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Vertical Distribution of the Tropospheric Ozone over Japan: The Origin of the Ozone Peaks

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Abstract

Vertical distributions of the tropospheric ozone over Wakasa Bay, Enshu-nada and some other places in Japan were measured by a series of aircraft measurements during 1987–1991. The backward trajectories of air parcels with the ozone peaks and potential vorticity distributions around them were then analyzed to study their origin. This result showed that the ozone peaks over Japan sometimes originated from the tropopause folding which occurred in the west of Japan. Two ozone peaks had different altitudes and concentrations over Enshu-nada on 21 January 1989 which originated from different tropopause foldings. They seemed to diffuse during the transport which resulted in a stratified structure forming over Japan. The two similar ozone peaks, which originated from the identical tropopause folding and were observed 300 km away on 21 January 1989, belonged to the identical ozone layer based on the potential temperature inclination. Two different types of air masses were simultaneously observed at different points over Japan on 8 August 1990. One was the continental air mass which showed a high ozone and a low water vapor concentration being transported from the northeast of China. The other was the maritime air mass which showed a low ozone and a high water vapor concentration staying over the sea for a few days that didn't come across the stratospheric air mass and pass over the metropolitan area. However, the ozone and water vapor showed a positive correlation even when the maritime air mass passed over the metropolitan or industrial areas, thus showing that the concentrations of the ozone and vapor were both high. The vertical distribution of ozone on 27 April 1991 over Tsukuba was almost uniform and the concentration was approximately 70 ppbv. This suggested that the air masses came from the folding area whose vertical inclination of potential vorticity was small and was transported shortly after this event. Not only intrusion from the stratosphere but diffusion during the transport was also important for the ozone distribution over Japan based on the relationship between the concentration of ozone and the distance of transport.

1. Introduction

Though various meteorological processes are thought to cause stratospheric/tropospheric exchanges (Shapiro, 1980), the dominant processes of stratospheric/tropospheric exchange are tropopause folding (Danielsen and Mohnen, 1977) or tropopause folding and mean meridional circulation (Singh *et al.*, 1980). On the other hand, a major loss process is the destruction on land and ocean surface (Fabian and Pruchniewicz, 1977; Newell, 1963; Danielsen and Mohnen, 1977; Galbally and Roy, 1980). Lately, in addition to the stratosphere-troposphere exchange processes noted above, photochemical processes in the troposphere can also be a significant source or sink of the tropospheric ozone (Fishman and Crutzen, 1978). Japan is located in the western Pacific rim region which is on the border of the continental and the maritime air masses. Besides, the most intense jet stream over Japan in the northern hemisphere is associated with repeated cyclogenesis in this area (Austin and Midgley, 1994). Muramatsu (1989) has also pointed out that the north of Japan is the region where the stratospheric air mass intrudes actively into the troposphere. Thus, the study of ozone in the free troposphere over Japan is an important subject. The ozone measurement in the free troposphere over Japan has been made at the top of Mt. Fuji since 1992 (Tsutsumi *et al.*, 1994). Though this measurement showed a significant seasonal change and a typical tropopause folding event in the free troposphere, the information about the vertical profile of ozone was limited. Since an aircraft measurement is useful for the study of vertical or horizontal

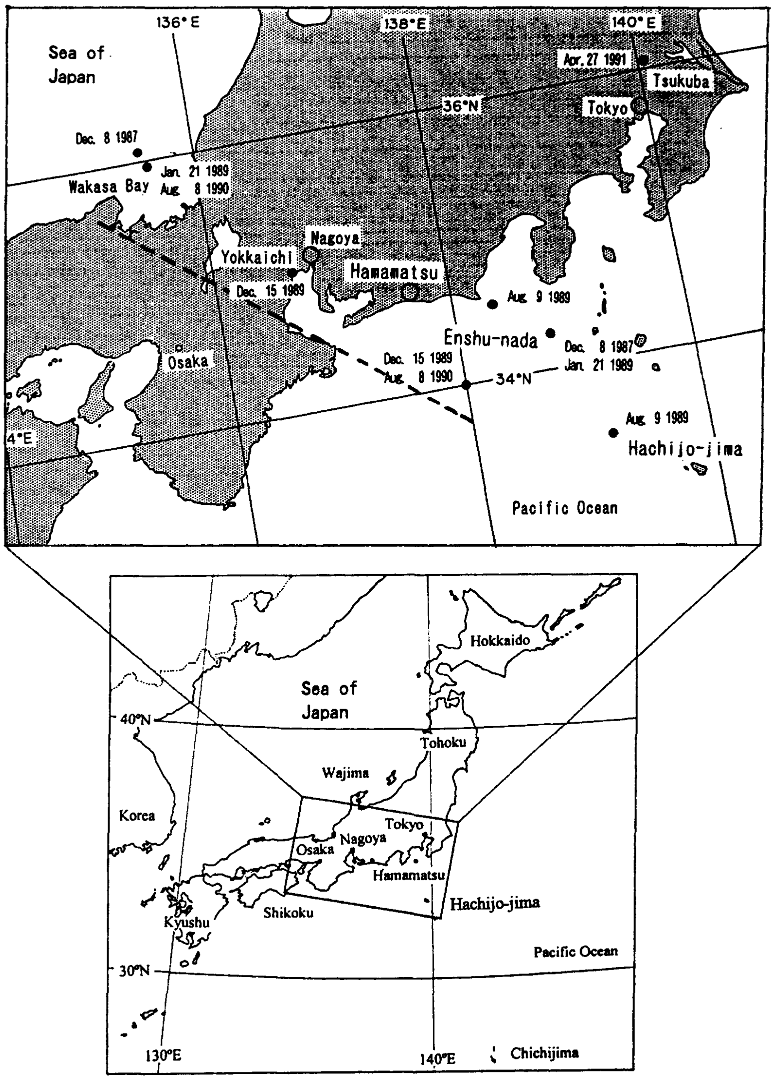

Fig. 1. Research map for a series of aircraft measurements. The closed circles show the research area where the vertical distribution was observed. The dashed line indicates the location of the cross section which is shown in Fig. 7.

distributions of the tropospheric ozone, numerous aircraft measurements on tropospheric ozone were performed, including projects like TROZ (Fabian and Pruchniewicz, 1977), GASP (Nastrom, 1979), GAMETAG (Routhier and Davis, 1980), TROPOZ (Marenco *et al.*, 1990) and INSTAC (Tsutsumi *et al.*, 1991; Kondo *et al.*, 1993). Though the aircraft measurement on the tropopause folding over Japan was made by Muramatsu *et al.* (1984), this measurement was focused on the folding structure. There are 4 ozone sonde stations in Japan and many vertical ozone profiles have been accumulated for more than 30 years. However the vertical resolution and accuracy of the concentration in the troposphere were not sufficient to analyze the details of the ozone profiles. Thus, the vertical structure of the tropospheric ozone over Japan has not been fully resolved yet. The vertical distribution of ozone over the tropical region in the Pacific Ocean showed a simple profile in the INSTAC project. For example, the concentration of ozone simply increased with altitude over Yap (138°E, 10°N) and, on the other hand, simply decreased with altitude over Davao (125°E, 7°N) (Tsutsumi *et al.*, 1991). How is the ozone distributed in the troposphere over Japan and where does it come from? A series of aircraft measurements from 1987 to 1991 were made to study the vertical distribution of the tropospheric ozone over Japan and the origin of their peaks.

2. Experimental and analysis

Air was sampled from an intake on the aircraft and was introduced to an ozone monitor. The ozone monitor was an UV absorption type (Dasibi: Model 1008 and later Dylec: 1006-AHJ) and measured every 12 seconds. The concentration of ozone was

Table 1. Flight data of a series of aircraft measurement from 1987 to 1991. The morning means around 11 A.M. and the afternoon means around 2–3 P.M.

Flight Number	Date	Time	Research Area	Ascent/ Descent	Remarks and Instrument
—	Dec. 8 1987	morning afternoon	Waksa Bay Enshu-nada	Descent Descent	high pressure over Japan model 1008
Flight A	Jan. 21 1989	morning afternoon	Wakasa Bay Enshu-nada	Descent Descent	typical winter mon-soon model 1006-AHJ
Flight B	Dec. 15 1989	afternoon afternoon	Enshu-nada Yokkaichi	Descent Descent	typical winter mon-soon model 1006-AHJ
Flight C	Aug. 9 1989	afternoon afternoon	Enshu-nada Hachijo-jima	Ascent Descent	typical summer model 1006-AHJ
Flight D	Aug. 8 1989	afternoon afternoon	Wakasa Bay Enshu-nada	Descent Descent	summer but typhoons were located at east and west of Japan model 1006-AHJ
Flight E	Apr. 27 1991	afternoon	Tsukuba	Descent	clear later cloudy model 1006-AHJ

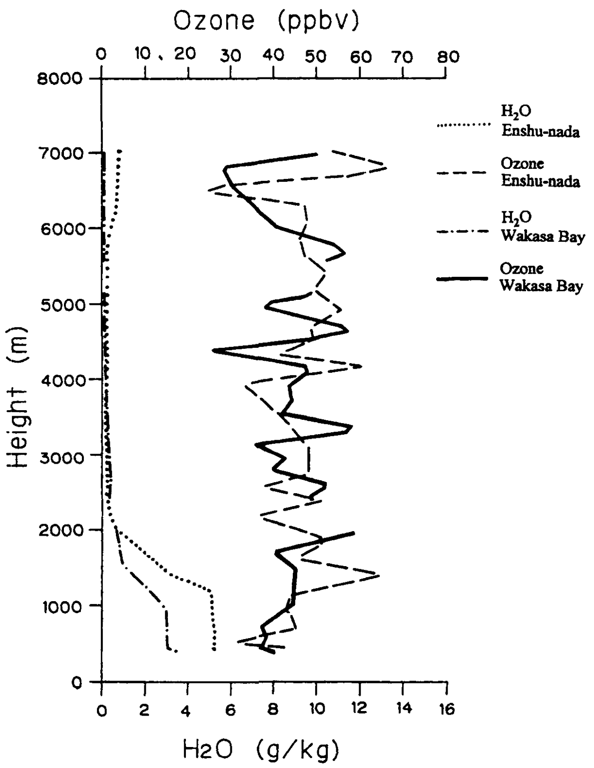

Fig. 2. The vertical distributions of ozone and water vapor over Wakasa Bay and Enshu-nada on 21 January 1989.

modified to calibrate the standard temperature and pressure. An altitude, air temperature and dew point were monitored simultaneously. The air temperature was measured by Model TS-051P (Makino Oyo Sokki) and the dew point was measured by Model 137-C3 (EG&G) whose significant range was from −50 to 70°C.

