Atmos. Chem. Phys., 3, 1541–1549, 2003
www.atmos-chem-phys.org/acp/3/1541/

Atmospheric Chemistry and Physics

# Rarity of upper-tropospheric low $O_3$ mixing ratio events during MOZAIC flights

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Received: 8 February 2003 – Published in Atmos. Chem. Phys. Discuss.: 24 March 2003
Revised: 21 September 2003 – Accepted: 25 September 2003 – Published: 29 September 2003

**Abstract.** Only a few previous observations of very low $O_3$ mixing ratios in the upper troposphere are available. The aim of this study was to examine the rich MOZAIC data set for more. Flights with at least 25 4 s averaged mixing ratios less than 8 ppbv at pressures lower than 500 hPa measured using commercial aircraft within the MOZAIC project have been analysed. There are eleven flights that fulfil these conditions (excluding artefacts as discussed below), representing about 0.001% of all measurements during the analysed period August 1994–December 1997. The low $O_3$ events occurred over Southeast Asia, Africa, Brazil and the sea area 200 km east of Florida (US) and were all likely to be associated with transport of air masses from tropical sea areas. These low mixing ratio events occur in the upper troposphere during periods with generally low mixing ratios. They are not only found over sea, but also over land at pressure levels as low as 179 hPa. It could well be that some of the low $O_3$ mixing ratio events measured during two or more flights belong to the same bigger low $O_3$ mixing ratio area.

## 1 Introduction

$O_3$ is central to the oxidizing capacity of the troposphere and is an intensively studied trace gas. For that reason it is important to know and understand the atmospheric distribution of $O_3$. In polluted areas emissions of $NO_x$ and hydrocarbons lead to in situ $O_3$ production. In the marine boundary layer far from anthropogenic and biomass burning sources, however, mixing ratios of $NO_x$ are so low that $O_3$ is destroyed (Singh et al., 1996). Kley et al. (1996) found not only low mixing ratios in the marine boundary layer in the equatorial Pacific, but also extremely low mixing ratios in the upper troposphere. While the photochemical lifetime of $O_3$ in the tropical marine boundary layer is less than 1 week, it increases rapidly with height to about 1 month at 6 km and 1 year at 10 km (Kley et al., 1996; Lawrence, 1996). For that reason one can expect that sometimes air parcels containing little $O_3$ formed in the tropical marine boundary layer are lifted up by convection and are then transported from tropical sea areas to other areas without large changes in mixing ratio, even to mid-latitude areas (Davies et al., 1998). Other possible explanations of very low upper tropospheric $O_3$ mixing ratios are catalytic destruction of $O_3$ by reactive iodine and bromine species that originate from the marine boundary layer (Kley et al., 1996; Platt, 2000) or $O_3$ destruction associated with the dissolution and dissociation of $HO_2$ in cloud droplets or possibly on the surface of ice particles followed by reaction with $O_3$ (Kley et al., 1996). The last possibility may explain that low upper tropospheric $O_3$ mixing ratios are sometimes associated with cirrus clouds (Reichardt et al., 1996).

Up to date there has been little systematic information about low $O_3$ mixing ratios in the upper troposphere. The aim of this paper is to make use of the extensive upper tropospheric $O_3$ observations collected by the MOZAIC project to see how frequently low $O_3$ levels are encountered in the upper troposphere and to investigate their origin. Within the MOZAIC project ("Measurements of Ozone and Water Vapour by Airbus In-service Aircraft"; Marenco et al., 1998; http://www.aero.obs-mip.fr/mozaic/) automated measurements are made on board five commercial aircraft. The aircraft are measuring the $O_3$ mixing ratio, relative humidity and temperature as a function of the pressure and the geographical position. Very recently also CO and $NO_x$ monitors were installed on board of some of these aircraft. The aircraft are based in Europe and have in the period studied (August 1994 - December 1997) being flying to North America (46% of the flights), South America (18%), Africa (6%), Southeast Asia (13%) and China and Japan (16%).

