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Comparisons of ozone measurements from the MOZAIC airborne program and the ozone sounding network at eight locations

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Abstract. Automatic ozone measuring devices have been operating continuously on board the five long-range aircraft of the Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) program since September 1994. This paper presents the main characteristics of the ozone system and the procedures followed to ensure its accurate calibration over long durations. Measurement accuracy was estimated at ±[2 ppbv + 2%], but much better in-flight levels were in fact observed: average discrepancy (between different devices) ranging from 1 ppbv at tropospheric concentrations to a few ppbv at stratospheric concentrations. This demonstrates the ability of the MOZAIC ozone data to produce accurate and reliable ozone climatologies. A 2-year ozone climatology (1994-1996) generated from MOZAIC data collected at between 0 and 12 km altitude was compared to longer and older measurements made at eight stations of the Ozone Sounding Network (OSN): Hohenpeissenberg, Wallops Island, Tateno, Palestine, Pretoria, Goose Bay, Biscarosse, and Poona. Despite the different nature of the programs (techniques, platforms, sampling frequencies, spatial distribution, and operation periods), the OSN and MOZAIC climatologies were found to show a reasonably high level of agreement. Mean concentrations derived from ozone sondes are about 3 to 13% higher than those obtained by the MOZAIC program in the free troposphere, in a similar geographic location. These differences are within the range of uncertainty of the two techniques. Larger discrepancies observed in the boundary layer and in upper layers are explained by the influence of local pollution and the distance between measurements, amongst other factors, limiting the reliability of comparisons. A comparison of OSD and MOZAIC data at Hohenpeissenberg/Frankfurt and Wallops Island/New York, over an overlapping period (1994-1995), shows good agreement in the free troposphere (800-300 hPa), no detectable bias for Hohenpeissenberg/Frankfurt, when taking into consideration the various causes of discrepancies (Dobson normalization, ozone geographical variations). Indeed, the results of this analysis support the hypothesis that it is not advantageous to scale the ozone sonde data to the overhead ozone column; the scaling appears to cause overestimation of the tropospheric O_3 concentrations, by about 3-6% at Hohenpeissenberg, and to cause more scatter in the sonde-MOZAIC differences. The correspondence between the OSN and MOZAIC climatologies obtained in very different conditions demonstrates that they are representative of the atmosphere and that, being complementary while each retains its own advantages, they are therefore both useful for validation studies.

1. Introduction

Ozone is obviously one of the most important chemical species involved in atmospheric chemistry, owing to its potential impact on the environment (chemical oxidation capacity, health, greenhouse effect, and vegetation). However, the ozone distribution within the whole troposphere is not sufficiently known, and its future evolution remains a matter of discussion, after the long-term period of increase observed since the beginning of this century has been followed by a recent leveling off [*Bojkov*, 1988; *Volz and Kley*, 1988; *Logan*, 1994; *Marenco et al.*, 1994; *Staehelin et al.*, 1994] (see discussion in subsection 7.3.3).

The budget, and therefore the trend, of tropospheric ozone are highly dependent on emissions in the atmosphere, by human activities, of carboneous and nitrogenous precursors [*Crutzen*, 1979, 1988; *Fishman and Crutzen*, 1977; *Thompson*, 1992], occurring mainly in the boundary layer. At upper altitudes, the

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release by subsonic aircraft of water vapor and nitrogen oxides in the upper troposphere (leading to ozone production) and also in the lower stratosphere raises the problem of their potential impact, especially when one considers the growth of air traffic forecast for the next century [*Marenco et al.*, this issue]. It is vital for the evaluation of future changes in climate and atmospheric composition that the processes affecting the distribution of ozone in these domains be fully understood.

Understanding and forecasting of the atmosphere cannot be achieved without the use of three-dimensional (3-D) models which are essential to combine the complex effects of natural and anthropogenic emissions, chemical transformations, horizontal and vertical transport, and removal of trace species. Several tropospheric 3-D models have been developed in recent years, and they need to be validated and improved by comparison with experimental data. Ozone variations recorded at ground level networks and from observations at altitude are being used for model evaluation. Until recently, the primary source of information on the vertical distribution of tropospheric ozone was the Ozone Sounding Network (OSN). The available data were analyzed recently by J.A. Logan (manuscript in preparation, 1998), who has provided climatologies for individual stations for use in model evaluation. Subsets of the ozone sonde data (OSD) have been used in the past to evaluate 3-D models [e.g., *Oltmans and Levy*, 1994; *Muller and Brasseur*, 1995; *Roelofs and Lelieveld*, 1995], and the more complete analysis of J.A. Logan (manuscript in preparation, 1998) is now being used by several modeling groups [*Friedl et al.*, 1997; *Wang et al.*, 1998; D.A. Hauglustaine et al., MOZART: A global chemical tracer transport model for ozone and related chemical tracers, 2, Model results and evaluation, submitted to *Journal of Geophysical Research*, 1998; hereinafter referred to as Hauglustaine et al., submitted manuscript, 1998].