Six flight measurements — three in winter, two in summer and one in spring — were made during 1987–1991. The research points and flight information are shown in Fig. 1 and Table 1. Five measurements which showed significant features were analyzed. The vertical distributions were acquired during the spiral ascent or descent from a few hundred meters to about 7000 m. Two different points for measuring the vertical distribution of ozone were selected. The first point was over Wakasa Bay (36°N, 135.5°E) — north of Osaka and usually the windward side of Japan in the winter monsoon. The second point was over Enshu-nada (34°N, 138°E) — southeast of Nagoya and the leeward side of Japan in the winter monsoon. These two points were about 300 km apart. Additional vertical measurements were also made over Yokkaichi (35°N, 136.8°E), Hachijo-jima (33.5°N, 137.1°E) and Tsukuba (36°N, 140°E). Every flight departed from Yao airport (34.5°N, 135.6°E) in Osaka and took about 30–90 minutes to observe the vertical distribution at each research point. There are two big cities around the research points of these aircraft measurements. Osaka has a population of about 250 million and is located to the west and the south sides of the research points. Nagoya has a population of about 200 million and is surrounded by an industrial belt. Nagoya is located southeast of Wakasa Bay and northwest of Enshu-nada.

The backward trajectories of air parcels along the isentropic surface were calculated using objective analysis data from the Japan Meteorological Agency (JMA) at several altitudes of each research point. The vertical levels for the objective analysis data were 1000 hPa, 850 hPa, 700 hPa and 500 hPa. The longitudinal and latitudinal resolution was 1.875° and the time resolution was 12 hours. The objective analysis data has not always fully reflected the real atmosphere, especially when a vertical distribu-

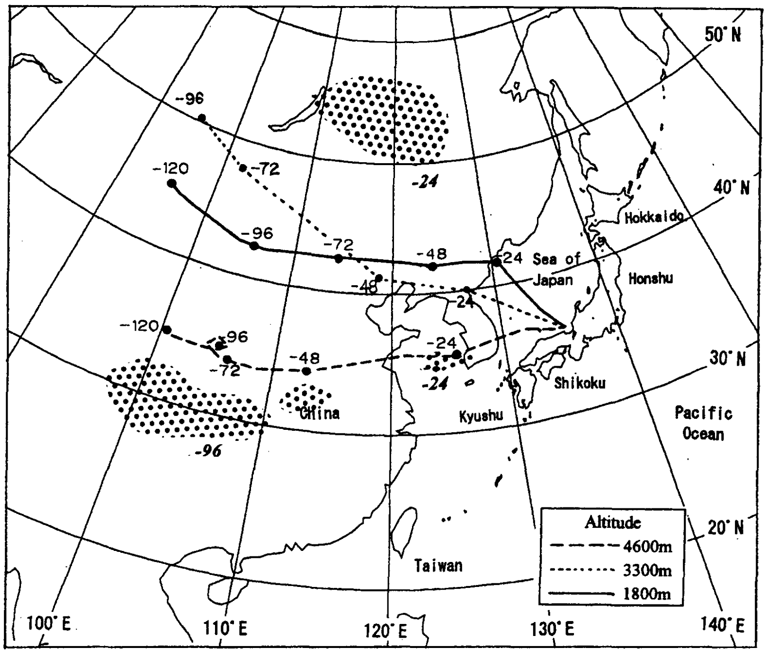

Fig. 3. The backward trajectories from the 4600 m, 3300 m and 1800 m levels over Wakasa Bay on 21 January 1989. The trajectories were calculated to 5 days (120 hours) before the measurement. The closed circles on the trajectories indicate the positions of the air parcels every 24 hours. Dotted regions indicate the region where the potential vorticiy was larger than 1.0E-6 $m^2s^{-1}Kkg$ at the 500 hPa level and italic numbers mean the hours before the measurement when the tropopause folding occurred.

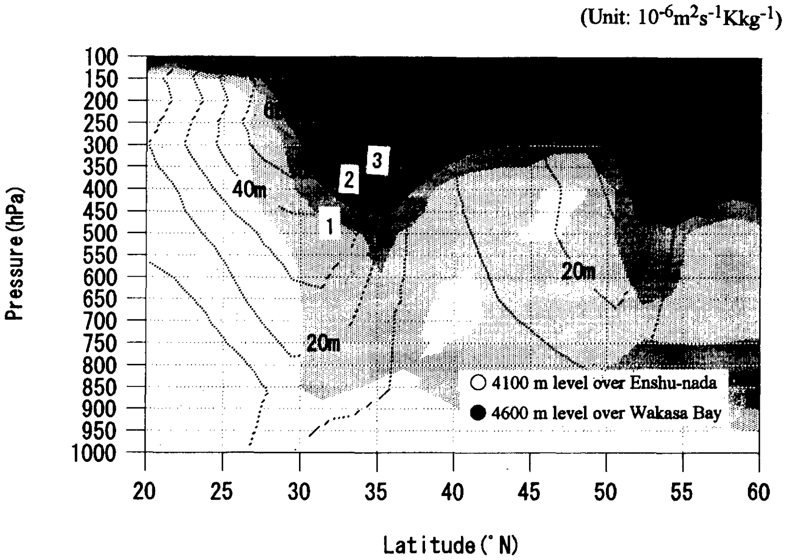

Fig. 4. The cross section chart of potential vorticity along 125°E at 9 JST on 20 January 1989 — 24 hours before the measurement. Break lines indicate isopleth of wind speed (m/s). The symbols indicate the points where the backward trajectories from each research area passed through. The numbers in the figure indicate the intensity of potential vorticity (10^{-6} $m^2s^{-1}Kkg^{-1}$).

tion of wind speed or wind direction has a singular point, then the calculated trajectory could have an error.

Potential vorticity $g(\zeta + f)\frac{\partial\vartheta}{\partial p}$ is a conservative quantity under an adiabatic condition and the potential vorticity in the stratosphere is higher than that in the troposphere. Reed (1955) has used the potential vorticity as a tracer for the stratospheric air mass. Shapiro (1974) has found a close relationship between the potential vorticity and con-

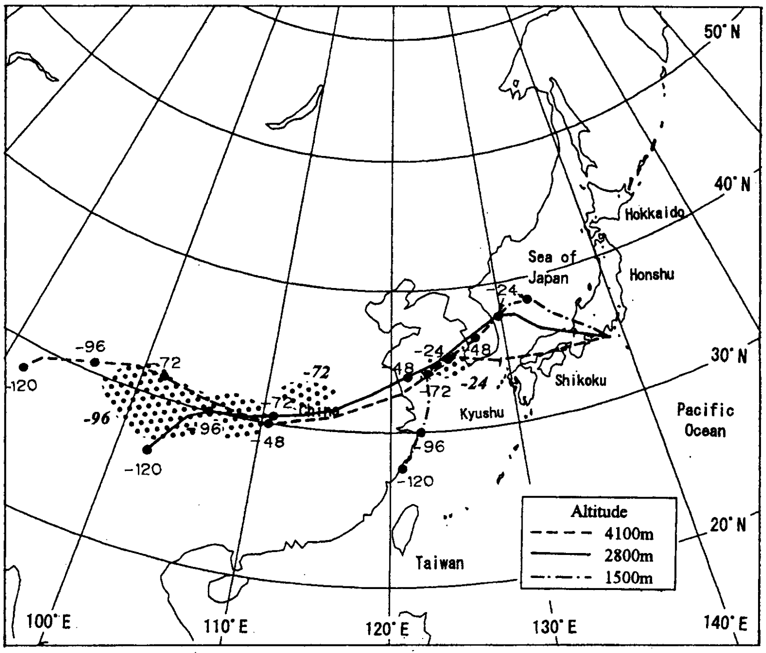

Fig. 5. Same as Fig. 3, but the backward trajectories from the 4100 m, 2800 m and 1500 m levels over Enshu-nada on 21 January 1989.

