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ACCURACY OF TRAJECTORY CALCULATIONS FOR LARGE-SCALE AIR POLLUTION IN EAST ASIA

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Abstract

Backward trajectory calculations have been used to understand the behavior of air pollutants and the results of chemical measurements. However the accuracy of trajectory calculations is not well known, especially for long-range transport in East Asia. In this study, the results of recent chemical measurements are used to determine the adequacy of conventional isentropic trajectory models. In the absence of observational data which indicate "true" trajectories, we have compared locations calculated by a conventional trajectory technique with those calculated by a three-dimensional trajectory technique using meteorological fields with high temporal and spatial resolution. Isentropic trajectories arriving at vertical levels close to the top of the boundary layer sometimes differ greatly from three-dimensional trajectories arriving at the surface measurement points. The differences are caused by differences in starting heights of computation and by differences in the meteorological data. Although a single trajectory does not necessarily reflect the actual path of motion of pollutants, the calculations suggest that the ensemble mode of trajectory analysis provides a useful tool for analyzing flow patterns at observatories and interpretation of surface chemical measurements.

KEYWORDS: *Trajectory analysis, Isentropic trajectory, East Asia, Long-range transport*

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1. Introduction

Increased attention to regional- and global-scale air pollution has resulted in numerous air quality monitoring stations and field studies to understand the behavior of air pollutants. However, the transport, dispersion, and removal processes are very complex and it is not easy to analyze the data. Backward trajectory calculations have been widely used to relate sources of air pollution to subsequent air quality measurements. However, air parcel trajectories can only be determined accurately if wind fields are known with a high degree of spatial and temporal resolution. For large-scale air pollution, the spatial and temporal resolution of meteorological data is insufficient and the accuracy of trajectories is not well known.

Recent economic development in East Asia has been accompanied by increased consumption of fossil fuels and emission of air pollutants such as carbon dioxide and sulfur dioxide. If current growth is maintained, East Asia will become a major area for emissions of air pollutants in the next century. It is thus important to study the characteristics of transport processes in East Asia. Long-range transport of air pollutants in East Asia has been investigated in several recent studies. Mukai *et al.*(1994) estimated the origin of Pb transported to Oki Island, and found that isobaric trajectories were more common than isentropic ones due to the frequent passage of fronts during the study period. Parungo *et al.*(1994) studied transport of black carbon aerosols from China to the East China Sea using trajectory analysis. There are also modeling studies investigating transport, chemistry and deposition of acidic species (Ichikawa and Fujita, 1995; Kitada and Lee, 1993; Murao *et al.*, 1993). The main difficulty in studying transport processes over East Asia is that there are sea areas without rawinsonde data.

The study here first examined the trajectory analysis technique applied to ground-based air quality monitoring or large-scale field studies performed near the surface in this region. To evaluate the accuracy of calculated trajectories, 'true' trajectories must be known. Several techniques have been employed to obtain 'true' trajectories. The most attractive technique is tracer experiment (Haagenson *et al.*, 1987; Kahl and Samson, 1988; Carhart *et al.*, 1989), but tracer gas experiments over large areas are difficult and expensive, and no study has been made for East Asia. An alternative to verifying trajectories is to compare them with tetroon (constant volume balloon) flight trajectories (Warner *et al.*, 1983; Kuo *et al.*, 1985). However, Lamb(1981) showed large differences between tetroon trajectories and fluid particle (such as air pollutant) trajectories in some situations. In the absence of observational data to indicate the "true" trajectory, it is difficult to determine which trajectory technique is superior. Thus without addressing accuracy, we compared locations calculated by a conventional trajectory technique with those calculated by a three-dimensional trajectory technique using meteorological field with high temporal and spatial resolution. It is presumed that three-dimensional or isentropic trajectories are superior to isobaric or isosigma trajectories, because it is important to take vertical motion into account in long-range transport. Next, the trajectory case studies were performed for the Hateruma baseline observatory and possible implications for the use of trajectory analysis are discussed.