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## 2 Instrumentation

The instrumentation consists of an $O_3$ monitor and a relative humidity/temperature monitor. The inlet for $O_3$ is at 7 cm distance from the skin, towards the front of the aircraft, and is well outside above the aircraft's boundary layer. It consists of a 6 mm stainless steel tube that is coated on the inner side with Teflon. The inlet system is periodically checked, cleaned or replaced to prevent destruction of $O_3$ by deposits collected in the tube. Pumps are used to pressurise the air to cabin pressure (700 hPa at cruise altitude). A dual beam UV photometer with two separate absorption cells and detectors is located in the electronic compartment within the aircraft and is used to measure $O_3$ (Thermo-Electron, Model 49-103). The response time is 4 s. The photometers operate 180° out of phase, i.e. when the first cell contains the zero mixing ratio, the second cell contains the sample or reference mixing ratio and vice versa. The measurements are corrected for variations in the light intensity and for variations in pressure and temperature. The detection limit of the system is 2 ppbv and the overall precision is ±(2 ppbv+2% of the measured mixing ratio). At cruise level the outside temperature is of the order of −50°C and the pressure is about 200 hPa. When the air used for $O_3$ measurements is brought to cabin conditions (700 hPa, 15°C) it attains a very low relative humidity due to the large temperature difference. The travel time from the inlet to the $O_3$ sensor is at maximum 2 s at cruise altitude, which strongly limits the possible $O_3$ destruction in the line. Zero $O_3$ air for the calibration is obtained by leading cabin air over a filter that destroys $O_3$. $O_3$ containing air for calibration is generated by an uv lamp. Before takeoff, about every 2 hours during the flight and after landing, reference mixing ratios of 0, 80 and 500 ppbv $O_3$ are measured in order to obtain information on potential drift of the instrument. Recalibration of the instruments in the laboratory shows that only in exceptional cases an instrument drift of more than 1% per year is observed. Detailed information on the $O_3$ measuring system and procedures can be found in Thouret et al. (1998).

The relative humidity and temperature are measured in situ outside the aircraft in a compact airborne sensing device ADFS2 (Aerodata, Braunschweig, Germany). The sensing elements in this device consist of a capacitative sensor with a hydroactive polymer film (Humicap-H from Vaisala, Finland) and temperature sensor (Pt100 resistor) and are mounted in an appropriate housing (Model 102 BX, Rosemount Inc., Aerospace Division, United States) with its own inlet (Helten et al., 1998). The signals are fed into a microprocessor-controlled transmitter unit (HMP230, Vaisala, 1993) and transferred to the MOZAIC computer on board the aircraft. The relative humidity and the temperature are calculated from the signals of the sensors. Due to the strong speed reduction in the sensor housing the sampled air is heated by adiabatic compression. The signals are corrected for this effect according to procedures described by Helten et al. (1998). Relative humidity values in excess of 100% are occasionally observed. These are most likely caused by a partial or complete evaporation of hydrometeors due to the adiabatic heating in the sensor housing.

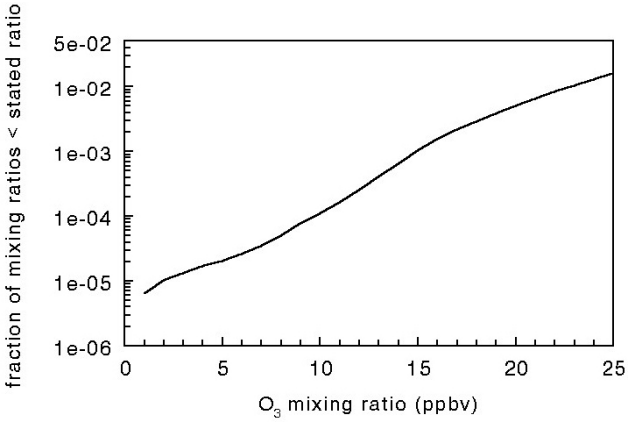


**Fig. 1.** Fraction of measured $O_3$ mixing ratios less than a stated mixing ratio for pressure levels lower than 500 hPa.

## 3 Observed low $O_3$ mixing ratio periods

MOZAIC measurements from August 1994–December 1997 were examined (7500 flights of on the average about 7 h, or 1000 times around the earth at the equator). We excluded measurements in the lower troposphere by selecting only events with a pressure lower than 500 hPa. On the average about 90% of the measurements were taken at pressure levels lower than 500 hPa. It was found that 2.3% of the mixing ratios at pressure levels lower than 500 hPa were less than 26 ppbv, 0.005% were less than 8 ppbv and 0.002% were less than 5 ppbv. Figure 1 shows the fraction of the $O_3$ mixing ratios at pressure levels lower than 500 hPa, which are lower than threshold mixing ratios of 1 to 25 ppbv; the fraction depends nearly log-linearly on the threshold mixing ratio. It should be noted here, that these statistics include very short periods with low mixing ratios as well as low mixing ratios that are caused by artefacts that are discussed below.