Amongst the various programs started recently for evaluating the impact of aircraft, the Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) program was specially designed to collect ozone and water vapor data, using fully automatic devices installed on five long-range Airbus A340s in normal airline service, flying regularly all over the world [*Marenco et al.*, this issue]. Between September 1994 and December 1997, 7500 flights were made over continents and the Atlantic Ocean. MOZAIC cruise data (90% of measurements) were used to build ozone climatologies between 9 and 12 km altitude; these are presented and analyzed by *Thouret et al.* [this issue]. Comparisons between MOZAIC data and the results of various 3-D chemistry and transport models (CTM) are now in progress [*Law et al.*, this issue].

OSN and MOZAIC data correspond to: (1) different techniques (electrochemical sondes and UV photometer, respectively); (2) platforms (balloon and aircraft) and sampling frequencies (2-12 soundings per month for OSD, and daily or weekly profiles in MOZAIC); (3) spatial exploration (vertical profiles in about 40 locations for OSD, and vertical (20 m resolution) and horizontal (1 km resolution) measurements in MOZAIC); and (4) operation periods, 3-16 year series for OSD, often starting more than 20 years ago, and since 1994 for MOZAIC. This may result in distinct climatologies with possible consequences for model evaluation. An intercomparison between climatologies derived from the sonde data and from MOZAIC is presented below.

In the following, we call "climatologies" the monthly mean statistics for ozone profiles obtained from the sondes and from MOZAIC. One can wonder if such nomenclature is appropriate because a certain amount of data is required to document the main features and variability of tropospheric ozone (J.A. Logan, manuscript in preparation, 1998). For most sonde stations, there are between 20 and 150 profiles each month obtained over several years, while for the MOZAIC data used here (September 1994 to August 1996) there are a similar number of profiles for about half the locations, but sparser measurements in some months at the other locations.

Evaluation of 3-D models generally focuses on their ability to reproduce average ozone concentrations, seasonal cycles and vertical profiles, regional patterns in concentrations, and variability. Three-dimensional models tend to underestimate the variability of ozone, and it is important that data with information about the variability are used to the full. The sonde and MOZAIC data provide different statistics, the sondes giving longer time series with lower measurement frequency, and MOZAIC giving higher-frequency measurements over a much shorter period.

The first goal of this paper is to present the performance of the MOZAIC ozone devices and their capability to produce reliable measurements in an operational program. We then compare the different data sets at eight locations for which both types of measurement are available (1) long-term (3-16 year) climatologies for ozone sonde stations and (2) 2-year (1994-1996) MOZAIC climatologies, primarily the data collected during ascents and descents. We begin with a discussion of the quality of both types of measurements.

2. Performance of MOZAIC Ozone Devices

The five A340 aircraft involved in the MOZAIC program were operated by different airlines: one by Air France from Paris, one by Sabena from Brussels (operated by Air France until March 1997), two by Lufthansa from Frankfurt, and one by Austrian Airlines from Vienna. The aircraft flew almost 18 hours per day, and O_3 and H_2O measurements were recorded continuously during the ascent, cruise (9-12 km), and descent phases. Since the beginning of routine measurements, 7500 flights, corresponding to 54,000 flight hours (up to December 1997), have been performed over North and South America, the Atlantic Ocean, Europe, Africa, and Asia [*Marenco et al.*, this issue].

The data collected by the five MOZAIC aircraft have been calculated/validated and incorporated in a database developed at Centre National de Recherches Météorologiques (CNRM), Toulouse, before their evaluation by the scientific investigators. To ensure homogeneity of the database and reliability in time and space of derived ozone products, such as climatologies, it is crucial to make measurements of high quality and reproducibility. In particular, accuracy must remain constant over the entire duration of the program (several years), and it must be independent of the following: (1) the flight phase (ascent, descent, and cruise), (2) the aircraft considered, (3) the outside conditions (clear sky, clouds, and turbulence), and (4) the atmospheric composition (pollution; wide range of ozone concentration between boundary layer and stratosphere).

This was achieved by careful adaptation of the equipment to in-flight conditions, a procedure for sampling the outside air only during flight, periodic reference to an internal ozone generator, a yearly inspection and absolute recalibration of each O_3 device at Laboratoire d'Aérologie (LA/CNRS), a research unit for the Centre National de la Recherche Scientifique in Toulouse, careful data validation, and, finally, verification of data reproducibility between the different MOZAIC aircraft.