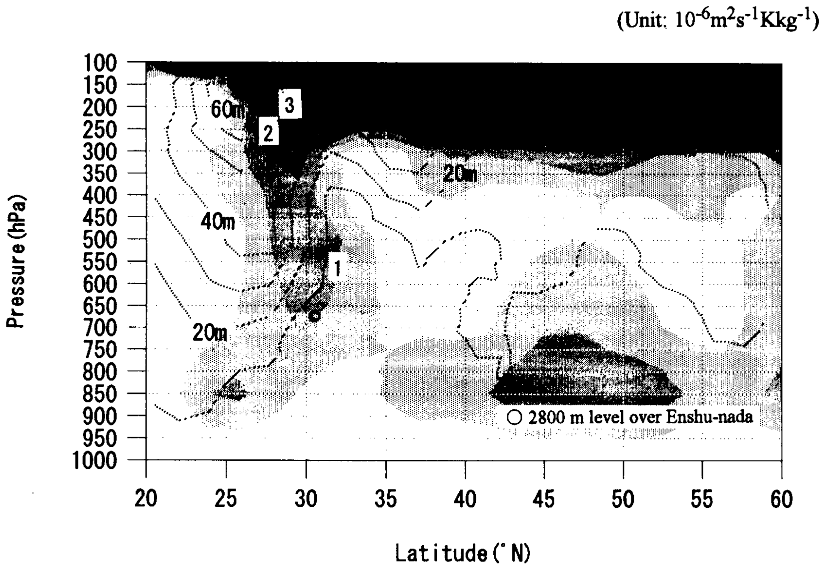

Fig. 6. Same as Fig. 4, but along 105°E at 9 JST on 17 January 1989 — 96 hours before the measurement.

centration of ozone in the baroclinic zone beneath a jet stream. Since the high potential vorticity region ($> 1.0 \times 10^{-6}$ $m^2s^{-1}Kkg^{-1}$) is usually located in the upper level, the region where this high potential vorticity falls down to the lower atmosphere has the possibility of having an intrusion from the stratosphere. As a result, the region with high potential vorticity larger than 1.0 (10^{-6} $m^2s^{-1}Kkg^{-1}$) at 500 hPa level was defined as a high potential vorticity region where the stratospheric air mass could have intruded into the troposphere. The main high potential vorticity regions around the trajectories were calculated using the objective analysis data from the JMA which are marked as the dotted areas in the trajectory charts to check whether the trajectories were affected by the folding or not. A tropopause folding occurs in a few hours, however the time resolution of the objective analysis data is 12 hours. Thus it is difficult to catch the instant when the trajectory is in the fold. Since the stratospheric air is transported to the lower troposphere by folding (Johnson and Viezee, 1981), we presume

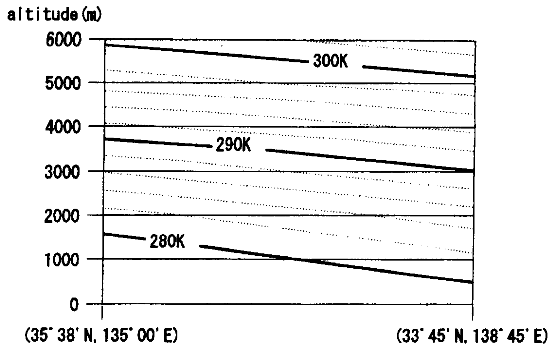

Fig. 7. The cross section chart of potential temperature (K) from Wakasa Bay (35°38′N, 135°00′E) to Enshu-nada (33°45′N, 138°45′E) at 9 JST on 21 January 1989.

the effect of the tropopause folding on the trajectory from the relationship between the cross section of potential vorticity and the location of the trajectory.

3. Results and Discussion

3.1 Flight A (21 January 1989)

Five days before this flight, an anticyclone covered Japan. After that, an active cyclone traversed Japan. On the measurement day, this cyclone was located off the east coast of Tohoku district — the northern part of Honshu. The pressure pattern was a typical winter monsoon type, and the wind direction was west to northwest over Japan.

Figure 2 shows the vertical distributions of ozone and water vapor over Wakasa Bay and Enshu-nada. The ozone concentrations were not measured from the 2000 m to the 2400 m levels and from the 5100 m to the 5500 m levels over Wakasa Bay as the instruments were being checked out. The ozone variation of the vertical profile over Wakasa Bay was as large as 30 ppbv and two distinct peaks appeared at the 4600 m and the 3300 m levels. Figure 3 shows the backward trajectories from the 4600 m, 3300 m and 1800 m levels (the last layer showed a lower ozone) over Wakasa Bay. As illustrated with the dashed line in this figure, the trajectory from the 4600 m level passed over the Yellow Sea (125°E, 35°N) — 24 hours before the measurement. Figure 4 is the cross section chart of the potential vorticity along 125°E at 9 Japan Standard Time (JST) on 20 January — 24 hours before the measurement. This figure shows that a tropopause folding occurred over the Yellow Sea. The closed circle in this figure indicates that the backward trajectory from the 4600 m level over Wakasa Bay passed through the high potential vorticity region ($\sim 1.0 \times 10^{-6}$ m^{2}s^{-1}Kkg^{-1}) in the fold. Hence, the ozone layer of high concentration around the 4600 m level over Wakasa Bay

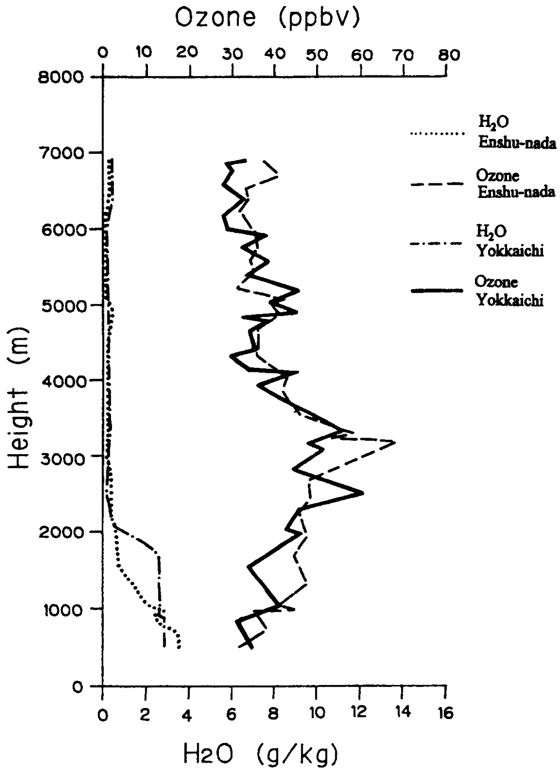

Fig. 8. The vertical distributions of the ozone and water vapor over Enshu-nada and Yokkaichi on 15 December 1989.

originated from the tropopause folding. The trajectory from the 1800 m level over Wakasa Bay (solid line in Fig. 3) shows that the air parcel came from the north of China, passing over the north of North Korea 24 hours before the measurement. Since the dotted region in Fig. 3 shows that the high potential vorticity region did not occur around this trajectory, the air parcel of this layer was not affected by the intrusion of the stratospheric ozone-rich air. The trajectory from the ozone peak at the 3300 m level also came from the north of China and North Korea (dotted line in Fig. 3), thus this trajectory was not considered to be affected by the stratospheric air directly as far as this figure shows. The origin of this ozone peak could not be identified.

The vertical distribution of ozone over Enshu-nada varied from 24 ppbv to 66 ppbv. The ozone peaks higher than 60 ppbv appeared at the 6800 m, 4100 m and 1500 m levels (Fig. 2), and a broad peak appeared at the 2800 m level. Figure 5 shows the backward trajectory analysis from the 4100 m, 2800 m and 1500 m levels over Enshu-nada. As illustrated with the dashed line in Fig. 5, the trajectory from the 4100 m level over Enshu-nada passed over the Yellow Sea 24 hours before the measurement as the same route as that from the 4600 m level over Wakasa Bay. The open circle in Fig. 4 shows that this backward trajec-

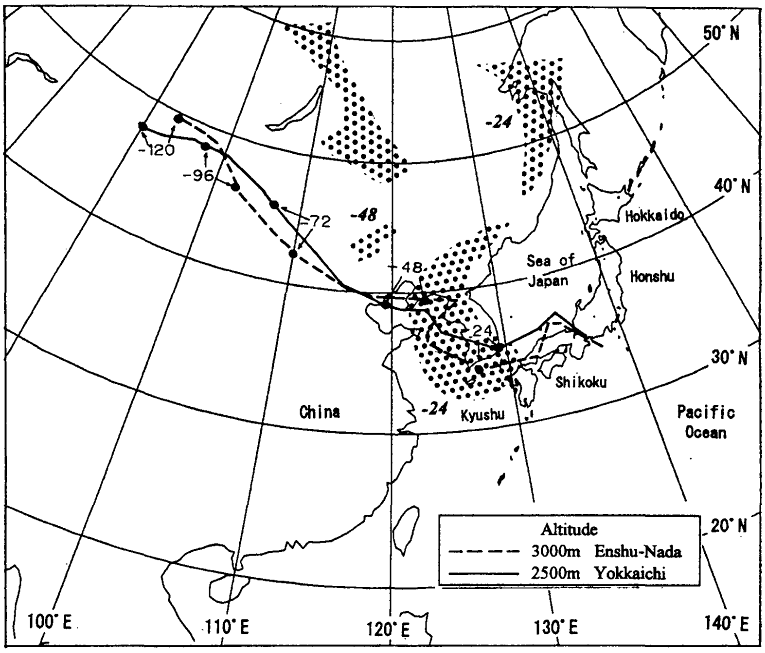

Fig. 9. Same as Fig. 3, but the backward trajectories from the 3000 m over Enshu-nada and 2500 m level over Yokkaichi on 15 December 1989.