What we are interested in are not events with accidental low mixing ratios, but events with consistently low mixing ratios during part of the flight, enabling a meteorological analysis of this low mixing ratio period. The number of data in the MOZAIC database is so large that not all events with mixing ratios less than 26 ppbv can be analysed in detail. Minimum modelled mixing ratios computed for the tropical Pacific upper troposphere are 5 ppbv (Lawrence et al., 1999) and the precision in the MOZAIC $O_3$ measurements is ±2 ppbv at this mixing ratio. To find a reasonable number of flights that could be analysed we experimented with different selection criteria. We finally selected a reasonable number of flights when we chose flights with at least 25 mixing

**Table 1.** Position of low $O_3$ mixing ratio events during events with generally low mixing ratios

| Flight no. MOZAIC [a] | Long. (°E) | Lat. (°N) | Pressure (hPa) | No. of consecutive low mixing ratio measurements | Trajectories over tropical sea areas? |
|---|---|---|---|---|---|
| 5010501 | −79.2 | 26.5 | 474–500 | 14 | yes |
| 6010206 | −45.7 | −22.2 | 395–500 | 85 | likely |
| 6022804 | −46.3 | −22.6 | 462–500 | 69 | yes |
| 6041201 | 102.0 | 7.0 | 217 | 31 | yes |
| 6062101 | 101.0 | 4.1 | 217 | 30 | yes |
| 6062201 | 104.6 | 0.3 | 357–382 | 42 | yes |
| 6062205 | 105.8 | −4.8 | 369–398 | 33 | yes |
| 6062207 | 101.5 | 2.8 | 323–342 | 61 | yes |
| 6082701 | 102.2 | 13.0 | 179–452 | 39 | yes |
| 6082704 | 104.1 | 12.0 | 197–486 | 34 | yes |
| 7053105 | 28.4 | −15.1 | 254–262 | 20 | yes |

[a] First digit: year (e.g. 5=1995), next four digits are month, and day within the month and the last two digits are the flight numbers on that particular day.

ratio measurements integrated over 4 s below 8 ppbv at pressures below 500 hPa. Measured mixing ratios of 8 ppbv are still very low and not very different from the reported lowest modelled mixing ratios of 5 ppbv. We have analysed the data for the period August 1994 to December 1997, for which we also had access to back trajectories. The analysis was for that reason restricted to this period. The selection criteria are to some extent arbitrary, so we could e.g. also have chosen a level of 10 ppbv or a minimum of 10 low mixing ratio measurements, though we do not expect that this would change the main qualitative results obtained here, and we would then have to analyse many more events. Not all the selected flights showed periods with consecutive low mixing ratios. During some flights the mixing ratios showed high-frequency variations around the 8 ppbv level. These flights were excluded. During other flights there was more than one period with low mixing ratios and the number of measurements during all these periods was at least 25. Moreover, some events were excluded for which no good humidity and temperature measurements were available for the low $O_3$ mixing ratio part of the flight or for which the mixing ratios apparently were set to 0 ppbv or flights that showed other artefacts, such as a drop the $O_3$ mixing ratio to low levels during the adjustment following $O_3$ spikes of the type that is discussed in Suhre et al. (1997). In all 11 flights with low $O_3$ mixing ratios were selected at the end, of which the low mixing ratio part represents 0.001% of the measurements.

To interpret the measurements, five-day 3-D back trajectories were calculated for 2-minute intervals for each low mixing ratio period and for a period of 1 h before and after it (Scheele et al., 1996). This can show whether the low mixing ratio period could be associated with changes in the origin of the air mass. A length of five days was chosen for the back trajectories. Longer period back trajectories are more uncertain and even with five day back trajectories some care has to be taken in particular in convective regions. The data used in the trajectory calculations were 6-hourly data at a 1 by 1 degree spatial resolution from the ECMWF MARS archive. These trajectories describe the effects of convection at a larger scale realistically, but fail in resolving the effects of local convection. For one special event (b6010206) also 13-day back trajectories were calculated.

In one case images from the stationary METEOSAT satellite for visible light (VIS; 0.5–0.9 $\mu$m), infrared (IR; 10.5–12.5 $\mu$m) and water vapour (WV; 5.7–7.1 $\mu$m) were used to obtain additional information. The resolution of the images is about 5 km (for IR and WV) and 2.5 km (for VIS) at the subsatellite point, but a lower resolution at other points. Each 30 min a new image is retrieved, which leads to a relatively high temporal resolution (EUMETSAT, 1999).

Detailed information on the 11 flights is presented below, in Table 1 and in Fig. 2. The geographical area mentioned first below is the area where the low mixing ratio event occurred. Unless otherwise indicated the back trajectories before and after the low mixing ratio event came from about the same pressure level over the same area. Although one criterion for the selection of the events was that the pressure should be less than 500 hPa during some events the low $O_3$ mixing ratio starts at pressures higher than 500 hPa. In such cases the highest pressure at which the low $O_3$ mixing ratios occurred is mentioned.