2. Method

2.1 Meteorological data

To construct accurate trajectories, meteorological data with high temporal and spatial resolution is essential. Some trajectory techniques use only rawinsonde measurements to define transport winds (Kahl and Samson, 1986). These measurements are made twice-daily at locations separated by approx.400 km. As the East Asia region has ocean areas such as the Sea of Japan and the East China Sea, rawinsonde station density is insufficient for establishing trajectories. An alternative is to use analyzed gridded datasets. These datasets are available from meteorological centers such as the Japan meteorology agency and the European Center for Medium-range Weather Forecasts (ECMWF). They show geopotential height, wind, temperature, and humidity with temporal resolutions of 6 to 12 hours. In this study, ECMWF fields are obtained for 11 pressure surfaces (1000, 925, 850, 700, 600, 500, 400, 300, 250, 200, 150, 100 hPa) on a 0.5×0.5 degree of latitude-longitude grid with a temporal resolution of 6 hours. This spatial resolution corresponds to approx.50 km in the vicinity of Japan. However the native model grid information only has a spatial resolution of approx.100 km. The meteorological field generated by the Japan Spectral Model (JSM) is also used in this study. This dataset is specially output and archived for shipboard chemical measurements described later and has spatial and temporal resolutions of approx.50 km and 20 minutes. There are 19 vertical levels, 10 in the Planetary Boundary Layer (PBL). Some advantages of the JSM dataset are: the meteorological data are available with high temporal and vertical resolutions; they contain information on scales resolved by the 50 km JSM model grid.

2.2 Trajectory models

The trajectories are constructed in a backward direction and two trajectory models are used: a three-dimensional sigma model and a two-dimensional isentropic model. The characteristics of both models are as follows.

(1) Three-dimensional standard trajectory

A three-dimensional trajectory, called a 'standard trajectory' in the discussion in this paper, was constructed by the JSM data described in 2.1. The trajectory is computed from the u, v, and, $\dot{\sigma}(d\sigma/dt)$ component wind. The time step(Δt) is 10 minutes and linear temporal interpolation is used. As described above, JSM wind fields are published for 20 minute intervals. The high vertical resolution in PBL allows the construction of reliable three-dimensional trajectories which start from near the surface where many chemical measurements are performed. The reason why three-dimensional trajectories are not generally used is that commonly available datasets like ECMWF do not have sufficient spatial and temporal resolution to calculate vertical motion accurately, especially in PBL.

(2) Isentropic trajectory

Isentropic trajectories are constructed by using the ECMWF(Δt = 20min.) and the JSM(Δt = 10min.) datasets based on the method described in Nakayama *et al.*(1994). The isentropic trajectory model assumes that air motion takes place on surfaces of constant potential temperature. The potential temperature θ is the temperature of a parcel of dry air at atmospheric pressure p (hPa) and temperature T (K) if moved adiabatically to a standard atmospheric pressure p_0 (usually 1000 hPa).

$$\theta = T(p_0/p)^{2/7} \tag{1}$$

If a parcel of dry air moves adiabatically, it will conserve its potential temperature. Thus, the movement of a parcel of dry air is limited on an isentropic two-dimensional surface. The advantage of considering isentropic surfaces is that three-dimensional motion of an air parcel can then be calculated only from the horizontal wind data. The application of isentropic trajectories have clear limitations however. The assumption of dry adiabatic motion is well satisfied for large-scale motion in the free troposphere; *i.e.*, above PBL and for periods of several days. However, in regions of cloudiness, precipitation, and strong mixing, the assumption does not hold. Further, because of limited spatial resolution, isentropic surfaces can not be specified for transport winds below the top of PBL (approx.850 hPa).

2.3 The case studied here

(1) Examination of conventional isentropic trajectories

In January 1993 shipboard chemical measurements were carried out on the Sea of Japan to obtain data to evaluate long-range transport of air pollutants from the Asian continent (Kaneyasu, 1994). Here, high concentrations of hydrogen peroxide (H_2O_2) and SO_2 were observed and transport from the Asian continent was suggested. On 26 January 1993 a H_2O_2 concentration of 0.6 ppb was measured at 128.0°E, 28.9°N under anticyclonic flow conditions (Takeuchi *et al.*, 1993). The concentration was significantly higher than that obtained in other areas. A rapid increase in SO_2, to a peak concentration above 10 ppb, was observed on 29 January 1993 at 131.0E, 34.8N. The ppb. This was observed under winter type flow conditions. The differences in the trajectories of the two cases, the 'H_2O_2 episode' and the 'SO_2 episode', were evaluated in four steps:

1) Using JSM wind fields, we constructed a backward three-dimensional standard trajectory from the observation height. For comparison, we also constructed the isentropic trajectory from the top of PBL using JSM wind fields. Further, the isentropic trajectories using JSM wind fields with a degraded temporal resolution of 6 hours was also constructed.
2) ECMWF wind fields were used to construct a backward isentropic trajectory on the isentropic surface close to the top of PBL (850 hPa) over the observation point. Then the three-dimensional standard trajectories were compared with the isentropic trajectory.
3) A three-dimensional trajectory which starts at the top of PBL was also constructed to evaluate the effect of differences in starting height on the trajectory calculations over the region. Trajectories which start from the top of PBL appear in many studies especially in the Lagrangian model studies of acid deposition (Eliassen and Altbones, 1983). This treatment may have been adopted largely due to data availability and computational cost. Here, it may be assumed that transported 'aged' pollutants are well mixed though the boundary layer. The validity of this assumption may be examined by comparing the two three-dimensional trajectories.
4) A comparison of the three-dimensional trajectory with the isentropic trajectories which start from the same height allows the exclusion of the influence of starting point height and an examination of how differences in the meteorological data and in the trajectory models affect the trajectory calculations.

The conditions of the trajectories are detailed in Table 1.

(2) Application of trajectory analysis

The Japan Environmental Agency established the Hateruma Island baseline observatory in 1993 (Utiyama *et al.*, 1994). It is located in the south of Japan (24.0°N, 123.8°E). The dominant meteorological feature in summer is southeasterly flows and the observatory is free from anthropogenic influence. However because it is located to the west of the Asian continent,

Table 1. Details of the trajectories utilized in this study

Met. Data	Temporal resolution (hr)	Trajectory Type	Starting height	Comment
JSM	20min.	3-Dim.	Surface	Standard trajectory, used in Fig.1 and Fig.3
JSM	20min.	3-Dim.	~850 hPa	used in Fig.3 and Fig.5
JSM	20min.	isentropic	~850 hPa	used in Fig.5
JSM	6 hr	isentropic	~850 hPa	used in Fig.5
ECMWF	6 hr	isentropic	~850 hPa	used in Fig.1 and Fig.5

the observatory is influenced by emissions from Taiwan and China in other seasons, when westerly winds prevail. Ozone and aerosol number concentrations (diameter of 0.3-0.5 μm) measured during April to December 1993 were used to test the applicability of isentropic trajectories. The method of the analysis is as follows.

1) Measurements for each month are classified into groups based on 1-hour ozone and aerosol concentrations.
2) The conventional isentropic trajectories which start from the top of PBL are developed for each group. If there are common characteristics in the grouped trajectories, the reasons for these are considered.

3. Results and discussion

3.1 Adequacy of conventional isentropic trajectory analysis

(1) Differences between three-dimensional standard trajectories and isentropic trajectories

To examine differences between three-dimensional standard trajectories and conventional isentropic trajectories using ECMWF wind fields, trajectories corresponding to the SO_2 episode and the H_2O_2 episode described previously were plotted. The results are shown in Fig. 1. For the H_2O_2 episode the three-dimensional standard backward trajectory during the first 36 h shows movement in the northerly direction with anticyclonic curvature reflecting the high pressure system located in southern China. Then, the trajectory moved slowly across Seoul. From the height profile of this trajectory, it appears that the air parcel which passed over Seoul at the height of 1000 m transported the photochemical oxidants to the observation point. This does not contradict the consideration that high concentrations of H_2O_2 were produced by photochemical reactions in the urban atmosphere.

The three-dimensional standard trajectory for the SO_2 episode showed movement in a northwesterly direction. The trajectory indicates that the air parcel originated in the Shantung peninsula 3 days earlier, descended and crossed the south end of South Korea and then arrived at the observation point.