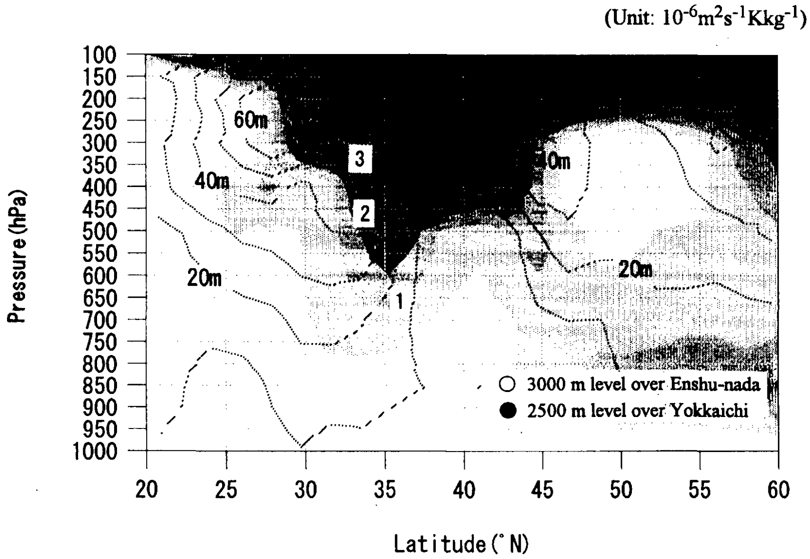

Fig. 10. Same as Fig. 4, but along 130°E at 9 JST on 14 December 1989 — 24 hours before the measurement.

tory passed through the high potential vorticity region ($\sim 1.4 \times 10^{-6}$ $m^2s^{-1}Kkg^{-1}$), consequently the ozone peak at this level also originated from the tropopause folding which occurred over the Yellow Sea on 20 January. As illustrated with the solid line in Fig. 5, the trajectory from the 2800 m level over Enshu-nada did not pass over the Yellow Sea 24 hours before the measurement but passed over the south part of China (105°E, 30°N) 96 hours before the measurement. Figure 6 is the cross section chart of the potential vorticity along 105°E at 9 JST on 17 January — 96 hours before the measurement. This cross section chart shows that a tropopause folding occurred over the south part of China (105°E, 30°N) on 17 January, and the open circle in this figure shows that the trajectory passed through the high potential vorticity region ($\sim 1.0 \times 10^{-6}$ $m^2s^{-1}Kkg^{-1}$). Hence, the broad ozone peak at the 2800 m level over Enshu-nada was affected by the other tropopause folding. The trajectory from the 1500 m level over Enshu-nada (dotted-dashed line in Fig. 5) shows that the air parcel

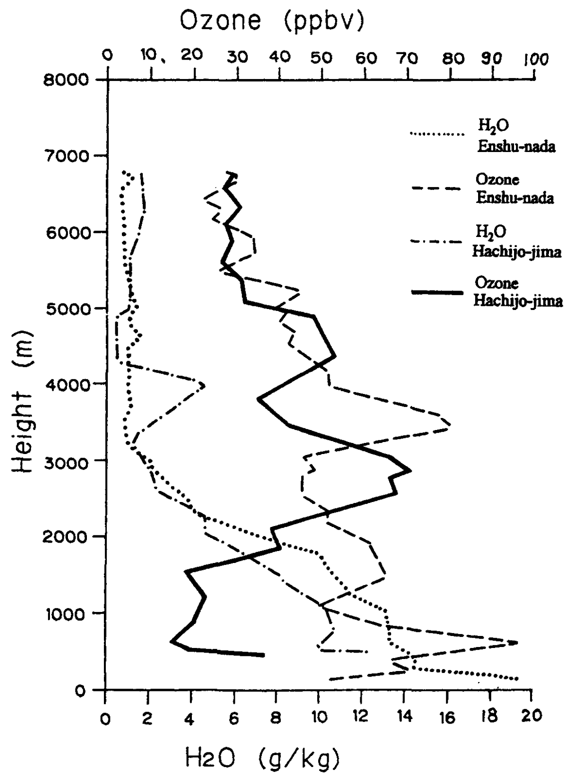

Fig. 11. The vertical distributions of the ozone and water vapor over Enshu-nada and Hachijo-jima on 9 August 1989.

passed over the north of Nagoya metropolitan and industrial area just before the measurement. No tropopause folding occurred around this trajectory, and the water vapor mixing ratio was high when below the 2000 m level over Enshu-nada (Fig. 2). For these reasons, the ozone peak at the 1500 m level was thought to be affected by the photochemical ozone production from the anthropogenic pollution. However, to confirm this, other data such as NOx and CO concentrations are necessary.

Consequently, the air parcels which contained a high concentration of ozone over Japan came from the stratosphere by tropopause folding in the west of Japan. Considering that the transported peak was broad, the ozone peaks diffused during the transport, building a stratified structure over Japan. Figure 7 is the cross section chart of the potential temperature from Wakasa Bay (35°38′N, 135°00′E) to Enshu-nada (33°45′N, 138°45′E) (see Fig. 1) on 21 January 1989. This cross section chart shows that the isentropic surface slowly descended from Wakasa Bay to Enshu-nada. The shape of the ozone peak at the 4600 m level over Wakasa Bay was similar with that of the ozone peak at the 4100 m level over Enshu-nada. Considering that the height difference of potential temperature between Wakasa Bay and Enshu-nada is approximately 500 m at the 4000 m level, the height difference of these peaks (~500 m) is reasonable. For these reasons, it is believed that these ozone peaks belonged to the identical ozone layer and the height difference of these peaks was due to the inclination of isentropic surface. Enshu-nada is 300 km distant from Wakasa Bay, so the horizontal scale of the tropospheric ozone layer was larger than 300 km in this case.

3.2 Flight B (15 December 1989)

Two days before this flight, a cyclone generated west of Japan and traversed over Japan, developing rapidly. This cyclone was located off the Pacific coast of the Tohoku district, and an anticyclone was located in the middle of China on the same day. The weather pattern was a typical winter monsoon type and a strong westerly covered Japan. The cyclone brought a cut-off low in the upper level of troposphere. This cut-off low also traversed over Japan with the cyclone.

Figure 8 shows the vertical distributions of ozone and water vapor over Enshu-nada and Yokkaichi. The vertical distribution of ozone over Enshu-nada had a sharp peak whose concentration was approximately 70 ppbv at the 3000 m level, and the water vapor mixing ratio at this level was lower than the detection limit of the instrument. Figure 9 shows that a large-scale tropopause folding occurred around the west and the south of Korea on 14 December — 24 hours before the measurement. The backward trajectory from the 3000 m level over Enshu-nada (dashed line) also passed over the south of Korea (128°E, 35°N) on the same day. Figure 10 is the cross section chart of the potential vorticity along 130°E at 9 JST on this day. The open circle in this figure shows that this trajectory passed through the high potential vorticity region ($\sim 1.6 \times 10^{-6}$ $m^2s^{-1}Kkg^{-1}$); consequently it appears that the high ozone peak at the 3000 m level over Enshu-nada originated from this tropopause folding.

There was an ozone peak whose concentration was 60 ppbv at the 2500 m level over Yokkaichi in Fig. 8. The solid line in Fig. 9 shows that the backward trajectory from the 2500 m level over Yokkaichi almost passed the same route as that from the 3000 m level over Enshu-nada 24 hours before the measurement when passing through the same high potential vorticity region ($\sim 1.6 \times 10^{-6}$ $m^2s^{-1}Kkg^{-1}$) as illustrated with the closed circle in Fig. 10. Hence, this ozone peak also originated from the tropopause folding that occurred on 14 December over the west and the south of Korea. Both of the ozone peaks — the 3000 m level over Enshu-nada and the 2500 m level over Yokkaichi — originated from the identical tropopause folding.

Austin and Midgley (1994) suggested that there was a high concentration of ozone in a 5 km layer above Tsukuba. They concluded that this thick ozone layer was caused by a low tropopause level

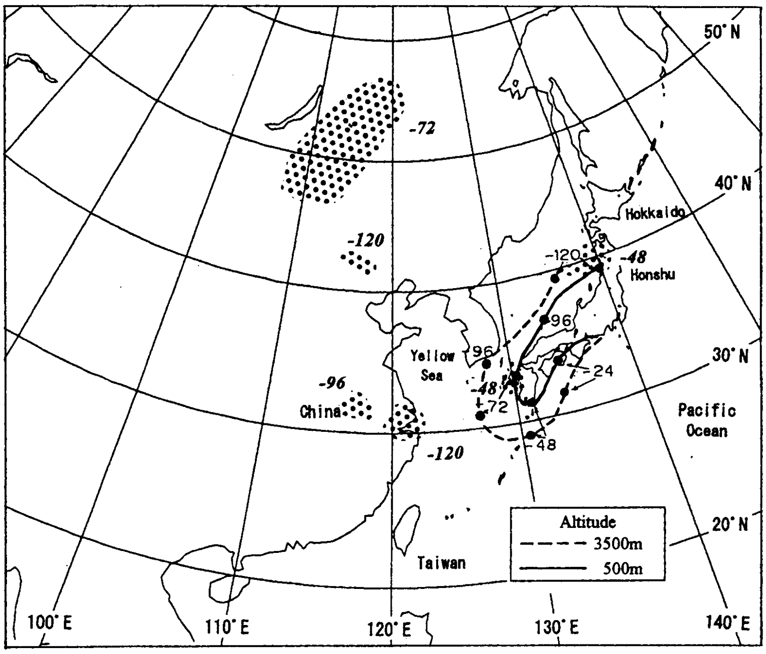

Fig. 12. Same as Fig. 3, but the backward trajectories from the 3500 m and 500 m levels over Enshu-nada on 9 August 1989.