### 3.1 Event 5010501 – Sea area 200 km east of Florida (US)

The low mixing ratio area was encountered while ascending from Miami at a pressure level of 643 hPa. It continued up to 474 hPa (a layer of about 2200 m). The horizontal extension of the area was about 70 km. The relative humidity was often higher than 100%, indicating the presence of clouds, but there was no relation between the $O_3$ mixing ratio and the

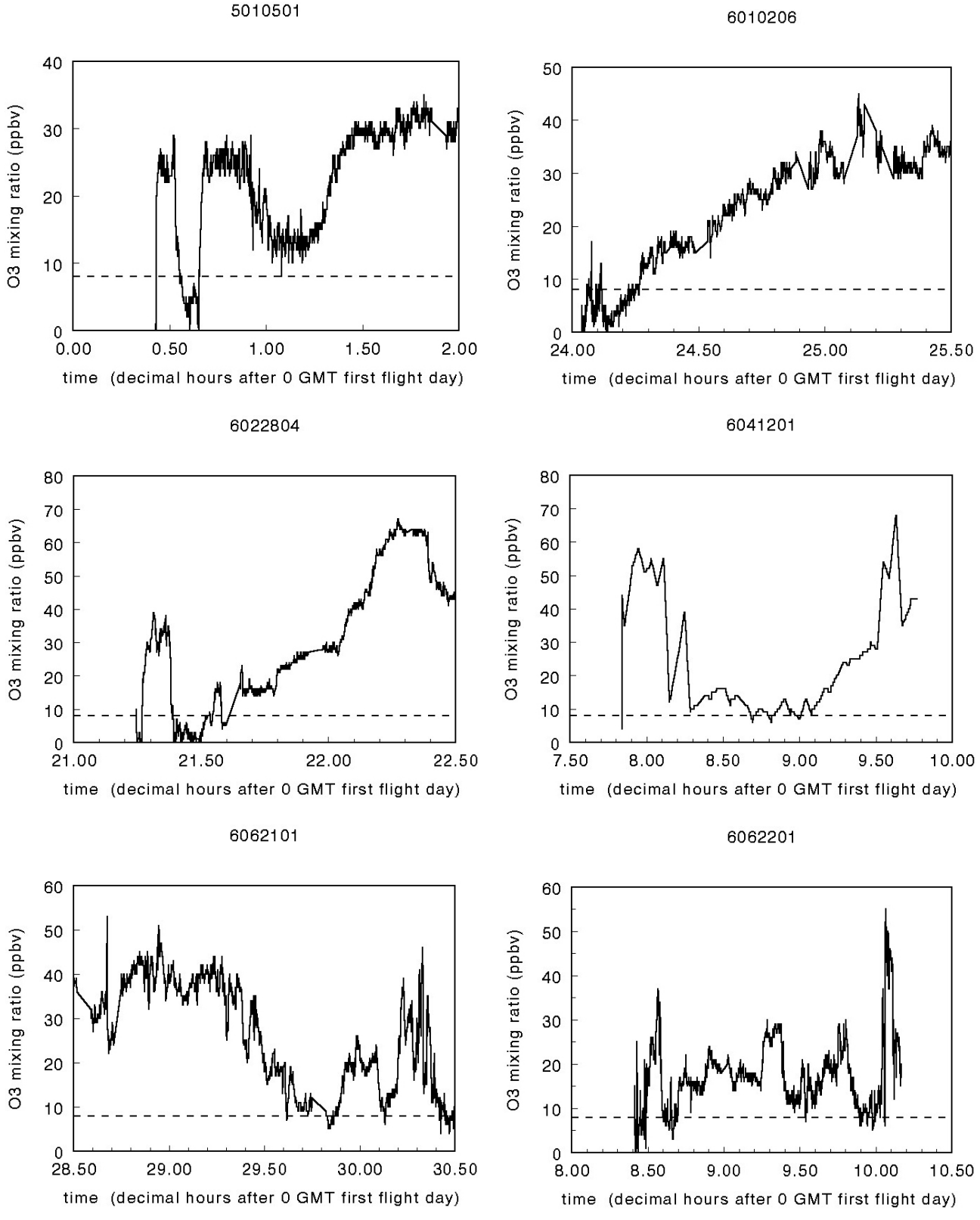


**Fig. 2.** $O_3$ mixing ratio vs. time for low $O_3$ events during flights with generally low mixing ratios. The dashed line indicates the 8 pbbv level. The data presented start some time before and end sometime after the low mixing ratio event. For that reason they may include measurements taken at pressures higher than 500 hPa. The flight number is indicated above the graphs.