2.1. Instrumentation

2.1.1. Sensor. The ozone analyzer installed on board each of the five MOZAIC aircraft is a dual-beam UV absorption instrument (Thermo-Electron, model 49-103). The response time

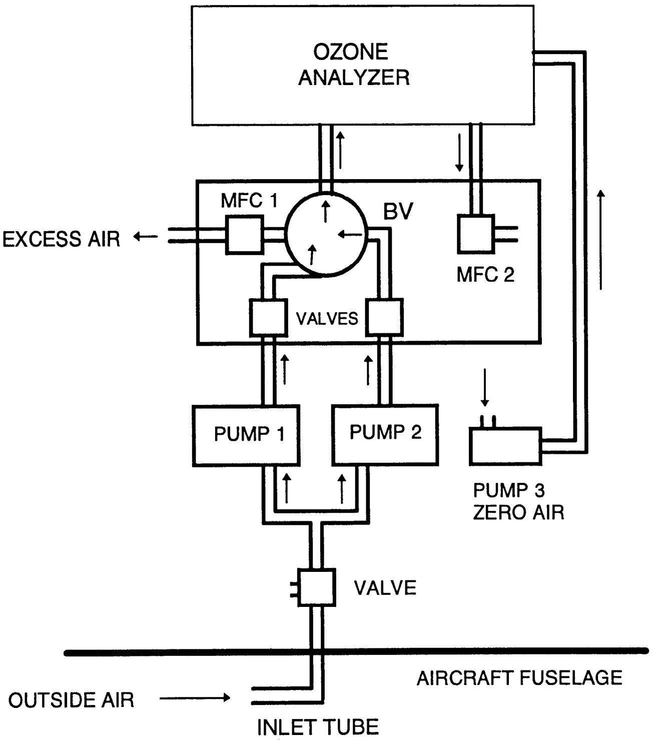

Figure 1. MOZAIC Ozone Pumping System (MFC, mass flow controller; BV, buffer volume).

is 4 s, and the concentration is automatically corrected for pressure and temperature influences. Because of its specifications, precision 2 ppbv, noise 1 ppbv, minimum detectable 2 ppbv, as given by the manufacturer, the operating principle, and the design of electronics, this model provides very good stability, making the measurements accurate and reliable over long time periods. It has been recognized by the U.S. Environmental Protection Agency (EPA) as an "Equivalent Method for the measurement of ambient concentrations of ozone pursuant with the requirements defined in 40 CFR Part 53 [40 FR 7049, February 18, 1975]".

2.1.2. Ozone measuring system. The instrument is mounted through shock absorbers on a shelf installed in a specially designed rack located in the electronic compartment, an area below the cockpit where temperature (20°C) and the aircraft's internal pressure are controlled. It is connected to the air inlet with Teflon lines (6 mm OD) through auxiliary devices (Teflon pump, Teflon valves; Figure 1) and it is controlled by the MOZAIC computer unit. The latter is also connected to the Air Data Computer (ADC) for the acquisition of aircraft parameters.

Outside air is sampled through a dedicated inlet tube consisting of a stainless steel pipe, Teflon-walled to avoid ozone destruction onto the steel. The probe plate is located on the fuselage 7 m back from the nose of the aircraft, such that the inlet (7 cm above the plate) is well outside the aircraft boundary layer which is 3 cm thick there, as calculated by the Department of Aerodynamics of Aérospatiale (B. Bauvieux, Airbus A340 engineering, private communication, 1997).

A Teflon KNF Neuberger pump (PUMP 1), providing a flow rate above the 2 l min^{-1} (STP) required, is used to pull the air through the analyzer from an outside pressure of 200-300 hPa at cruise altitude to the internal aircraft pressure. It is controlled by mass flow controllers (MFC)1,2 (Figure 1). An intermediate Teflon flask (BV, volume 100 cm^3), installed on the Teflon line upstream of the ozone analyzer, is used as a buffer volume to equilibrate the pressure with the internal aircraft pressure at the entrance to the ozone analyzer. A second Teflon pump (PUMP 2) is used as emergency equipment and is automatically operated in the case of failure of the other pump.

The residence time between the inlet and the ozone analyzer is about 2 s, which limits the possible ozone destruction in the lines. The inlet Teflon line and the pumps are checked frequently for cleanliness during aircraft ground maintenance and are changed periodically. In addition, a remote-controlled Teflon valve, activated after takeoff and before landing, is used to sample the external air for ozone measurements above 20 m altitude only, in order to prevent contamination of the input line by deposition of organic compounds and dust while the aircraft is on the ground and subject to local pollution. Laboratory and ground tests during maintenance have demonstrated that the ozone losses through the whole line (inlet, lines, pump, and buffer volume) remain less than 1%.