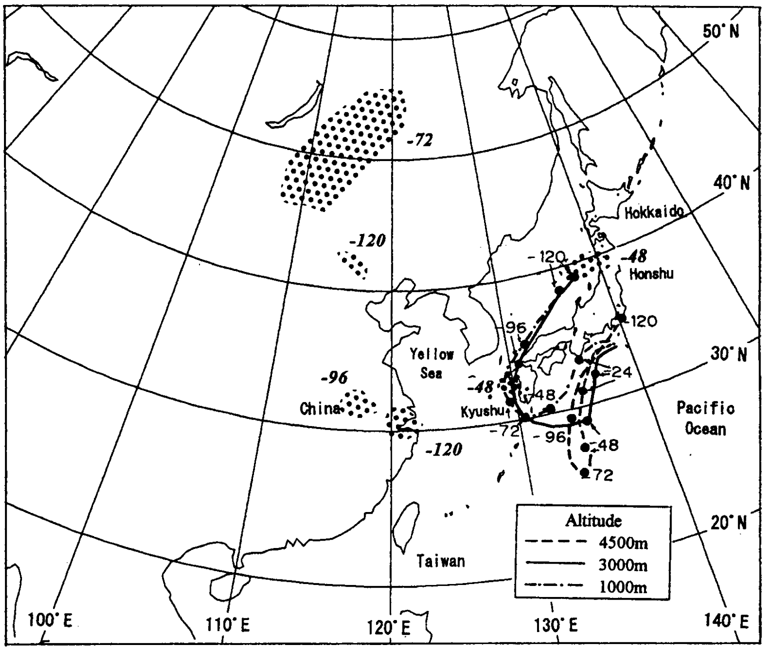

Fig. 13. Same as Fig. 3, but the backward trajectories from the 4500 m, 3000 m and 1000 m levels over Hachijo-jima on 9 August 1989.

with a cut-off low. The thickness of the ozone layers whose concentrations were higher than 40 ppbv over Yokkaichi and Enshu-nada were 2 and 3 km, respectively. As mentioned above, the weather chart at the 500 hPa level at 21 JST on 14 December showed that a cut-off low was located over Japan. Though this cut-off low resulted in a low tropopause level (~6030 m) at Hamamatsu (137°42′E, 34°45′N) which is close to Enshu-nada at 9 JST on 15 December 1989, this altitude was higher than those at Yokkaichi and Enshu-nada. Thus, these ozone layers originated from the tropopause folding rather than from the cut-off low.

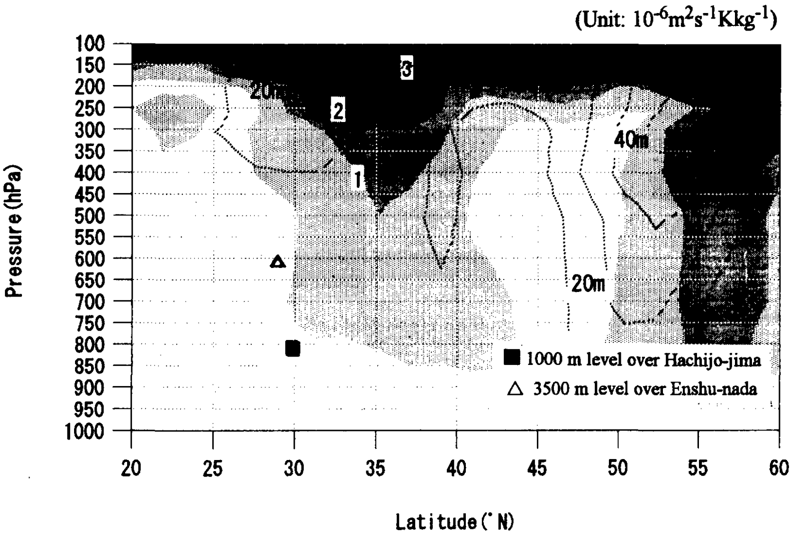

Fig. 14. Same as Fig. 4, but along 130°E at 9 JST on 7 August 1989 — 48 hours before the measurement.

3.3 Flight C (9 August 1989)

Two days before this flight, a typhoon passed over Japan. After that, it became a cyclone with a cold front to the north of Hokkaido. Though this cold front was passing through Hokkaido, other areas of Japan were clear and covered with an anticyclone on this day. It was a typical summer weather pattern in the middle part of Japan.

Figure 11 shows the vertical distributions of the ozone and water vapor over Enshu-nada and Hachijo-jima. Though the concentration of ozone increased between the 4000 m and the 5000 m levels over Hachijo-jima, the concentrations of ozone decreased when the altitudes were increasing above the 3000 m level over Enshu-nada and the 4000 m level over Hachijo-jima. Both of the concentrations of ozone from the 5000 m to the 7000 m levels were approximately 30 ppbv. On the other hand, the ozone profiles at the lower level were different. Namely, the concentration of ozone at the lower level over Enshu-nada was higher than 90 ppbv at the 500 m level, however that over Hachijo-jima was less than 20 ppbv. There was an ozone peak whose concentration was approximately 80 ppbv at the 3500 m level over Enshu-nada, and the water vapor mixing ratio was low (about 1 g/kg or lower than the detection limit) at this level. There were two ozone peaks at the 3000 m and the 4500 m levels over Hachijo-jima whose concentrations were approximately 70 ppbv and 50 ppbv and the water vapor mixing ratios were both below 2 g/kg.

Figure 12 shows the backward trajectory analysis from the 3500 m and the 500 m level over Enshu-nada and Fig. 13 shows the backward trajectory analysis from the 4500 m, 3000 m, 1000 m levels over Hachijo-jima. These figures also show that a high potential vorticity region was located at the west of Kyushu (130°E, 35°N) on 7 August 1989 −48 hours before the measurement. The backward trajectories from the 3500 m level over Enshu-nada and the 4500m and the 3000 m levels over Hachijo-jima passed to the south of this high potential vorticity region 48 hours before the measurement. On this day, a typhoon was located in the northern part of Japan, on the other hand, an anticyclone was located on the east coast of China. Though Danielsen (1980) pointed out that the tropopause folding occurred over the Pacific Ocean even in summer, this case was not caused by the tropopause folding because the location of the jet stream was far north of Japan (see Figs. 14 and 15). As noted above, the typhoon was located over the northern part of Honshu (140°E, 40°N) on 7 August, thus the subsidence occurred around the eye of the typhoon (see Fig. 12), as pointed out by Palmén and Newton (1969). However, this subsidence area was too far from the trajectories. Figure 14 is the cross section chart of the potential vorticity along 130°E at 9 JST on this day and Fig. 15 is same as Fig. 14 but along 135°E. These figures show the high potential vorticity regions fell down to the lower layer around 35–40°N. The typhoon brought a low at the 500 hPa level over the western part of Japan, and a cold air flowed into this low from the north. This low at the upper level might bring the subsidence over Kyushu. Considering that the water vapor mixing ratio was low at the 3500 m level over Enshu-nada and the trajectory from there passed close to the high potential vorticity region (Fig. 14), the ozone peak at the 3500 m level over Enshu-nada might be affected by this subsidence. The situation was almost same for the trajectories from the 4500 m and the 3000 m levels over Hachijo-jima. However, two days before the flight, a typhoon passed over Japan. Then the

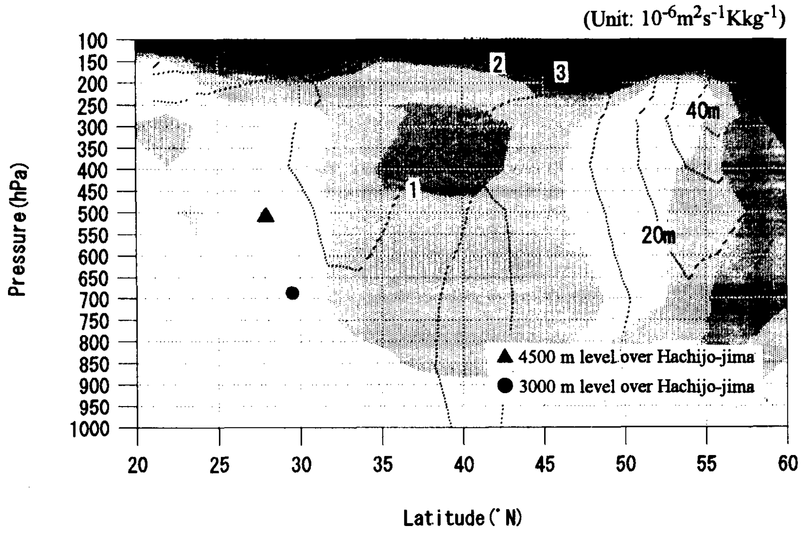

Fig. 15. Same as Fig. 4, but along 135°E at 9 JST on 7 August 1989 — 48 hours before the measurement. The symbols indicate the points where the backward trajectories from each research area passed through.