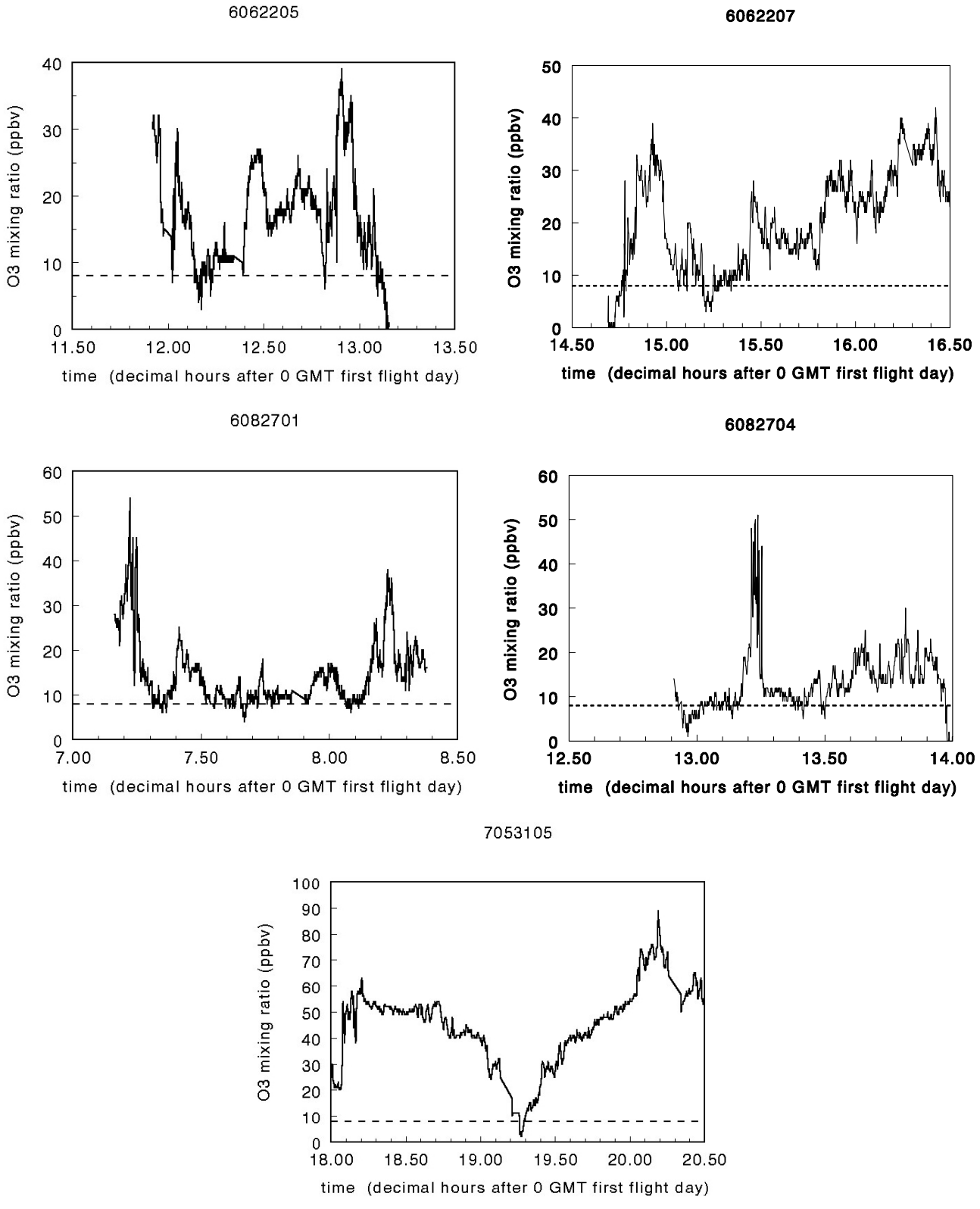


**Fig. 2.** Continued.

relative humidity. The back trajectory came over Caribbean Sea at pressure levels of 500–850 hPa.

### 3.2 Event 6010206 – Inland Eastern Brazil, 150 km from the coast

The aircraft was leaving from Sao Paulo and from 929 hPa (the lowest height at which measurements are available) up to a pressure level of about 395 hPa (about 7000 m height, about 200 km from the airport) the mixing ratios were below 8 ppbv most of the time. The relative humidity was higher than 100%, indicating the presence of clouds. The five-day back trajectories indicated that the air came at low speed over inland Brazil and Bolivia at pressure levels of 400–850 hPa. As this was the only low $O_3$ event trajectory for which the five day back trajectory did not come over a tropical sea area, also thirteen day back trajectories were calculated to get an impression of the possible route of the air mass further back in time. It appeared that two out of the three back trajectories that were associated with this event came over the tropical Atlantic Ocean at pressure levels of 800–1000 hPa about ten to at least thirteen days back. The other one was thirteen days back still somewhere over Brazil at similar pressure levels. It is therefore concluded that it is not unlikely that this air mass came over a tropical sea area, but that the uncertainty in the trajectory calculations is so large that no firm conclusion can be drawn.

