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Scientific note

In-situ measurement of the ozone concentration in the Arctic Airborne Measurement Program 2002 (AAMP 02)

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Abstract: From 5 to 14 March, 2002, the Arctic Airborne Measurement Program 2002 (AAMP 02) was conducted on a round-trip flight between Nagoya, Japan, and Longyearbyen, Svalbard, via Anchorage and Barrow, U.S.A. and the North Pole using a chartered twin-jet aircraft, the Gulfstream-II (G-II). On board the G-II, *in-situ* measurements of the ozone concentration were carried out every 12 s in order to obtain information on air mass differences and advection. Vertical profiles of the ozone concentration observed over Longyearbyen agreed well with those observed by ozone sonde launched around the same time from Ny-Ålesund, about 100 km north of Longyearbyen. The ozone variations observed in the upper troposphere and lower stratosphere showed negative correlation with the CO_2 concentration, suggesting vertical displacements of air masses. However, it was also observed that the ozone concentration fluctuated considerably with little consistency with the meteorological field.

key words: stratospheric ozone, carbon dioxide, airborne measurement, arctic, transport

1. Introduction

The ozone concentration in the lower stratosphere and upper troposphere is determined by long-range advection from its source region, *in-situ* photochemical production/destruction, and troposphere-stratosphere exchange. Such processes produce the characteristic vertical profile of ozone in the atmosphere; low concentration (several tens ppbv) throughout the clean troposphere and abrupt increase over the tropopause. Since the lifetime of ozone is on the order of several months in the lower stratosphere and the ozone concentration increases rapidly with height over the tropopause, ozone is thought to be a suitable tracer of stratospheric air masses and their vertical motions. Therefore, various aircraft measurements with the ozone observation have been conducted to elucidate the tropopause folding structure (*e.g.* Browell *et al.,* 1987), air mass distinction near fronts (*e.g.* Bethan *et al.,* 1998), and tropospheric air transport into the lower stratosphere (*e.g.* Fischer *et al.,* 2002), for example.

In March 2002, the Arctic Airborne Measurement Program 2002 (AAMP 02) was conducted to investigate temporal and spatial variations of atmospheric minor constituents, ozone and CO_2 as well as other greenhouse gases and aerosols, using a chartered twin-jet aircraft "Gulfstream II (G-II)" (Yamanouchi *et al.,* 2003; AAMP 98, Shiobara *et al.,* 1999). The AAMP 02 flight was a round-trip between Nagoya (35°N, 137°E), Japan and Longyearbyen (78°N, 15°E), Svalbard via Kamchatka, Russia, Alaska, U.S.A., and the North Pole. *In-situ* ozone and CO_2 measurements were carried out continuously to detect air mass differences and horizontal and vertical advection of minor constituents along the flight route.

In this paper, spatial distributions of the ozone concentration during the flight of AAMP 02 and the relationship between the ozone distribution and meteorological field will be presented. These analyses and data can also be used as background information to help interpret the greenhouse gas and aerosol data obtained on board (Yamanouchi *et al.,* 2003).

2. Measurement system

Since a detailed description of our ozone measurement system on board the G-II aircraft has already been reported by Morimoto (2002), only brief and supplemental information will be given here. Sample air taken from the air inlet was, at first, introduced into a pre-pressurized pump (Enomoto Pump, FX7700) of which the inner walls and diaphragm were coated with Teflon sheet. The sample air pressurized above the cabin pressure of the G-II (about 850 hPa) was supplied to a three-port manifold made of Pyrex glass. A Dasibi-type ozone monitor (Dylec, model 1100), sucking in the air sample from the manifold by a diaphragm pump, determined ozone concentrations every 12 s. To obtain precise concentration data, variations and fluctuations of the air amount in the detector cell of the ozone monitor were compensated using pressure and temperature data of the air sample monitored at the manifold and at the detector cell, respectively. Tubing was made of Teflon except for the manifold to prevent ozone decomposition before the air sample was introduced into the ozone monitor. It was confirmed in our laboratory that the ozone concentration did not change in the pump and tubing of the system by analyzing an air sample of which ozone concentration was previously determined. The observed data and house keeping data, such as pressure and temperature of the air sample, were stored in a PCMCIA RAM card on the data logger (Morimoto, 2002) every 12 s. Also stored were GPS position and time data obtained by a GPS receiver.