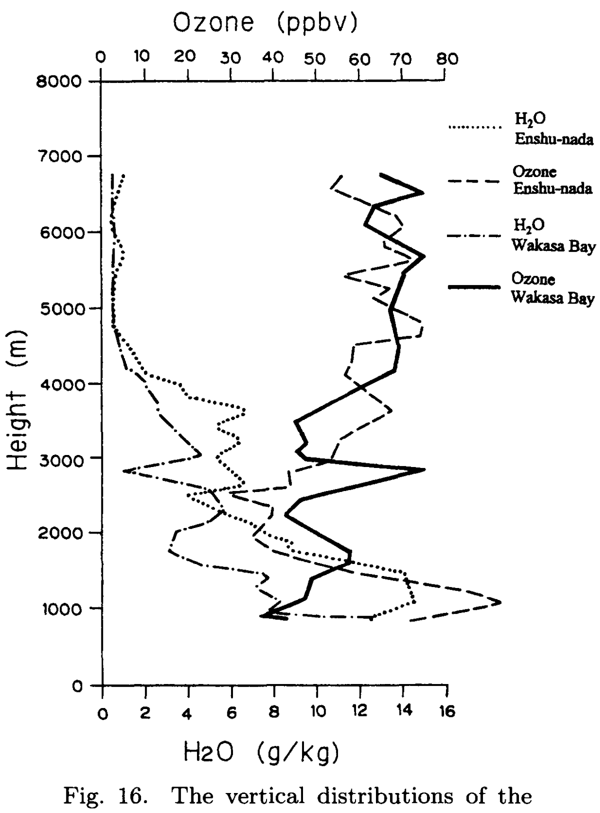

Fig. 16. The vertical distributions of the ozone and water vapor over Wakasa Bay and Enshu-nada on 8 August 1990.

adiabatic process might break down. Hence, trajectories older than 48 hours before the measurement possibly have an error. For this reason, we were not able to exclude the possibility that these ozone peaks over Hachijo-jima and Enshu-nada originated from photochemical production or the other tropopause foldings. Anyway, when the aircraft cruised back from Hachijo-jima to Osaka at about the 3000 m level, the air parcel which had a ozone concentration of approximately 90 ppbv and a low water vapor mixing ratio was observed over Hamamatsu with a horizontal scale of about 120 km. For certain there was an air parcel which had a high concentration of ozone and a low mixing ratio of water vapor over the middle of Japan on the south coast of that day.

There was a steep ozone peak which had a high ozone concentration of approximately 100 ppbv and a high water vapor mixing ratio ($\sim$14 g/kg) at the 500 m level over Enshu-nada (Fig. 11). As illustrated with a solid line in Fig. 12, the trajectory from the 500 m level over Enshu-nada passed over Osaka and Nagoya — the industrial and metropolitan area only 12 hours before the measurement, and the altitude of the trajectory was too low to be affected by the subsidence. The assumption that a given air parcel moves on a constant isentropic surface becomes doubtful in the lower layer where the diabatic effects from the radiative processes and turbulent mixing are important. However, we think that the isentropic trajectory is suitable when being applied over a short distance to this lower layer when the clear sky is over the sea because of no latent heat release and less turbulence. For these reasons, this ozone peak must be produced from the photochemical reactions of the anthropogenic gases in the metropolitan or industrial areas.

As illustrated in Fig. 11, the concentration of ozone at the 1000 m level over Hachijo-jima was very low (about 20 ppbv). Figure 13 shows the backward trajectory analysis from the 1000 m levels over Hachijo-jima. As illustrated with the dotted-

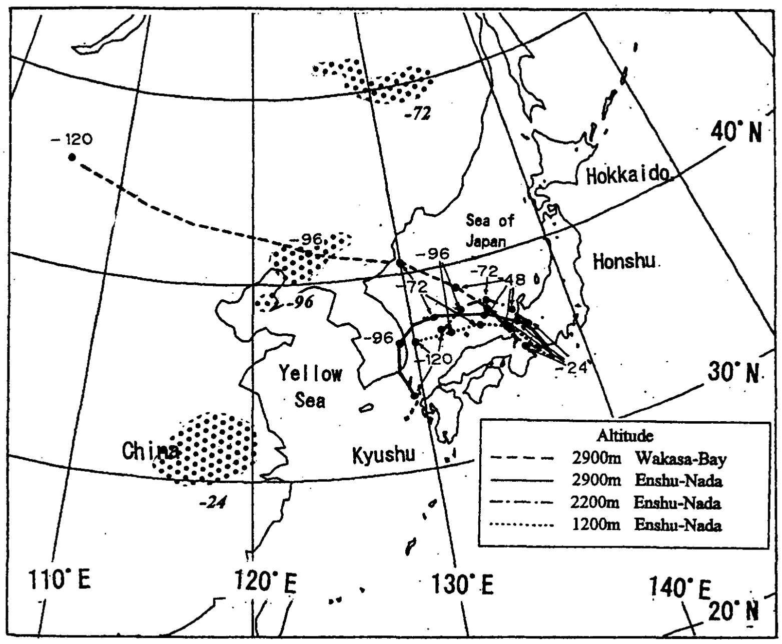

Fig. 17. Same as Fig. 3, but the backward trajectories from the 2900 m level over Wakasa Bay and the 2900 m, 2200 m, 1200 m levels over Enshu-nada on 8 August 1990.

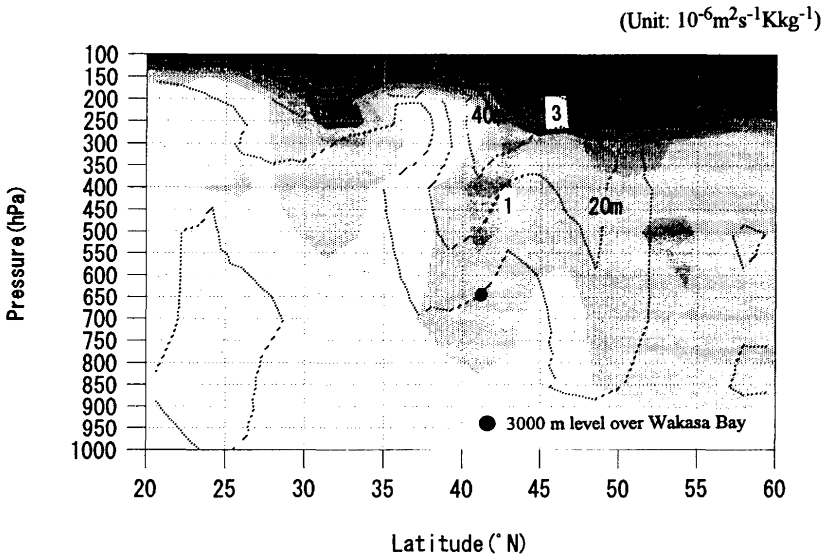

Fig. 18. Same as Fig. 4, but along 125°E at 9 JST on 4 August 1990 — 96 hours before the measurement.

dashed line in this figure, the trajectory from the 1000 m level passed over the south of Kyushu 48 hours before the measurement by essentially the same route as the 3500 m level over Enshu-nada mentioned above. The reason why the concentration at the 1000 m level over Hachijo-jima was low was that the altitude of the trajectory was too low to be affected by the subsidence, as illustrated with the closed square in Fig. 14, and the air parcel might have mixed with the oceanic ozone-poor air at this level.

Consequently, low ozone concentrations in the upper troposphere and high ozone concentrations in the middle troposphere were observed in this measurement. Considering the high ozone concentration above the tropopause, the vertical distribution over Hachijo-jima exhibits the same characteristic "S" shape as pointed out by Marenco and Said (1989) over the Atlantic Ocean in the Northern Hemisphere. Wakamatsu *et al.* (1983) suggested that there was a high concentration of ozone and other pollutants distributed about the 500 m$\sim$1 km level over industrial or metropolitan areas, and that they were locally advected by the land-sea breeze.

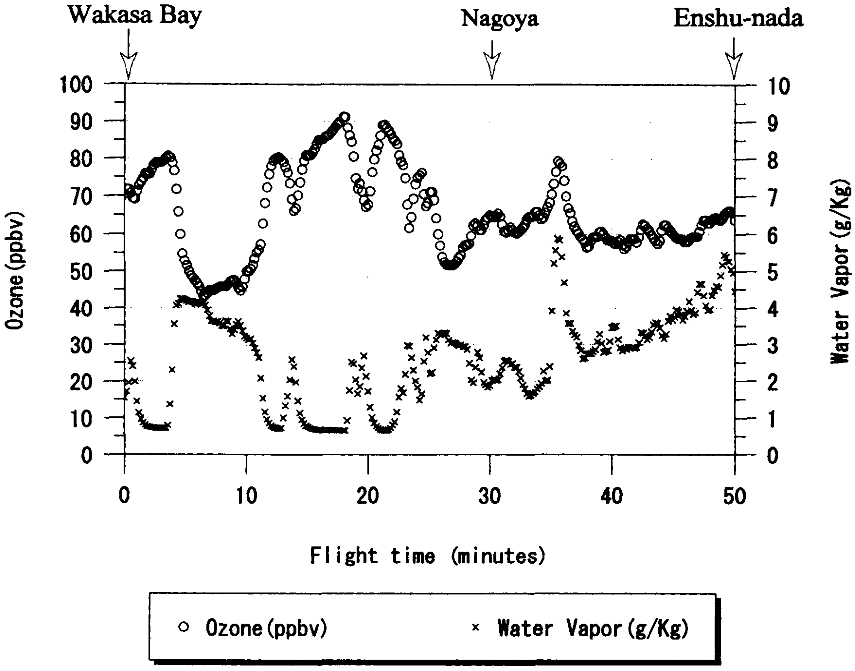

Fig. 19. The ozone and water vapor concentration data during the level flight from Wakasa Bay to Enshu-nada on 8 August 1990. The altitude is from 2800 m to 3500 m. It took about 30 minutes to reach Nagoya from Wakasa Bay.

