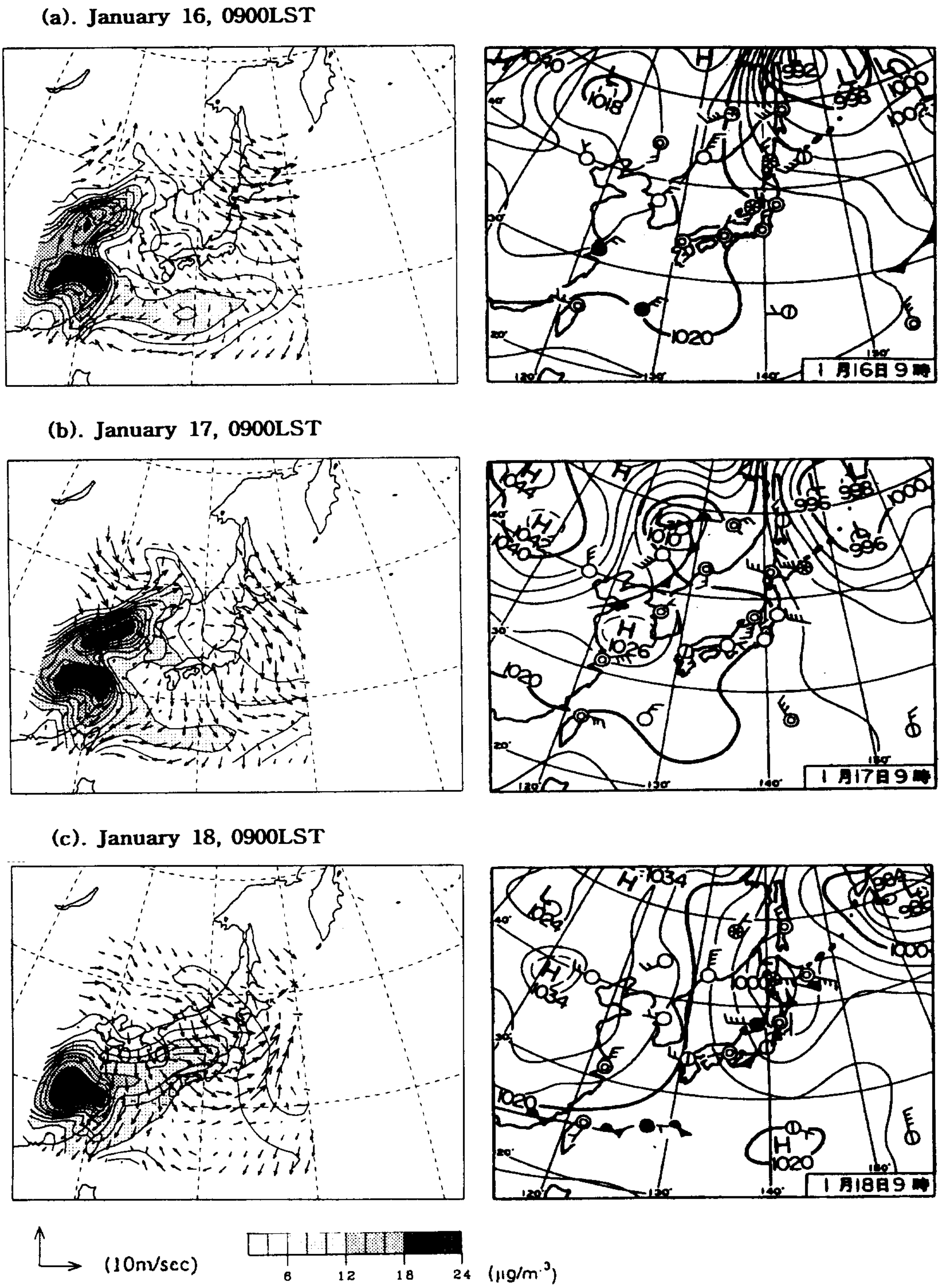

Fig. 4. Simulated sulfate concentration distributions(contour interval is 2ug/m^3, z=200m) with wind fields and synoptic weather conditions at that time from January 16th to 18th, 1997 0900LST(Stretching cone type).