The ozone monitor was calibrated by using the gas phase titration (GPT) system of the National Institute of Polar Research (Aoki, 1997) before and after the AAMP 02 flight. It was confirmed from repeated calibrations that there had been no drift in the zero point or span of the ozone monitor. Taking the accuracy of the GPT system, the temperature and pressure sensors, and the ozone monitor into account, the precision of the ozone measurement system was estimated to be within 2 ppbv (Morimoto, 2002).

3. Vertical profiles of ozone over Svalbard

Vertical profiles of the ozone concentration over Longyearbyen, Svalbard, observed on board G-II on 7 and 11 March are shown in Fig. 1. Also plotted in Fig. 1 are the ozone data obtained by balloon-borne ozone sondes launched from Ny-Ålesund, about 100 km north of Longyearbyen, by the Alfred Wegener Marine and Polar Institute (AWI), Germany, on the same days. As can be seen from these figures, the profiles are

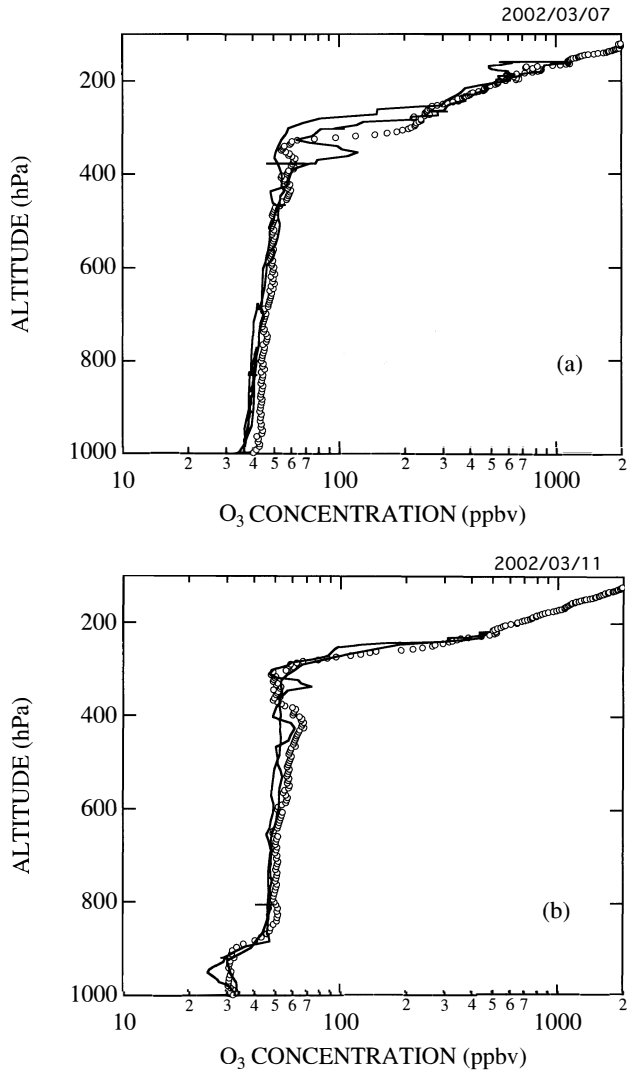

Fig. 1. Vertical profiles of the ozone concentration observed over Svalbard, Norway, on 7 March (a) and 11 March (b), 2002. Solid lines and open circles represent the ozone data obtained by our aircraft in-situ measurement system over Longyearbyen and by ozone sondes over Ny-Ålesund, respectively.

similar to each other for the two days. This fact shows that our ozone measurement system on board G-II worked well including the pressure and temperature compensations described in Section 2. It is noticeable in Fig. 1 that the ozone concentration measured by our system is several ppbv lower than AWI's value in the lower and middle troposphere on 7 March, although such a difference cannot be seen in the profile on 11 March. This difference might be ascribed to the air mass difference between Longyearbyen and Ny-Ålesund.