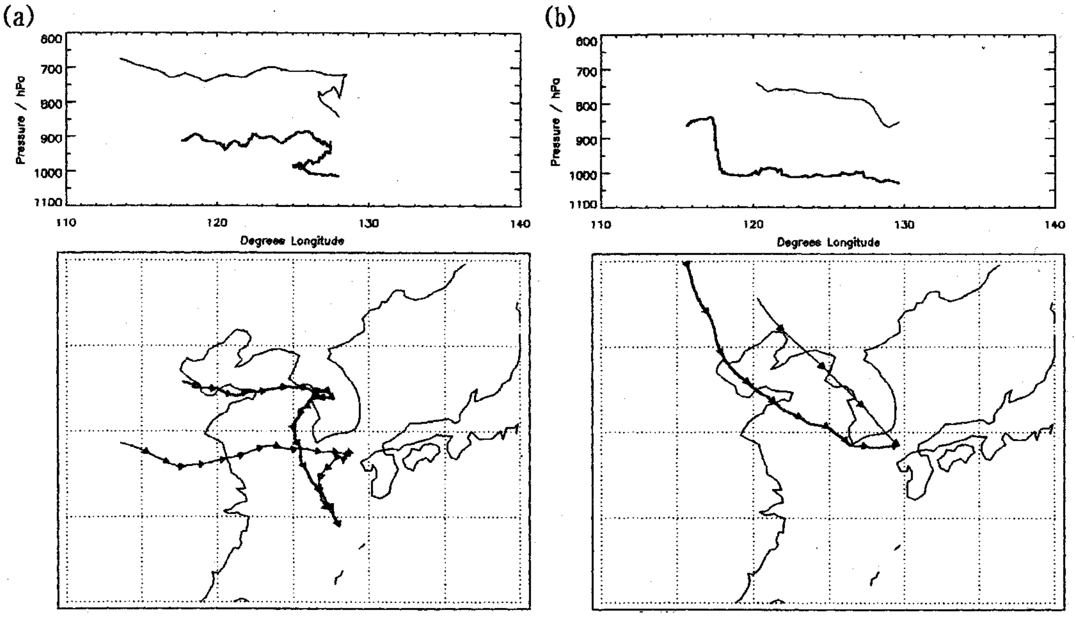

Figure 1. Three-dimensional standard (thick line) and isentropic (thin line) trajectories using ECMWF wind fields for (a) the H_2O_2 and (b) the SO_2 episodes described in section 2.3. Arrows are for 6-hr increments.

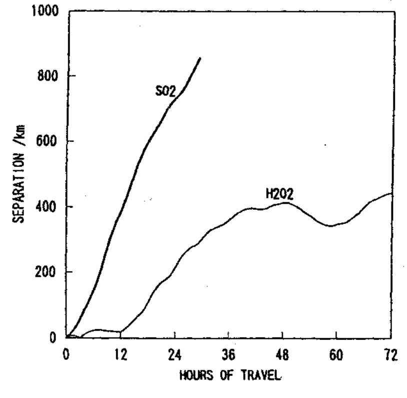

Figure 2. Horizontal Separation (km) between the three-dimensional standard trajectory and the isentropic trajectory using ECMWF wind fields for the H_2O_2 and SO_2 episodes.

The isentropic trajectories behaved quite differently, particularly in the case of the H_2O_2 episode. For the SO_2 episode, strong northwesterly wind prevailed over the region, and the resultant isentropic trajectory showed a fast transport of air from Asia. Although the two trajectories for the SO_2 episode followed very similar paths, there is a marked difference in position when they are compared at the same travel times as shown in Fig. 2. Because the starting point of the isentropic trajectory is set at the top of PBL (850 hPa), the faster progress of the isentropic trajectory than that of the three-dimensional trajectory may be largely due to the strong wind in the upper layers.