The air parcel at about the 500 m level over Enshu-nada which must have been affected by the photochemical ozone production seemed to be advected by the synoptic scale motion.

3.4 Flight D (8 August 1990)

Four days before this flight, a typhoon stayed off the south coast of Japan for 48 hours. Though this typhoon moved slowly to the east the next day, another typhoon approached Japan from the south. Japan was covered with an anticyclone which had a center in the middle of the west Pacific Ocean but, on the other hand, there were two typhoons to the east and the south of Japan on the same day. Owing to the surface pressure pattern, which showed a saddle point over Japan, the wind speed was weak and the wind direction was changeable over Japan.

Figure 16 shows the vertical distributions of ozone and water vapor over Wakasa Bay and Enshu-nada. The vertical distribution over Wakasa Bay clearly shows an anti-correlation between the ozone and water vapor from the 1000 m to the 4000 m levels. Namely, the ozone-poor wet layers and the ozone-rich dry layers are stratified alternately. There was a steep ozone peak about 80 ppbv at the 2900 m level over Wakasa Bay. Figure 17 shows the backward trajectory analysis from the 2900 m level over Wakasa Bay and the 2900 m, 2200 m and 1200 m levels over Enshu-nada. As illustrated with the dashed line in this figure, the trajectory from the 2900 m level over Wakasa Bay passed over the northeast part of China

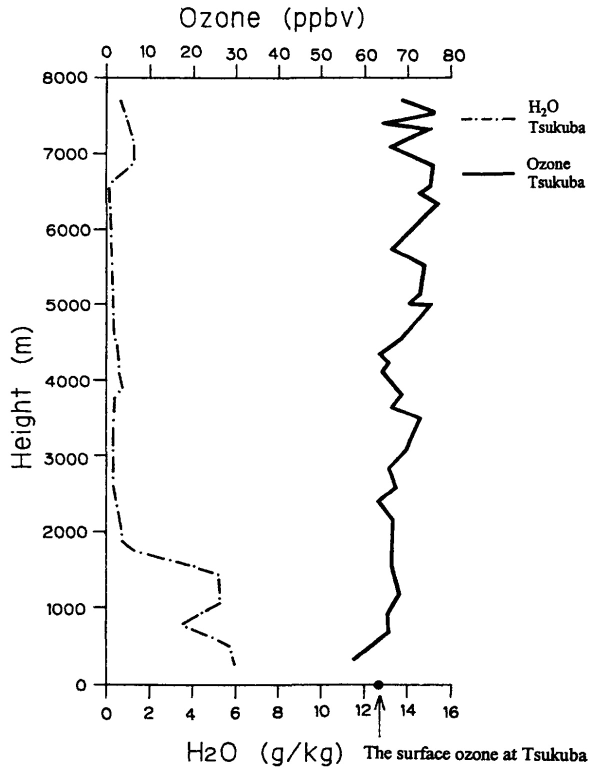

Fig. 20. The vertical distributions of the ozone and water vapor over Tsukuba on 27 April 1991. The surface ozone at Tsukuba was 63 pptv.

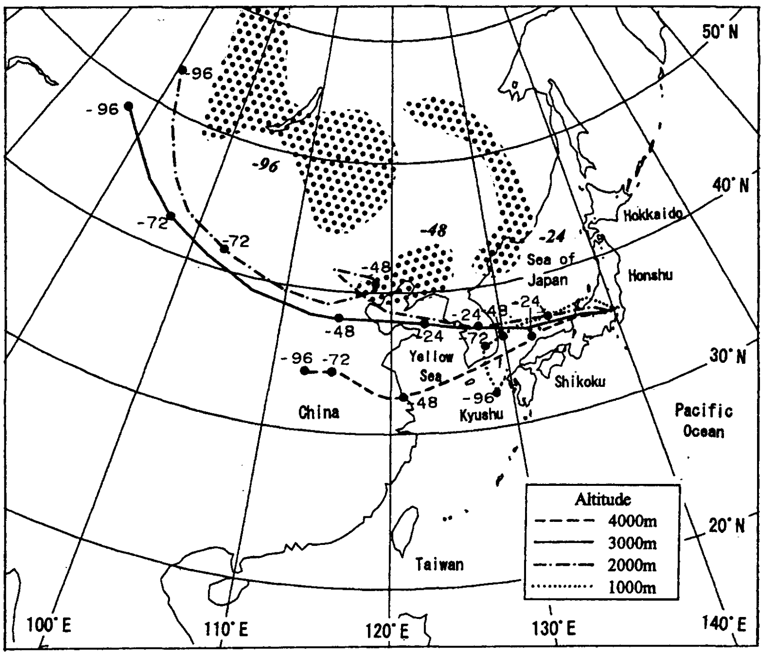

Fig. 21. Same as Fig. 3, but the backward trajectories from the 4000 m, 3000 m, 2000 m and 1000 m levels over Tsukuba on 27 April 1991.

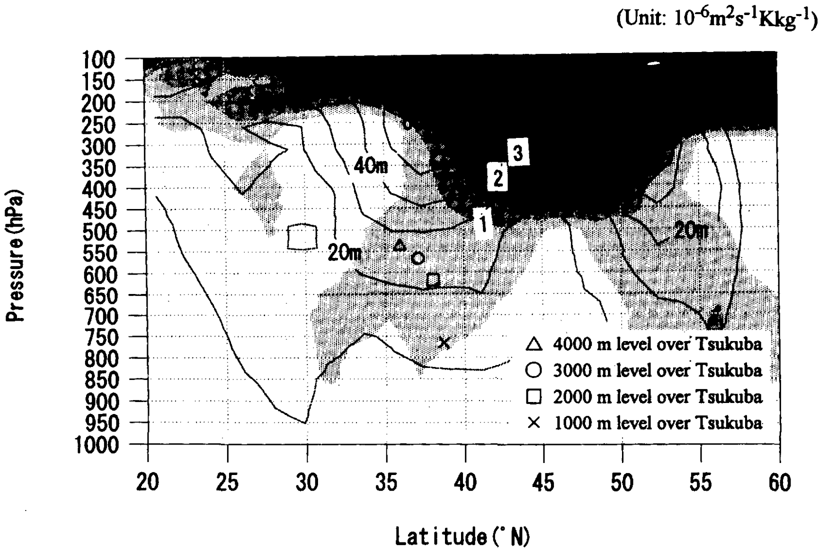

Fig. 22. Same as Fig. 4, but along 135°E at 21 JST on 26 April 1991 — 12 hours before the measurement.

(125°E, 42°N) on 4 August–96 hours before the measurement. Figure 18 is the cross section chart of the potential vorticity along 125°E at 9 JST on this day. The closed circle in this figure shows that this trajectory passed through a high potential vorticity region ($\sim 0.7 \times 10^{-6}$ $m^2s^{-1}Kkg^{-1}$) — just below the tropopause folding. The dry and ozone-rich air of this level originated from the tropopause folding which occurred over the northeast part of China.

The concentration of ozone at the 2200 m level over Enshu-nada was low (30–40 ppbv). However, the concentrations of ozone and the water vapor mixing ratio at the 1200 m level over Enshu-nada were both high (90 ppbv and 14 g/kg, respectively) in Fig. 16. As illustrated with the dotted-dashed line in Fig. 17, the trajectory from the 2200 m level over Enshu-nada where the ozone concentration was low, stayed over the Sea of Japan for 48 to 120 hours before the measurement. The concentration of ozone at this level over Enshu-nada is considered to be almost a background concentration, because there was no tropopause folding event during the transport and the trajectory was far from the polluted area. Tsutsumi *et al.* (1994) also suggested that

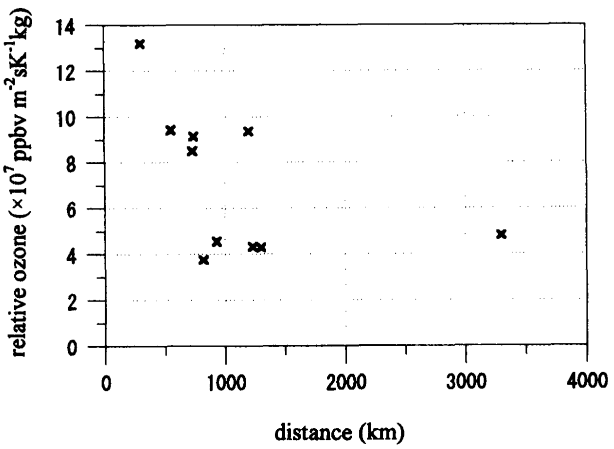

Fig. 23. The correlation between the relative ozone and the transport distance from the tropopause folding region where the trajectory passed. A relative ozone is defined as the peak concentration of ozone in the measurement divided by the potential vorticity where the trajectory passed through the folding.

there was a low concentration of ozone in summer at the top of Mt. Fuji. This contrast between the continental air mass over Wakasa Bay and the maritime air mass over Enshu-nada reflects that Japan is located in the rim region of these two kinds of air masses. As illustrated with the dotted line in Fig. 17, the trajectory from the 1200 m level over Enshu-nada passed over Nagoya metropolitan area 24 hours before the measurement. Considering the high ozone concentration, the high water mixing ratio and the trajectory that passed over the industrial area, this high ozone concentration is thought to originate from photochemical production.