It is seen in Fig. 1 that the ozone concentrations in the lower troposphere below 900 hPa on 11 March are about 20 ppbv lower than those of the above layers. To examine the temporal and spatial variability of the ozone near the surface, a time series of the vertical profile of the ozone below 500 hPa obtained when the G-II departed from and arrived at the Longyearbyen airport from 6 to 12 March is shown in Fig. 2. From 6 to 10 March, the ozone concentration in the lower troposphere was stable around 35–40 ppbv. And then, the concentration decreased to about 30 ppbv below 900 hPa and increased to 45–55 ppbv above 850 hPa on 11 and 12 March. Surface ozone concentration observed in Ny-Ålesund (our unpublished data) showed rather constant values around 35–38 ppbv before 11 March and decreased to 20 ppbv on 11 and 12 March. Such a coincidence between Longyearbyen and Ny-Ålesund indicates that the cause of the ozone decrease could not be local ozone destruction but intrusion of an ozone depleted air mass into the lowermost troposphere over Svalbard on those days. Three-dimensional 5-day backward trajectories calculated by using HYSPLIT (Draxler and Rolph, 2003; Rolph, 2003) indicated that the air mass that arrived at altitudes of 850 and 925 hPa over Longyearbyen on 12 March was transported over the Arctic Ocean from eastern Siberia. This fact suggests that the ozone in the air mass had been

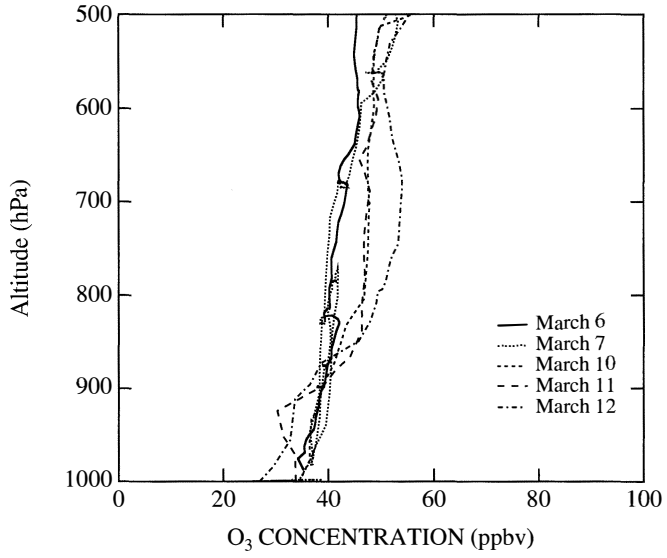

Fig. 2. Vertical profiles of the ozone concentration observed on board the aircraft over Longyearbyen from 6 to 12 March, 2002.

destroyed initially over eastern Siberia and/or during transport to Svalbard by interactions with sea-salt particles and sea-salts contained in surface snow over sea ice (*e.g.* Hara *et al.,* 2002).

4. Horizontal distributions of ozone observed on the long-range flight

The ozone concentration data observed during level flights from Nagoya to Longyearbyen and back are shown in Fig. 3, together with the pressure altitude of the flight, CO_2 concentration data measured on board G-II, potential temperature, and potential vorticity (PV). The potential temperature and PV were calculated using temperature and pressure data measured on board and using NCEP/NCAR reanalysis data, respectively. It was confirmed that the wind speed and temperature data described in the NCEP/NCAR reanalysis data were consistent with those measured on board G-II. For instance, the differences of the zonal and meridional wind speed and temperature data between the reanalysis data along the cruise track and those measured on board G-II were only 0.4 ± 3.6 m/s, -2.1 ± 2.8 m/s, and $+3.7 \pm 2.8$ K, respectively, on 5 March. Therefore, the PV values calculated with the reanalysis data have enough reliability for the following discussion.

As shown in Fig. 3, the ozone and CO_2 concentrations showed very similar variations with opposite signs throughout the AAMP 02 flight. The scatter diagram of the CO_2 and ozone concentrations during the level flights in AAMP 02 indicates a clear negative correlation between them, as shown in Fig. 4. A similar relationship was also observed on the AAMP 98 flight (Machida *et al.,* 2002). Since the sources and sinks of CO_2 are on the surface (land biosphere, ocean, and fossil fuel combustion) and CO_2 is chemically stable in the troposphere and stratosphere, the CO_2 concentration decrease with increasing height above the upper troposphere (Nakazawa *et al.,* 2002). On the other hand, as noted in Section 1, the source region of the ozone is mainly in the upper and middle stratosphere. Therefore, the negative correlation between the CO_2 and ozone variations reflects differences of the source region for both gases and the vertical displacements of air masses observed in the upper troposphere and lower stratosphere. In addition, it is suggested from the clear negative correlation that *in-situ* ozone production and/or destruction made little contribution to the ozone distribution during AAMP 02.