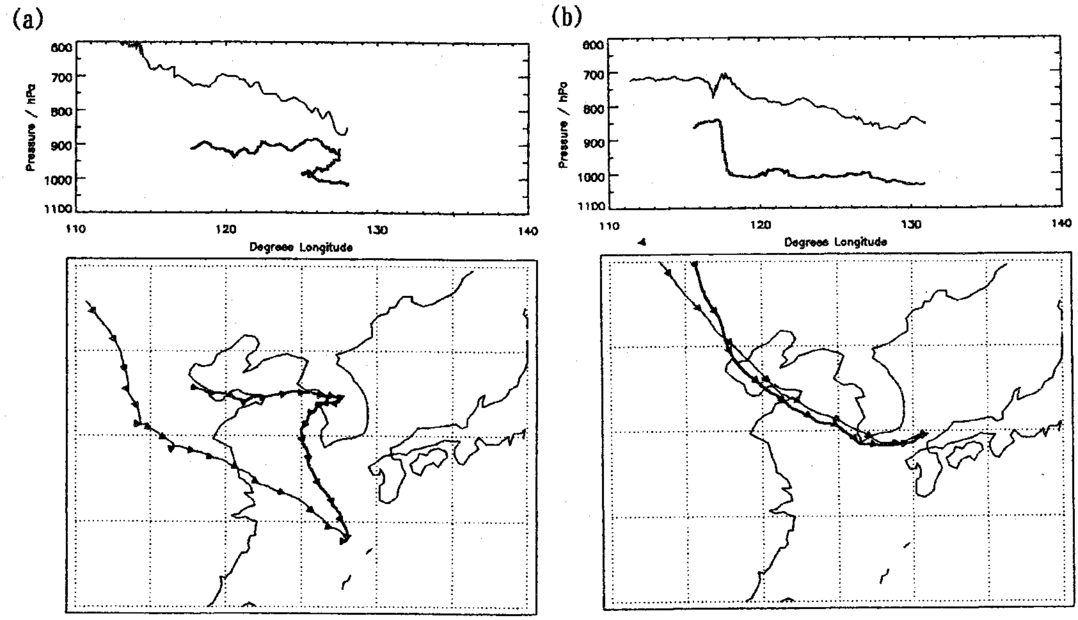

Figure 3. The three-dimensional trajectory starting from the observation point (thick line) and that starting from the top of PBL (thin line) for (a) the H_2O_2 and (b) the SO_2 episodes. Both trajectories are computed from JSM wind fields. Arrows are for 6-hr increments.

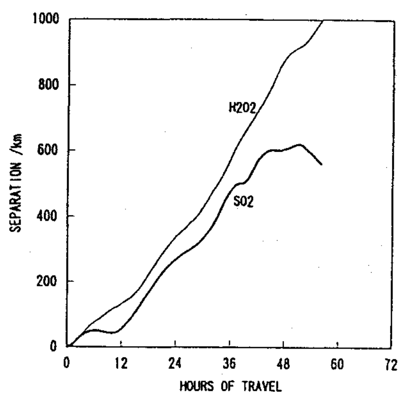

Figure 4. Horizontal Separation (km) between the three-dimensional trajectory starting from the observation point and that starting from the top of the PBL for the H_2O_2 and SO_2 episodes. Both trajectories are computed from the JSM wind fields.

(2) Reasons for differences in trajectories - How differences in starting point height affect the trajectories -

The differences between the trajectories may be attributed to 1) the differences in starting point height, 2) differences in trajectory models, and 3) differences in the meteorological data. To establish how the differences in starting point height affect the trajectories, we compared three-dimensional standard trajectories with three-dimensional trajectories starting at the top of PBL using JSM wind fields. The results are shown in Fig. 3. This demonstrates that the

(a)