### 3.3 Event 6022804 – Inland Eastern Brazil, 150 km from the coast

There were two periods with low mixing ratios. The low mixing ratios of the first period were observed at pressure levels of 619–453 hPa. The horizontal distance travelled during this low mixing ratio period was about 70 km and the vertical distance about 2300 m. The relative humidity was higher than 100% from 687–442 hPa, indicating the presence of clouds at about the same pressure interval as the low $O_3$ mixing ratios were observed. The second period occurred at pressure levels of 399–391 hPa and at a relative humidity of 35–40%. The horizontal distance travelled during this low mixing ratio period was about 13 km and the vertical distance about 130 m. During the first period back trajectories came at very low speed over sea and further backwards over land at pressure levels of 450–550 hPa. During the second period the trajectory came over the same area at pressure levels of 400–500 hPa.

### 3.4 Event 6041201 – Sea area east of Thailand and land area of Malaysia

There were three short low mixing ratio periods when the aircraft was flying at cruise level at 217 hPa. The low mixing ratio areas for these periods occurred over distances of 9, 15 and 5 km. The relative humidity was less than 100% during all low mixing ratio periods. During the first two periods back trajectories came over the Indonesian Archipelago as far as Irian Jaya at pressure levels of 150–250 hPa. During the last period the back trajectories came over Borneo and Celebes (Indonesia) at pressure levels of 200–600 hPa.

### 3.5 Event 6062101 – Malaysia

The aircraft was flying at a cruise altitude of 217 hPa when the low mixing ratio area was encountered and its extension was about 30 km. The relative humidity was less than 100%. The back trajectories came over the sea area east of Malaysia over the southern Philippines and the sea area east of the Phillipines at levels of 150–300 hPa.

### 3.6 Event 6062201 – Sea area east of Sumatra (Indonesia)

The aircraft was flying at a pressure level of 357–382 hPa during the event. The horizontal extension of the low mixing ratio area was about 12 km and the aircraft had a vertical displacement of about 500 m. The relative humidity was less than 100%. The back trajectories came over the sea area east of Sumatra, Celebes and Irian Jaya (Indonesia) at levels of 200–900 hPa.

### 3.7 Event 6062205 – Sumatra (Indonesia)

The low mixing ratio event occurred when the aircraft was descending (369 to 398 hPa). The horizontal extension of the event was about 14 km and the vertical travel distance was about 500 m. The relative humidity was lower than 100%. The back trajectories came over the sea area east of Sumatra, Celebes at pressure levels of 300–900 hPa. The back trajectories before the event came over the same area, but the back trajectory after the event came over the Indian Ocean.

### 3.8 Event 6062207 – Sea area between Sumatra (Indonesia) and Malaysia

The low mixing ratio area occurred when the aircraft was ascending from 342 to 323 hPa. The horizontal extension of the area was about 70 km and the aircraft was ascending about 400 m. The relative humidity was below 100%. The back trajectories came over the sea area east of Sumatra, Celebes and the sea area north of Irian Jaya at pressure levels of 250–700 hPa.

### 3.9 Event 6082701 – Thailand. Route from Bangkok to Saigon

While ascending a first low mixing ratio area was found at 456–435 hPa (a layer about 600 m thick). During the passage through this layer the aircraft was flying about 8 km in the horizontal direction. The second low mixing ratio area was found when the aircraft was flying at cruise altitude 181–179 hPa and had a horizontal extension of about 25 km. During both periods the relative humidity was below 100%.

During the first period with low mixing ratios the back trajectories came from the sea area south east of Vietnam at pressure levels of 400–1000 hPa. Before this period the back trajectories came from the sea area south of India. After this first low mixing ratio period the back trajectories came from the sea area south east of Vietnam (i.e. no change in origin). During the second period the back trajectories came over the sea area southeast of Vietnam at pressure levels of 200–900 hPa. Before this low mixing ratio period the back trajectories came from the same area but go further back to the Bay of Bengal. No change in back trajectory direction after this low mixing ratio period was observed.