To acquire information about meteorological conditions during the AAMP 02 flight, the potential vorticity (PV) on the 340 K isentropic surface around the flight level (approximately 200 hPa) and contour lines of PV with the value of 3 to 4 PVU (10^{-6} m^2 s^{-1} K kg^{-1}) on 320 K on 5, 6, 12, and 14 of March are calculated using the NCEP/NCAR reanalysis data as shown in Fig. 5. PV is well conserved in the upper troposphere and lower stratosphere and could be used as a dynamical tracer (*e.g.,* Vaughan, 1988; Holton *et al.,* 1995). The contour lines in Fig. 5 present approximate positions of the polar jet stream, since the maximum gradient of PV was calculated around the value of PV between 3 and 4 PVU on the 320 K isentropic surface, as also shown by Fischer *et al.* (2002). In the following subsections, we will compare the ozone and CO_2 variations observed on board with the meteorological data.

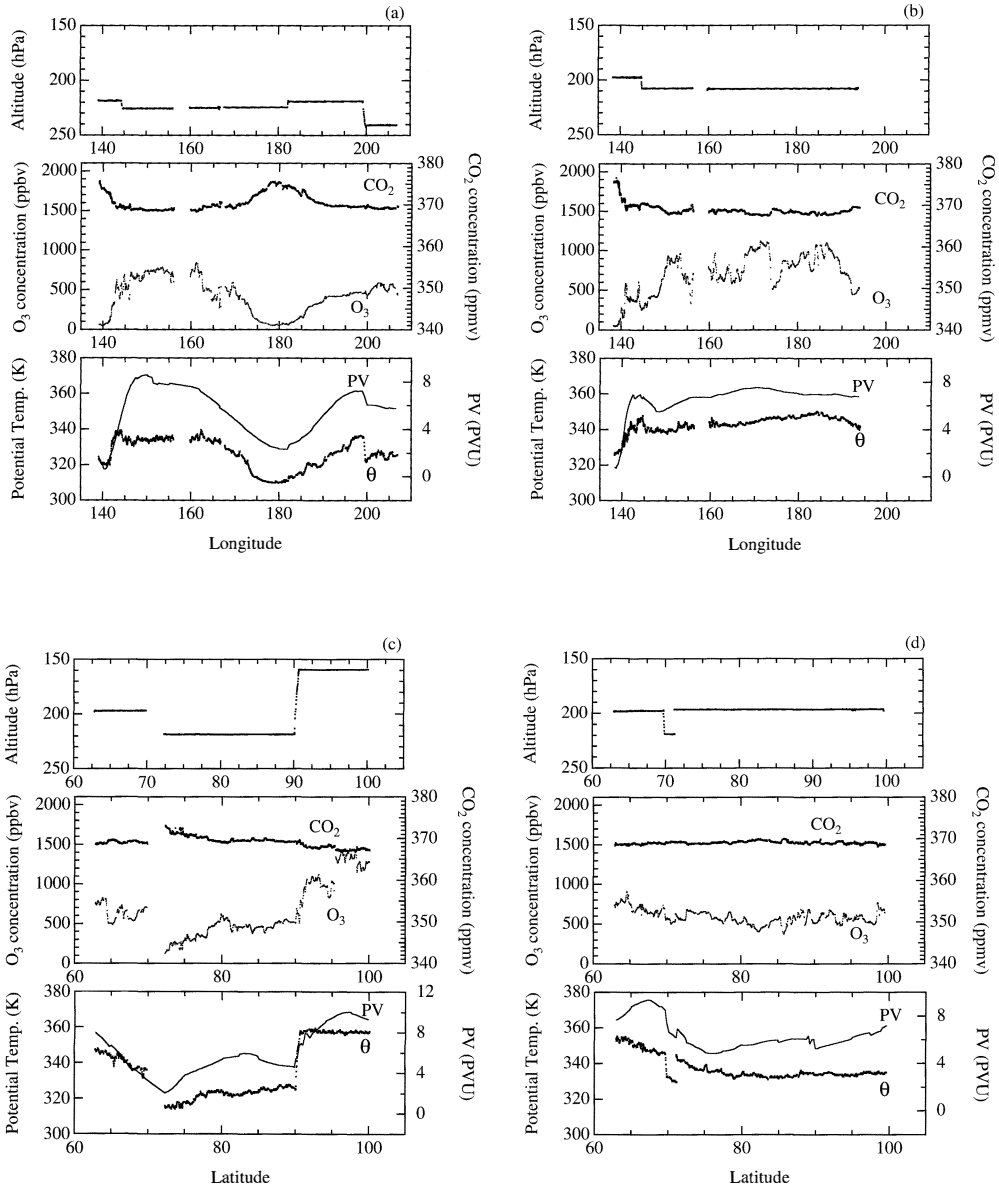

Fig. 3. Pressure altitude (top), ozone and CO_2 concentrations (middle), and potential temperature (noted as θ) and potential vorticity (bottom) obtained on the level flight in AAMP 02 from Nagoya to Anchorage on 5 March (a), Anchorage-Nagoya on 14 March (b), Anchorage-Longyearbyen on 6 March (c) and Longyearbyen-Anchorage on 12 March (d), 2002. The potential vorticities were calculated using NCEP/NCAR reanalysis data.

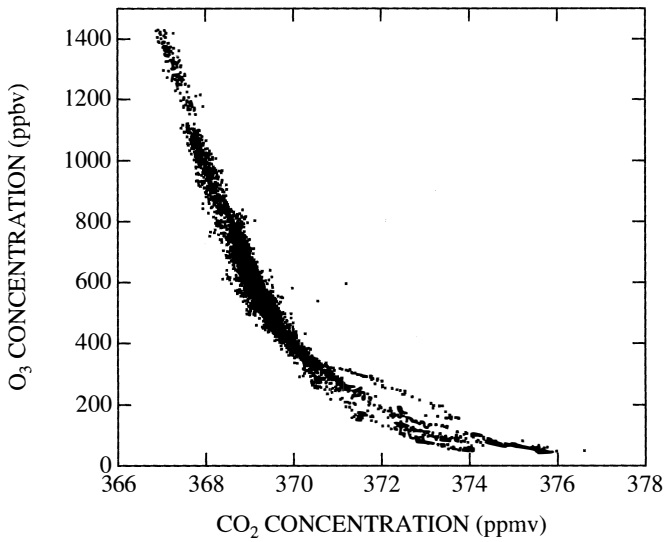

Fig. 4. Relationship between the ozone and CO_2 concentration observed on the level flights in AAMP 02 from 5 to 14 March, 2002.

4.1. Longitudinal distribution between Nagoya and Anchorage