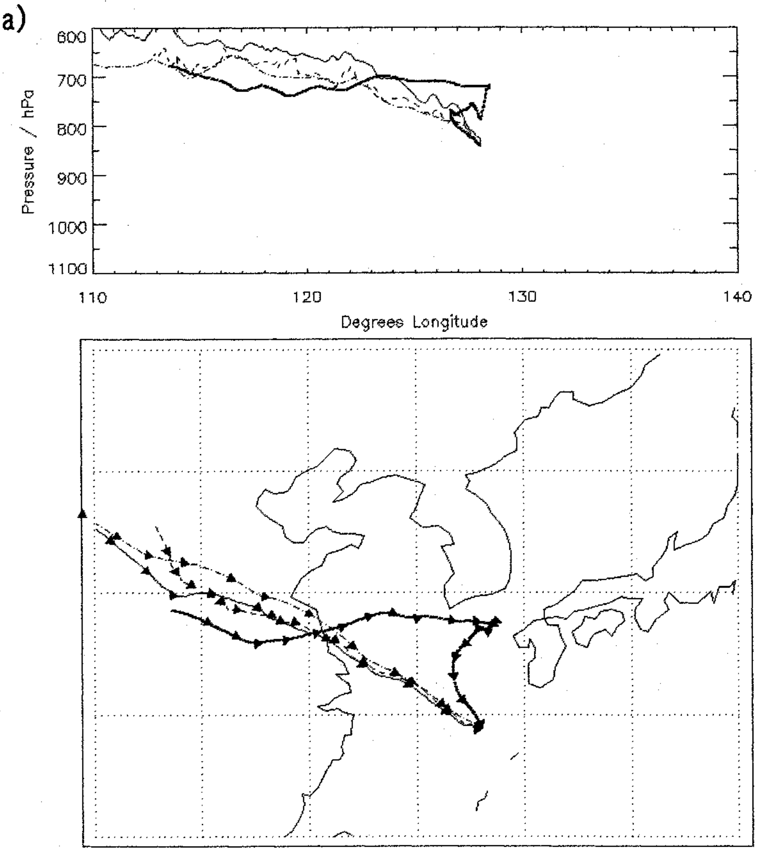

Figure 5. Three-dimensional and isentropic trajectories starting from the same height for (a) the H_2O_2 and (b) the SO_2 episodes. Heavy line: isentropic trajectory using ECMWF. Thin line: 3-dim. trajectory using JSM. Dashed line: isentropic trajectory using JSM. Chained line: isentropic trajectory using JSM with 6 h resolution. Arrows are for 6-hr increments.

trajectory starting from the surface is very different from that starting from the top of PBL. As shown in Fig. 4, the 24h horizontal separation between the trajectories are approx.300km for both episodes.

(3) Reasons for differences in trajectories - How differences in the meteorological data and the trajectory models affect the trajectories -

Another source of differences comes from the differences in the meteorological data and the differences in trajectory calculations. Pickering *et al.*(1994) showed large differences between trajectories over the South Atlantic based on wind fields from different analysis centers. To determine how differences in the meteorological data affect the trajectories, we have compared the isentropic trajectories with the three-dimensional trajectories which start from the top of PBL. The results are shown in Fig. 5. The 24h horizontal separation between the three-dimensional trajectory using JSM wind fields and the isentropic trajectory using ECMWF wind fields exceeds 500 km for both episodes (Fig. 6). This separation is greater than those

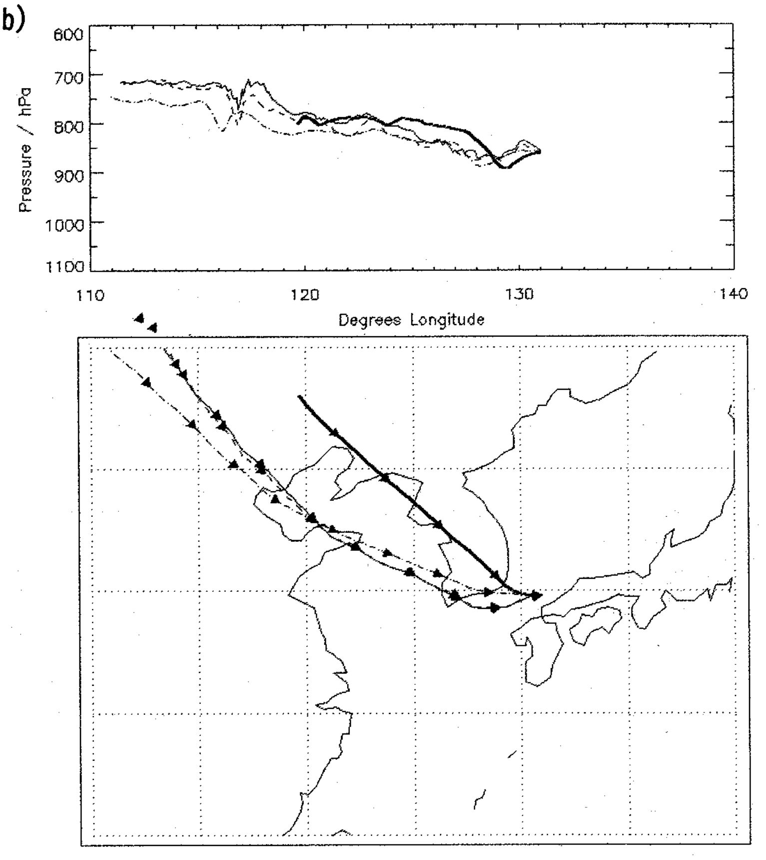

Figure 5. (Continued)

caused by the differences in starting point height. However the separation between the three-dimensional trajectory and the isentropic trajectory using the same JSM wind fields is quite small. Moreover, the isentropic trajectory using JSM wind fields with degraded temporal resolution showed almost identical movement. These results strongly indicate that differences in meteorological data affect the differences in the trajectories.