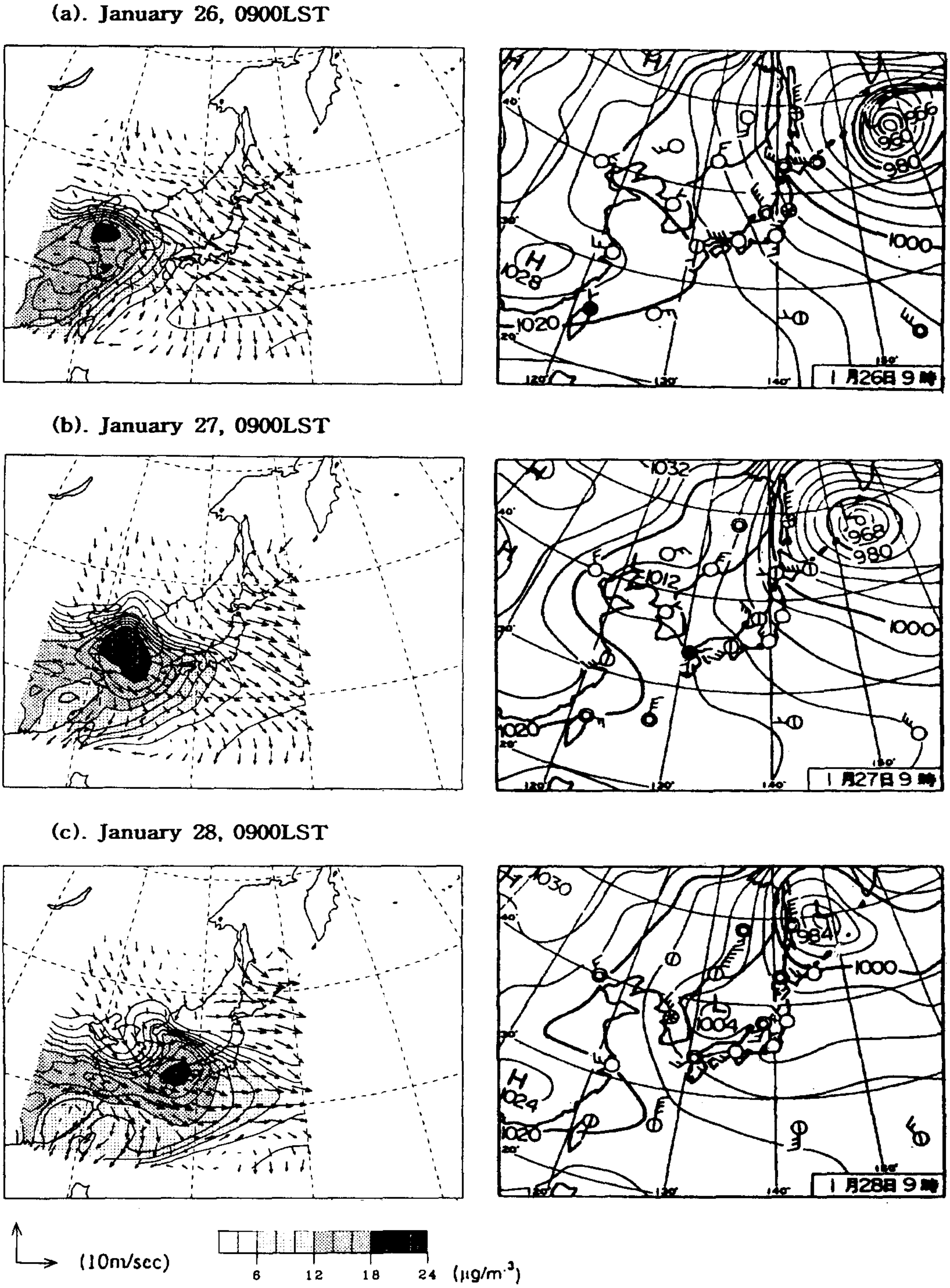

Fig. 5. Simulated sulfate concentration distributions(contour interval is 2ug/m^3, z=200m) with wind fields and synoptic weather conditions at that time from January 26th to 28th, 1997 0900LST(Giant puff type).

The small intermittent peaks(Figure 3, "a") can be explained by the stretching-cone type. In addition, it was found that a gradual time lag occurred from Kanghwa to Cheju Island, Fukue Island, and Dazaifu. This indicates long-range transport from the west side of the Asian mainland in the model region.

4.3.2. Giant Puff Type

When the high-pressure system was located in the southern part of mainland China during the winter monsoon season, the polluted air mass was transported from China to the Korean peninsular and Japan horizontally like a giant puff(Figure 5). On January 26th, the polluted air mass(above 16ug/m^3) located on the Asian mainland moved in a clockwise direction under a high-pressure system which was located on the south-west side of the mainland. The next day, the main polluted air mass continued to be transported in an eastward direction. On January 28th, the huge polluted air mass arrived in Korea, Kyushu, and the western part of Japan which were all affected by the high-pressure system from the south-west side of the mainland. The large peaks(Figure 3, "b") in the sulfate concentrations were observed when this puff-type transport occurred. The huge polluted air mass was formed under the control of a high-pressure system on the Asian mainland and then transported eastward. High concentrations were revealed almost simultaneously at each observation site(Figure 3). Accordingly, when compared with the cone-type transport, the sulfate concentration peaks were higher with a puff-type transport because the moving length of the polluted air mass was shorter.

5. Conclusions

An atmospheric transport model which included detailed chemical reactions, STEM-II, was carried out for the entire month of January 1997. The model successfully simulated the time variations of the observed sulfate concentrations at four sites (Kanghwa Island, Cheju Island, Fukue Island, Dazaifu). As a result, the horizontal and spatial scale of the heavily polluted air masses were analyzed based on numerical results, and the mechanism of the observed intermittent transport of air pollutants was clearly explained based on a comparison of the observed data with the numerical output. The transport of air pollutants emitted on the Asian mainland to the eastern region was found to be associated with synoptic scale weather conditions. Accordingly, a polluted air mass will travel with a low pressure system and the meso-front line movements pass by the Yellow Sea, Korea, and Japan in a cone-like shape. The polluted airmass is then driven southeastward progressively on the backside of a moving low pressure system. Otherwise, the polluted air mass moves directly to the eastern part of Asia under the influence of a continental high pressure system from the southern part of the mainland. But when the pressure gradient of the north-south direction is strong, the polluted air masses from the Asian mainland were seldom transported to the eastern region. This fact indicates that changes in the synoptic scale pressure system play an important role in the long-range transport in the East Asia region. In further research, an analysis of the vertical distribution of the air pollutant concentrations and the amounts of inflow and outflow fluxes will be performed to better understand the long-range transport mechanism of air pollutants in this region

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