The ozone and CO_2 concentrations obtained on the outward and homeward flights between Nagoya and Anchorage on 5 and 14 March, 2002 are presented in Fig. 3a and 3b, respectively.

In these figures, it is noticeable that the ozone and CO_2 concentrations changed rapidly near Japan around 142°E on 5 March and 140°E on 14 March, respectively; the tropospheric and stratospheric ozone concentrations were observed west and east of those longitudes, respectively. Since the flight routes cut across the polar jet stream at these longitudes as indicated in Fig. 5a, there could be tropopause gaps, which are between mid-latitudes and polar latitudes.

The most characteristic feature of the ozone and CO_2 variation on 5 March was their extremely low and high concentrations, respectively, observed around the longitude of 180° (date line) where the potential temperature and PV decreased. The lowest and highest values for the ozone and CO_2 concentrations, 58 ppbv and 375 ppmv, respectively, observed at 180° were close to those tropospheric values. The horizontal distribution of PV on the 340 K surface shown in Fig. 5a indicates that a large scale intrusion of a mid-latitude air mass into polar latitudes occurred around that longitude. Figure 6 presents 5-day backward trajectories from the flight route. It is clearly seen from this figure that the air masses observed at around 180° are transported from the troposphere (about 350 hPa).

In Fig. 5a, an equatorward tongue of high PV can be found at longitude of 150°E –180°. Inside the PV tongue, a region of higher PV is found at 145°–160°E along the flight route on which higher ozone concentrations were observed (Fig. 3a). Some diabatic processes could induce a downward displacement of the ozone-rich air mass