In this study, trajectory analysis was applied to the chemical measurements in the Sea of Japan where the density of meteorological observations is low. Moreover, the development of a mixing layer may be limited in this region for the period of the study. Although the conditions studied make it difficult to apply the isentropic trajectory, it is necessary to understand the above-mentioned difference when isentropic trajectory analysis is used with the surface chemical measurements in this region.

3.2 Application of isentropic trajectory analysis to Hateruma baseline observations

As discussed above, a single isentropic trajectory which uses routine meteorological data for surface chemical measurements does not always indicate the exact pathway of an air

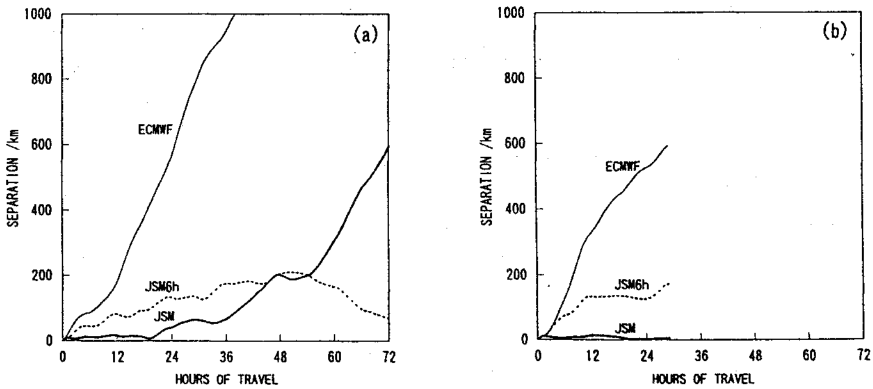

Figure 6. Horizontal separation (km) between the three-dimensional trajectory and the isentropic trajectories starting from the same height for (a) the H_2O_2 and (b) the SO_2 episodes.

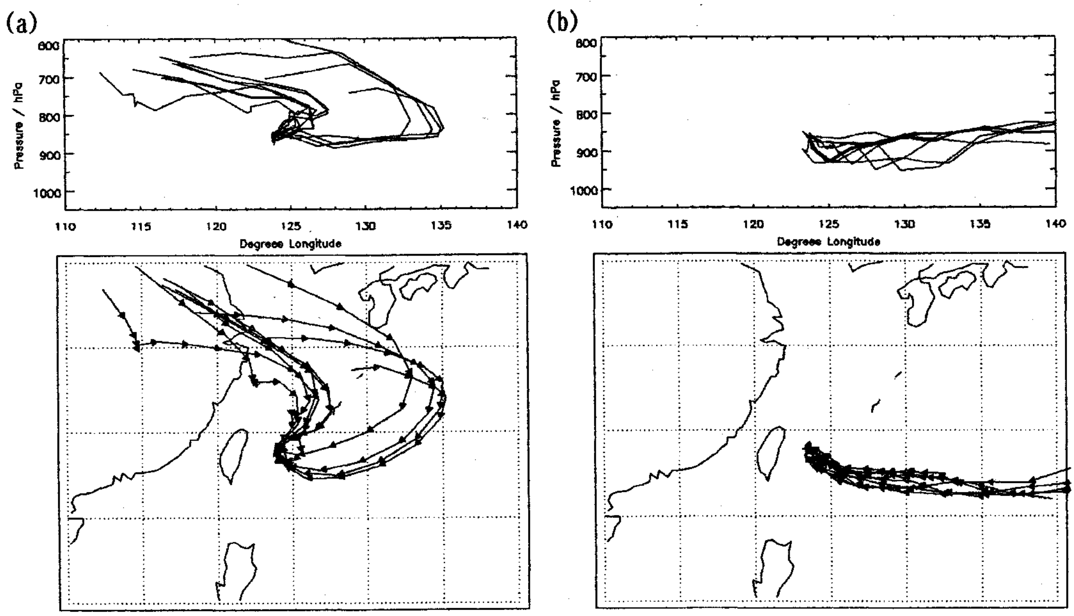

Figure 7. Isentropic trajectories classified by ozone concentration observed at Hateruma (a) Ozone concentrations above 40 ppb, (b) ozone concentrations below 30 ppb. Arrows are for 6-hr increments.

parcel. However, it is hoped that an analysis of a large number of trajectories will provide a useful representation of general flow patterns so that interpretation of the chemical measurements becomes possible. Constructing a large number of trajectories, several recent studies investigated the flow climatology for baseline observatories (Miller *et al.*, 1993; Harris and Kahl, 1990; Moody *et al.*, 1995). Instead of classifying trajectories, the 1-hr observations for each month in 1993 were grouped by using ozone and aerosol concentrations. Then, using ECMWF wind fields we constructed the isentropic trajectories for the grouped observations and examined how the trajectories could explain the results of the surface measurements.