### 3.10 Event 6082704 – Thailand

During event 6082704 the aircraft was flying from Saigon to Bangkok. While ascending the mixing ratios at pressure levels larger than 500 hPa were often below 8 ppbv. The first low mixing ratio that fulfilled our criteria occurred at pressure levels of 486–376 hPa (a layer of about 2000 m thick). During the passage through this layer the aircraft was flying about 60 km in the horizontal direction. The second low mixing ratio period occurred when the aircraft was flying at altitudes between 217 and 215 hPa. The horizontal extension of this area was about 6 km. The third low mixing ratio area occurred when the aircraft was flying at cruise altitude at 197 hPa and it had a horizontal extension of 15 km. The distance between the centre of this low mixing ratio area and the centre of the second low mixing ratio area of event 6082701 was about 30 km. During all periods the relative humidity was almost always below 100%. The back trajectories during the first period came over the sea area southeast of Vietnam and then back to the sea area north of Sumatra (Indonesia) at pressure levels of 400–800 hPa. For the second period the back trajectories came over the sea area south east of Vietnam at pressure levels of 200–500 hPa. For the third period the trajectories came also over the sea area south east of Vietnam at pressure levels of 200–600 hPa.

### 3.11 Event 7053105 – Zambia

During this flight the aircraft was at cruise altitude at a pressure level of 262 hPa when the low mixing ratio event occurred, but the aircraft had started to change to a new cruise altitude to 238 hPa when the low mixing ratio area was encountered. When the low mixing ratio area was left the pressure was reduced to 255 hPa. The area occurred when the aircraft was moving about 25 km in the horizontal direction and about 200 m up. Shortly before the change in height occurred, the aircraft had also changed flight direction, but it does not look like as does the pilot was trying to avoid an adverse situation, because the flight trajectory was not changed back to the original one. The turbulence had not increased much during the low mixing ratio event. The air temperature was about $-39$°C. The relative humidity was very low (about 1%) and satellite images indicate a temperature of the surface of about 15°C (infrared channel) and $-22$°C (water vapour channel). The water vapour channel mainly observes water vapour and a temperature of $-22$°C indicates that low clouds were present. The back trajectories followed the route Atlantic Ocean, South America and the Pacific at pressure levels of about 200–250 hPa. The low mixing ratio period was associated with a change in flight level from a pressure of 262 hPa to 238 hPa level.

## 4 Discussion and conclusions

Air masses with at least 25 $O_3$ mixing ratios, integrated over 4 seconds, with values less than 8 ppbv at pressures below 500 hPa were rarely encountered during the MOZAIC flights from August 1994 to December 1997 (about 0.001% of the measurements at pressures lower than 500 hPa). As the low mixing ratio events are rare, no general conclusions can be drawn on the frequency of occurrence of low mixing ratio events as a function of the geographical position and pressure level. This emphasizes the uniqueness of the low mixing ratio events.

The low $O_3$ events, characterized by generally low mixing ratios, was in 10 out of 11 cases associated with 5-day back trajectories that came over (sub)tropical sea areas (for the last case it was concluded that it was not unlikely that some of the back trajectories came over a tropical sea area 10–13 days before the event). It is, however, not so that the $O_3$ mixing ratio is always very low if the air comes over a tropical sea area. This is illustrated by the fact that the trajectories before and after the low mixing ratio events in most cases came over the same sea area, but had higher mixing ratios. This points towards a stochastic event such as deep convection as the cause of the low $O_3$ events, and this suggests transport of air masses from tropical sea areas where low $O_3$ mixing ratios have been found, not only in the boundary layer but also between 10 km and the tropopause (Kley et al., 1996). The 5-day back trajectories showed that the air masses associated with the low $O_3$ events sometimes came from the marine boundary layer and sometimes from the upper marine troposphere 5 days back. If air masses came from the boundary layer they have been lifted up and transported for several days in the upper troposphere. The question is then whether in general upper tropospheric air with low $O_3$ mixing ratios can be transported from the tropical sea areas to other regions without too much change in mixing ratio.

Though we cannot know the real chemical nature of the observed low-$O_3$ airmasses based on the limited measurements available, we can examine some constraints on the relationship between initial and observed conditions in the parcels. In the UT, the net chemical tendency for $O_3$ (production minus loss) is nearly always positive, averaging about 0.5 ppbv/day (see e.g. Fig. 6 in von Kuhlmann et al., 2003). Thus, it is likely that the initial mixing ratios in the air parcels