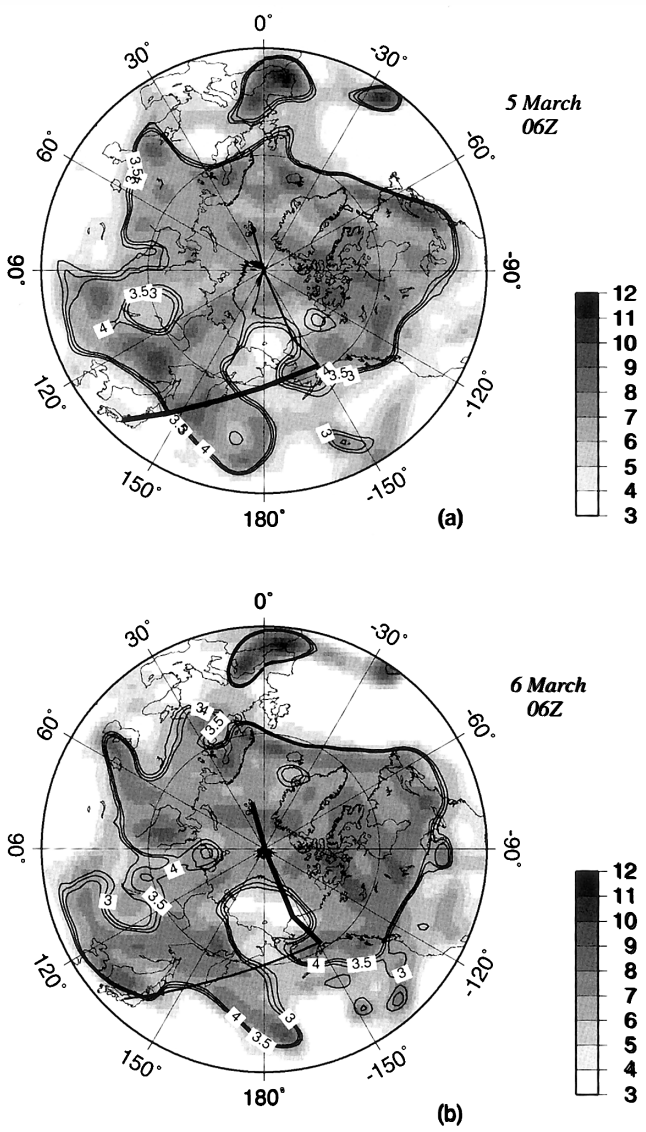

Fig. 5. Potential vorticities (PV) on the 340 K isentropic surface (shaded contours) at 06Z on 5 March (a), 06Z on 6 March (b), 18Z on 12 March (c) and 06Z on 14 March (d), 2002. Also shown are the contour lines of which PV values are between 3 to 4 PVU ($10^{-6}\,m^2 s^{-1}\,K\,kg^{-1}$) on the 320 K isentropic surface. Cruise tracks corresponding to PV maps are represented as bold lines.

from higher altitudes to the flight level at that longitude. The backward trajectories also indicated the downward transport of air mass at longitudes of 145–160°E (the lower panel of Fig. 6; blue lines).

The ozone concentrations on the flight from Anchorage to Nagoya on 14 March, 2002 showed more complicated variations (Fig. 3b), although potential temperature and