The result for December 1993 is shown here as an example. The 1-hr ozone concentrations ranged from 20 to 50 ppb, and 1 hr aerosol concentrations ranged from 0 to 60,000 l^{-1}. Because the objective of this analysis is to provide a test of grouped trajectories, we first omitted observations for which interpretation is difficult; *i.e.*, we selected observations associated with aerosol concentrations below 10,000. Then observations were grouped by ozone concentration into two groups: (a) less than 30 ppb; and (b) more than 40 ppb. Isentropic trajectories using ECMWF meteorological fields were constructed for each group. The results are shown in Fig. 7. Because clean-air observations were selected and photochemical activity is low in winter, the high concentrations of ozone may be caused by transport from above the PBL rather than by photochemical reactions in the PBL. In addition, the low concentrations of ozone may be caused by transport of clean marine air. The results of the isentropic trajectory analysis in Fig. 7 correspond to these expectations. The results of the analysis are summarized as follows.

1) For observations where both ozone and aerosol concentrations were low, all trajectories to the observatory showed travel over the Pacific ocean. These observations may be understood to be a result of the transport of clean oceanic air. The reliability of the trajectories for this case seems high.
2) For observations with high ozone and low aerosol concentrations, many trajectories showed descending transport from the free troposphere. These observations may be understood to be caused by transport from the free troposphere where ozone concentrations are high and aerosol concentrations are low. The reliability of the trajectories for this case is also high.
3) For observations with low ozone and high aerosol concentrations, some trajectories indicate transport over the industrial region of southern China. They suggest influences of human activity. However, for these observations there is the possibility of transport within the PBL where the isentropic model is less reliable due to adiabatic effects. It is not possible to estimate the source region from the trajectories.
4) The trajectories also indicated transport over the industrial regions of Japan, Taiwan, and China for observations where both the ozone and aerosol concentrations were high. However, there may be problems with the reliability of individual trajectories described in case 3 above.

4. Conclusions

This study examined the adequacy of conventional isentropic trajectory analysis applied to surface chemical measurements to determine background concentrations and establish long-range transport of pollutants. In the absence of observations which indicate a 'true' trajectory, we first compared locations calculated by isentropic trajectories with three-dimensional trajectories using meteorological fields with high temporal and spatial resolutions. Then, trajectory case studies were performed for the Hateruma baseline observatory and the implications of trajectory analysis are discussed. The results can be summarized as follows:

1) A comparison of two trajectory models was made for shipboard measurements in the Sea of Japan in January 1993. There were large differences between the three-dimensional trajectory which starts from the observation point and the conventional isentropic trajectory starting from the top of the PBL. The difference in 24h location exceeded 200 km. A comparison of two three-dimensional trajectories using the same meteorological data but starting at different heights showed that the difference in starting height is a large source for the differences in trajectories. The results clearly indicate the limits to the use of conventional isentropic trajectory analysis for chemical measurements performed in the

PBL. Comparing three-dimensional trajectories with isentropic trajectories starting at same height showed that the differences were caused by differences in starting height and by differences in the meteorological data. Thus, the use of a single isentropic trajectory for the surface chemical measurements to determine transport pathways is not adequate and isentropic trajectory analysis or interpretation of the results of the analysis with a clear understanding of the limitations discussed in this study is necessary.

2) Using ozone and aerosol concentrations observed at Hateruma observatory, we examined grouped isentropic trajectories to interpret the behavior of ozone. It was shown that the grouped trajectories for clean-air observations when aerosol concentrations were low could show large-scale atmospheric motion responsible for the ozone behavior at the observatory. However because of the possibility of transport in the PBL, the adequacy of grouped isentropic trajectories for polluted air mass detection in this region can not be finally determined.

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