over their region of origin were even lower than the mixing ratios when they were observed by the MOZAIC aircraft, supporting the tropical MBL origin suggested by the back trajectories. Given the relatively small net chemical tendency in the UT (0.5 ppbv/day average, ~1 ppbv/day max), travel times of several days would easily be possible for intact parcels (no mixing), which start off with typical tropical MBL $O_3$ mixing ratios of a few ppbv. Transport of parcels from the MBL to the UT with virtually no entrainment has been shown to occur in cloud resolving model simulations (Lu et al., 2000), though this is not always the case, and depends on parameters such as updraft intensity, atmospheric stability, and wind shear. Mixing of air parcels with surrounding air in the UT is a more difficult issue. Mixing rates vary greatly as a function of altitude and time, which can easily be witnessed in the variable expansion and dissipation rates of aircraft contrails. Typical entrainment rates of 1–$3\times10^{-4}\,s^{-1}$ (e.g., Gerz et al., 1998) dilute a parcel with its background within less than a day. Entrainment rates of two orders of magnitude lower than this are needed to maintain a low $O_3$ mixing ratio over 10 days, even in the presence of only a 20 ppbv background of $O_3$. This extreme requirement may be one of the reasons for the rarity of these parcels in the MOZAIC data. Finally, unknown chemical losses cannot be ruled out, which could help offset the effects of mixing or even lead to in situ depletion of the $O_3$ in the airmasses. However, such a loss would have to be extreme, around an order of magnitude larger than the gross UT loss of ~0.2 ppbv/day due to known chemistry (von Kuhlmann et al., 2003), in order to overcome the known gross production of ~0.7 ppbv/day and to deplete a parcel from ~20 ppbv to <8 ppbv in 10 days. Such a large missing sink would have to be highly localised, otherwise it would cause a large general bias for CTMs to overestimate UT $O_3$ observations, which has not been noted. Thus, though it cannot be concluded definitively, we find the most likely origin of these observations to be $O_3$-depleted MBL airmasses lifted by deep convection that then remain unusually well intact and undergo only moderate $O_3$ production during their travel times.

Low $O_3$ events occurred often in Southeast Asia, but also events in Africa, Brazil and the southeast of Florida (US) were observed. The number of low $O_3$ events differs from year to year, but this may be due to variations in the destinations of the aircraft. They do not only occur over sea, but also over land. Low mixing ratio events were not only observed close to the selected maximum pressure level of 500 hPa, but also at pressure levels as low as 179 hPa. The horizontal extent of the low mixing ratios areas at cruise altitude varied from 5 to 30 km. During ascent and descent layers with low mixing ratios were encountered that were at least 100–600 m thick.

During flights 6062101, 6062201, 6062205, 6062207 that occur at almost the same time in adjacent areas (Sumatra, Malaysia), low mixing ratio areas were observed in areas that are 400 to 1000 km apart and at pressure levels varying from 217 to 370 hPa. The associated back trajectories came from about the same sea area near Indonesia. It cannot be excluded that they are part of one large low mixing ratio area, but this cannot be concluded from the available measurements.

The events 6082701 and 6082704 are likely to be part of one event. The geographical coordinates for their flight trajectories are very similar. The geographical position of the second low mixing ratio period of event 6082701 is almost the same as the geographical position of the second period of event 6082704, but there is a difference in pressure level (179 hPa for event 6082701 and 197 hPa for event 6082704). It should be noted that if two or more events mentioned above belong to the same event, there would be less than 11 low $O_3$ mixing ratio events in total.

During some low $O_3$ events the relative humidity was less than 100%. This indicates that the low $O_3$ mixing ratio in these cases is not caused by destruction of $O_3$ by reaction with ice particles at the moment of observation, but it cannot be excluded that this process has played a role during transport of the air mass.

Given the large number of flights and the geographical spread of the routes flown, we conclude that low $O_3$ mixing ratio events as defined here are exceedingly rare. It should be noted, however, that only the MOZAIC aircraft that were flying to South America (12% of the flights) came over the tropical Atlantic, while aircraft going to southeast Asian destinations crossed over tropical sea areas only for a small fraction of the time. There were no flights across the tropical Pacific. It is also worthwhile to note that the low mixing ratios in the upper troposphere reported by Kley et al. (1996) were measured by balloon-borne $O_3$ sondes at a pressure level of about 100 hPa, which is much lower than the lowest pressure level during the MOZAIC low $O_3$ events reported here (179 hPa). So it might well be that low $O_3$ mixing ratios occur more often than reported here, but at lower pressure levels and in other regions than the aircraft are cruising.

*Acknowledgements.* Manfred Helten, Forschungszentrum Jülich, Germany is gratefully acknowledged for detailed information on the relative humidity instrumentation. Gilles Athier, Laboratoire d'Aérologie, Toulouse, France is acknowledged for making the data available in an easily accessible way. We are grateful to Alain Marenco, Laboratoire d'Aérologie, Toulouse, France for his help throughout the years. EUMETSAT, Darmstadt is gratefully acknowledged for supplying METEOSAT images. The authors acknowledge the MOZAIC project that is funded by the European Communities, CNRS, Forschungszentrum Jülich, University of Cambridge, and Météo-France and supported by EADS Airbus and the Airlines: Air France, Deutsche Lufthansa, Sabena, and Austrian Airlines who carry free of charge the MOZAIC instruments. This work was supported in part by the German BMBF project 07ATC02.

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