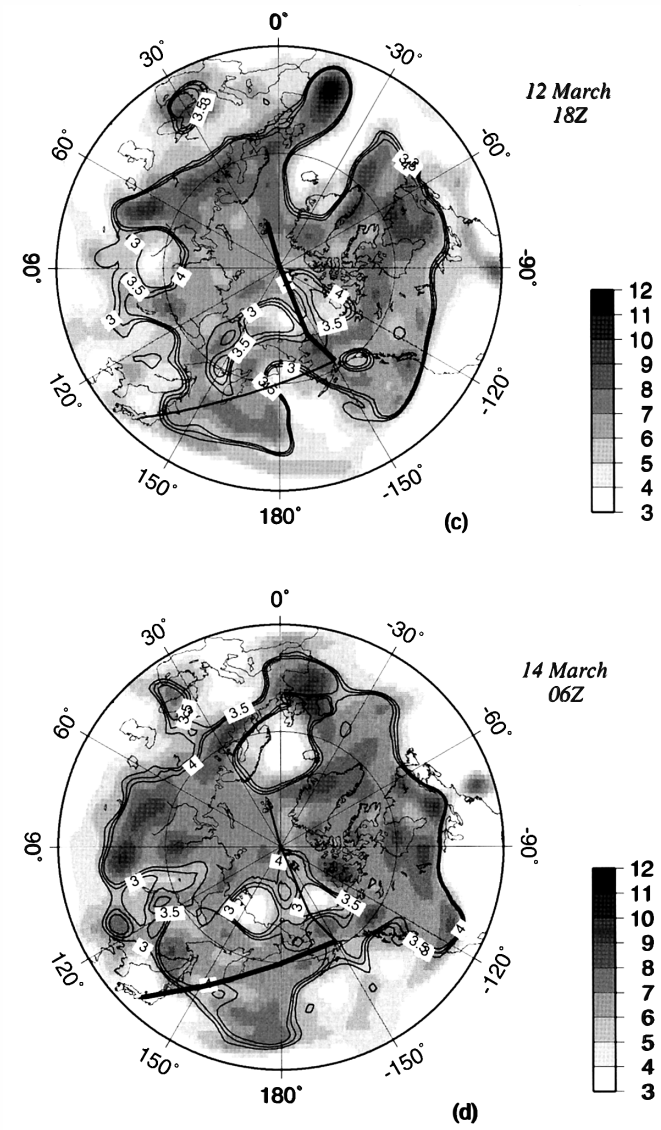

Fig. 5 (Continued).

PV along the flight track had been stable (Fig. 5d). In particular, a relatively low concentration of ozone and its abrupt increase in the westward direction were observed at 174°E; the ozone concentration increased from 510 ppbv at 174.2°E to 1100 ppbv at 173.4°E within a horizontal range of 50 km. The backward trajectories could not explain the ozone concentration variations along the cruise track; the air parcels that arrived at the flight level had stayed at altitudes around 200 hPa for the proceeding 5 days in the calculations. The ozone variations were correlated negatively with the CO_2 concentration variation also in the flight, and the relationship between the ozone and CO_2 variations were similar to those of other flights as shown in Fig. 4. Accordingly,