Figure 19 shows the ozone and water vapor mixing ratio during the level flight at the 3000 m altitude from Wakasa Bay to Enshu-nada. When the aircraft cruised over Nagoya (about 30 minutes from Wakasa Bay), the correlation between the ozone and water vapor changed from negative to positive. As illustrated with the solid line in Fig. 17, the trajectory from the 2900 m level over Enshu-nada came from the north of Kyushu, passing over Nagoya. On the other hand, the trajectory from the 2900 m level over Wakasa Bay came from the Asian Continent, as mentioned above. In consequence, two different types of air masses covered central Japan at the same 3000 m level on 8 August 1990 and the border was located over Nagoya. One is the air parcel which showed negative correlation between the ozone and water vapor. This air parcel came from the Asian Continent directly and was located on the northwest side of Nagoya. The other is the air parcel which showed positive correlation between the ozone and water vapor. This air parcel passed over Nagoya and was located on the southeast side of Nagoya. This positive correlation must be attributed to the effect of the anthropogenic pollution in the boundary layer.

3.5 Flight E (27 April 1991)

Three days before this flight, a cyclone generated to the south of Kyushu. This cyclone traversed Japan, developing rapidly on 26 April. Though there was a stationary front to the south of Japan, Japan was covered with an anticyclone centered on the Sea of Japan. Almost all of Japan was clear on the flight day but it became cloudy in the late afternoon over Tsukuba.

Figure 20 shows the vertical distributions of the ozone and water vapor over Tsukuba on 27 April 1991. The surface ozone at Tsukuba was also measured simultaneously and was recorded at 63 ppbv in the afternoon on the same day. The vertical distribution of ozone over Tsukuba was approximately 70 ppbv and gradually decreased with decreasing altitude in this measurement. Figure 21 shows the trajectory analysis from the 4000 m, 3000 m, 2000 m and 1000 m levels over Tsukuba. These trajectories did not pass the folding regions except for the 2000 m level over Tsukuba −48 hours before the measurement. However, a tropopause folding occurred over the west part of Japan (135°E, 35°N) 12 hours before the measurement, as shown in Fig. 22 which is the cross section chart of the potential vorticity along 135°E at 21 JST on 26 April. The region where the potential vorticity strength was 0.5∼1.0 (10^{-6} $m^2s^{-1}Kkg^{-1}$) descended and spread widely from the 500 hPa to the 850 hPa level. As illustrated in Fig. 22, all of these trajectories from over Tsukuba passed through the region where the vertical inclination of the potential vorticity was small. The high concentration of ozone over Tsukuba must originate from this tropopause folding region. Thus, the vertical distribution of ozone is considered to be uniform.

3.6 Correlation of the concentration of ozone and the distance of transport

The concentration of ozone over Japan whose origin is in the stratosphere, depends on the strength of intrusion, the diffusion during the transport and the destruction by photochemical reactions with NOx. Since a distinct ozone peak in the troposphere can diffuse with the turbulent mixing in the vicinity of the fold (Shapiro, 1980), the concentration of ozone would be determined from the strength of folding and diffusion during the transport in the free troposphere. Here, a relative concentration was defined as the peak concentration of the measured ozone divided by the potential vorticity of the point where the trajectory from this peak passed through the

Table 2. List of the observed ozone peaks and their origins.

ozone peak				origin		
date	place	altitude (m)	ozone (ppbv)	kind	time before (hours)	place
21 Jan. 1989	Wakasa Bay	4600	56	folding	24	125°E, 35°N
21 Jan. 1989	Wakasa Bay	3300	56	undefined		
21 Jan. 1989	Wakasa Bay	1800	40	—	—	—
21 Jan. 1989	Enshu-nada	4100	60	folding	24	125°E, 35°N
21 Jan. 1989	Enshu-nada	2800	48	folding	96	105°E, 30°N
21 Jan. 1989	Enshu-nada	1500	65	pollution	6	137°E, 35°N
15 Dec. 1989	Yokkaichi	2500	60	folding	24	130°E, 35°N
15 Dec. 1989	Enshu-nada	3000	69	folding	24	130°E, 35°N
9 Aug. 1989	Enshu-nada	3500	80	subsidence	48	130°E, 29°N
9 Aug. 1989	Enshu-nada	500	95	pollution	12	136°E, 35°N
9 Aug. 1989	Hachijo-jima	4500	50	subsidence	48	135°E, 28°N
9 Aug. 1989	Hachijo-jima	3000	69	subsidence	48	135°E, 29°N
9 Aug. 1989	Hachijo-jima	1000	20	—	—	—
8 Aug. 1990	Wakasa Bay	2900	75	folding	96	136°E, 35°N
8 Aug. 1990	Enshu-nada	2900	45	—	—	—
8 Aug. 1990	Enshu-nada	2200	39	—	—	—
8 Aug. 1990	Enshu-nada	1200	91	pollution	24	137°E, 35°N
27 Apr. 1991	Tsukuba	4000	65	folding	12	137°E, 36°N
27 Apr. 1991	Tsukuba	3000	68	folding	12	132°E, 37°N
27 Apr. 1991	Tsukuba	2000	66	folding	12	135°E, 37°N
27 Apr. 1991	Tsukuba	1000	65	folding	12	138°E, 38°N

folding. Figure 23 shows a correlation between the relative concentration and the distance of transport from the point of the tropopause folding. A negative correlation can be seen in the figure, thus diffusion also plays an important role for the tropospheric ozone during the transport.

4. Summary

A lot of the ozone peaks were observed in the series of aircraft measurements during 1987–1991 over Japan, as listed in Table 2. Compared with the results of the tropical region (Tsutsumi *et al.*, 1991), the vertical distributions over Japan were complex. There were many ozone peaks, most of which were transported from the stratosphere to the troposphere by tropopause foldings to the west of Japan a few days before the measurement. The intense jet stream over Japan may produce many folding origin peaks, as pointed out by Austin and Midgley (1994). The horizontal scale of the tropospheric ozone layer over Japan was estimated to be larger than 300 km and some ozone peaks in different areas originated from the identical tropopause folding. The high concentration of ozone in the boundary layer is thought to originate from the ozone production by the photochemical reactions even when the observation points were a few hundred kilometers from the industrial or metropolitan areas. On the contrary, the air parcel showed a low concentration of ozone when the trajectory stayed over the sea for a few days without coming across the stratospheric air or passing over the metropolitan area. Two different types of air mass were observed simultaneously at the 3000 m level over central Japan in the summer measurement. One was the continental air mass which showed a high concentration of ozone and a low concentration of water vapor. The other was the maritime air mass which showed a low concentration of ozone and high water vapor. However, if the maritime air mass passed over the metropolitan or industrial areas, it showed a positive correlation; the concentrations of ozone and water vapor were both high. The distance of transport from the intrusion region was an important factor for the concentration of ozone over Japan.

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日本上空における対流圏オゾンの鉛直分布：オゾンピークの起源

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1987年から1991年にかけて行った日本上空の一連の航空機観測によって、若狭湾、遠州灘、その他いくつかの地域の上空で、対流圏オゾンの鉛直分布を観測した。そして観測されたオゾンピークの起源を探るために、それぞれのピークに対して流跡線解析とその流跡線付近の渦位分布を調べた。その結果、今回日本上空で観測されたオゾンピークのうちの多くは、日本西方で起こったトロポポーズフォールディングに起因していることがわかった。

1989年1月21日に遠州灘上空で観測された2つの異なった高度、濃度のオゾンピークは、別のトロポポーズフォールディングを起源としていた。そして輸送中に拡散されながら、日本上空で層状構造をなしていた。同じ日に、同一のトロポポーズフォールディングを起源に持つ似た形のオゾンピークが約300 km離れた地域で観測された。それらのオゾンピークは、高度は異なるが温位の傾きから、同じ対流圏オゾン層に属していたと考えられる。1990年8月8日に2つの異なった性質の大気が日本上空のそう離れていない2地点で観測された。1つは高濃度のオゾンと低濃度の水蒸気を含んだ大陸性の大気で、中国東北部から輸送されて来ていた。もう一つは、低濃度のオゾンと高濃度の水蒸気を含んだ海洋性の大気で、成層圏大気に出合わずまた都市域も通過せずに数日間海上を漂っていた。海洋性気団でも都市域や工業地帯を通過したものは、オゾンと水蒸気が正の相関、すなわちオゾン、水蒸気ともに高濃度を示した。1991年4月27日の筑波上空でのオゾンの鉛直分布はほぼ一様で70 ppbvの高濃度を示した。これは、筑波上空の大気が鉛直方向の渦位勾配が緩やかなフォールディング領域から来ており、しかもフォールディングが起こって間もなく輸送されてきたためであろう。オゾン濃度と輸送された距離の関係から、対流圏中のオゾンの分布には成層圏からの流入だけでなく、輸送中の拡散も重要であると考えられる。