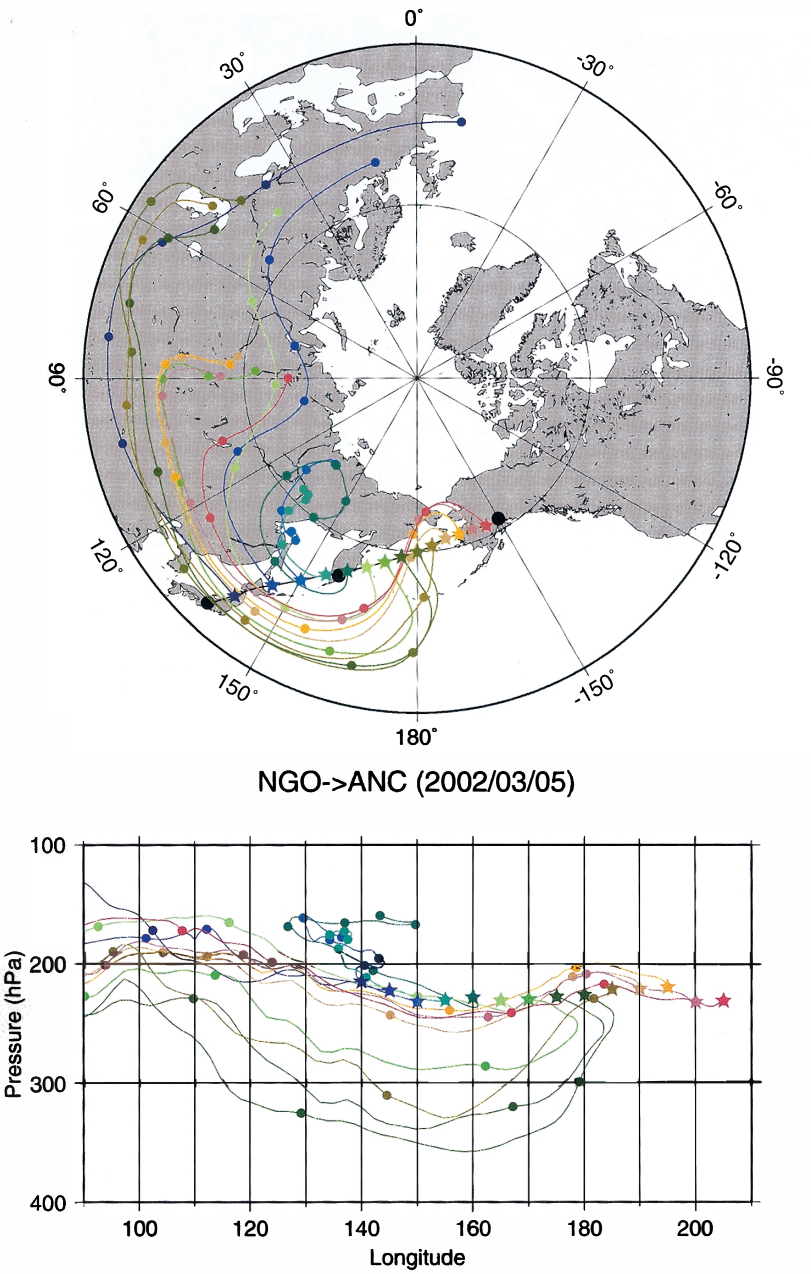

Fig. 6. Five-day backward trajectories from the flight route between Nagoya and Anchorage on 5 March, 2002. The lower panel shows an altitude-longitude cross section of the trajectories. Each star and closed circle represents a starting point of the calculation and the position of the air mass every 24 hours, respectively. Closed circles shown in black represent the positions of Nagoya, Petropavlovsk, and Anchorage.

the ozone variations showed that there could be vertical displacements of air masses. The inconsistency concerning the air mass movements estimated from the meteorological data and that from the trace gas data has not been solved yet.

4.2. Latitudinal distribution between Anchorage and Longyearbyen

The ozone and CO_2 concentrations observed from Anchorage to Longyearbyen on 6 March, 2002 and that from Longyearbyen to Anchorage on 12 March, 2002 are shown in Fig. 3c and 3d, respectively. In these figures, latitudes on the Europe side are shown as 90–100°N (actually 90–80°N) to prevent confusion with those on the Pacific side.

On 6 March, the ozone concentration increased gradually from 100 to 600 ppbv between 72°N and 80°N, where the potential temperature and PV increased and the CO_2 concentration decreased. As seen in Fig. 5b, poleward intrusion of lower latitude air, which was influenced by tropospheric air, have developed more than that of 5 March shown in Fig. 5a and arrived at the flight route at 65–80°N. Since the center of the intrusion along the flight route located around 72°N, the lowest concentration of ozone was observed at 72°N and the ozone concentration recovered toward the North Pole as the tropospheric influence diminished.

Abrupt increase of ozone from 900 to 1300 ppbv and decrease of CO_2 from 368.3 to 367.3 ppmv were observed at latitude 84.5°N on the Europe side, although the flight altitude and the potential temperature were constant, as shown in Fig. 3a. In addition, a sudden change of concentration occurred within a horizontal range of 20 km. As can be seen in Fig. 5c, a region of higher PV values was located between 85° and 80°N where the anomalous concentration changes were observed. Therefore, advection of an ozone-rich and CO_2-depleted air mass into the latitudes along the higher PV region could be responsible for the concentration anomalies. Such an air mass might be produced by downward transport of air from higher altitude into the lower stratosphere at another location.

In the flight from Longyearbyen to Anchorage on 12 March, the ozone and CO_2 concentration showed smaller variations than on other days in AAMP 02 as a whole (Fig. 3d). The potential temperature, ozone, and CO_2 concentrations correlated well with each other; potential temperature and ozone concentration gradually increased, and CO_2 gradually decreased, from 80°N to 65°N on the Pacific side. The PV distribution on the 340 K surface shown in Fig. 5c indicates that the poleward intrusion of the lower PV air mass was disappearing. These facts suggest that the lower stratosphere along the flight route became calm and well mixed by 12 March. It is confirmed by the backward trajectories that the air mass observed on board had mostly stayed around altitude 200 hPa and latitude north of 60°N for the preceeding 5 days (not shown).

5. Concluding remarks

The Arctic Airborne Measurement Program 2002 (AAMP 02) was conducted using a chartered twin-jet aircraft from Nagoya, Japan to Longyearbyen, Svalbard from 5 to 14 March, 2002. In the AAMP 02, *in-situ* ozone measurement was carried out throughout the flight as well as continuous CO_2 concentration measurement

(Yamanouchi *et al.,* 2003). The time series of the vertical profile of ozone over Longyearbyen from 6 to 12 March, 2002, showed an intrusion of ozone-depleted air mass into the lowermost troposphere on 12 March, suggesting that the ozone had been destroyed during its transport over the Arctic Ocean. The ozone variations during the level flight in the upper troposphere and lower stratosphere have a reasonable correlation with those of potential temperature and PV at the flight level. However, it was found in the flight between Anchorage and Nagoya on 14 March that large fluctuations of ozone occurred without the corresponding potential temperature and PV variations. To clarify the cause of the ozone variation, it is necessary to do more precise analyses together using vapor and aerosol data obtained on board.

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