Considering the intensive operation of the A340 by the airlines, the MOZAIC system is quasi permanently powered. The ozone analyzer is therefore always equilibrated and ready to function, thus avoiding any perturbations occurring during the warming-up period (1 hour).

2.2. Calibration

2.2.1. Reference. Despite the high stability of the ozone sensor, confirmed by the experience of continuous functioning at ground-based stations over several years, a special procedure is followed to detect any drift in instrument efficiency during flight. This is achieved by using the remote-controlled ozone generator of the analyzer along with the supply of "zero air" provided by an auxiliary pump (PUMP3; Figure 1) sucking ambient air (in the electronic compartment) through an ozone filter. The ozone generator is automatically activated, and references are made at three levels, 0, 80, and 500 ppbv: before takeoff, about every 2 hours during cruise, and after landing.

The precision of the internal ozone generators for in-flight conditions is estimated at 3%, which includes UV lamp stability, auxiliary pump flow rate stability, and the limited time of activation (5 min) not allowing for complete stabilization. An example of variations over a 1-year period is shown in Figure 2. This information is used as an indication of the history of the ozone analyzer performance during the flight period (see following section).

2.2.2. Calibration. Seven ozone analyzers were used in the MOZAIC program. While five of them were flying on the aircraft, the sixth one, acting as a spare instrument, was being recalibrated and refurbished in the laboratory before reinstallation on board another aircraft.

At the beginning of the MOZAIC program, these six ozone analyzers were intercalibrated against a seventh one, remaining at LA/CNRS and acting as the "reference analyzer" (RA). Their factory calibrations agreed with that of the reference, within 0.5% range. The pressure transducers for pressure correction also showed agreement within the 0.5% range. Finally, the linearity of each analyzer was checked in the 0-1000 ppbv range. The absolute accuracy of the calibration of the reference analyzer (RA) was evaluated at 0.5%.

After this initial validation of the instruments, each ozone analyzer was exchanged yearly and recalibrated against the RA. Most of the time, 12 cases over a total of 15 analyzer operations between 1994 and 1997, ozone analyzers did not exhibit any significant variation (less than 1%) after 1 year of in-flight operation. Only three exceptional drifts have been detected since the beginning of the program. In one case, it corresponded to a slight but continuous evolution (6% over 6 months); in the two other cases, the same analyzer was concerned and suddenly

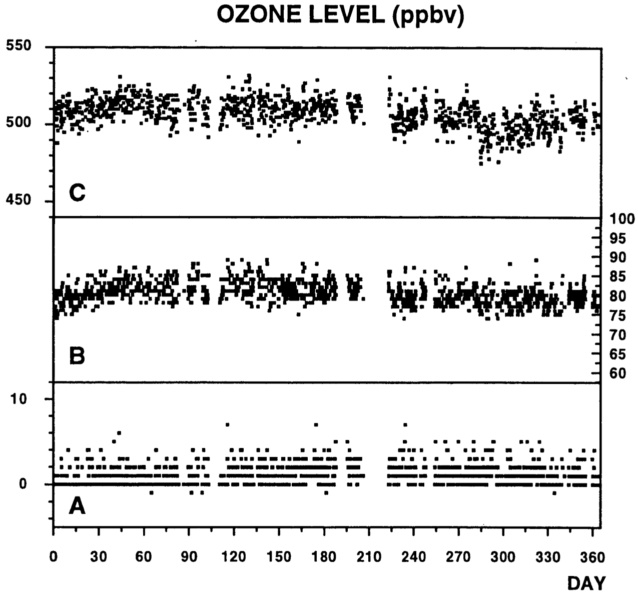

Figure 2. Variations in the response of the ozone analyzer 2 over an operation period (March 28, 1996 to March 26, 1997) on board aircraft MSN-053 for three levels of the internal ozone generator (A, 0 ppbv; B, 80 ppbv; C, 510 ppbv). Accurate preflight and postflight calibrations, made at the laboratory, were in agreement within a 0.5% range.

presented greater drift (25% over 3 months) which was attributed to the partial blocking, by particles, of one of the two capillary tubes controlling the airflow rates.

The internal ozone reference is not used for accurate on-line calibration but to identify the chronology of the main events in the case of significant drift in the calibration. The average curve describing the evolution of analyzer efficiency is deduced afterward, using the accurate preflight and postflight calibrations made at the laboratory and any main trends as revealed by the internal ozone reference; if necessary, corrections are applied to the data.

2.2.3. Precision of the measurements. Under these conditions, by integrating the possible losses in the lines (1%), absolute accuracy (1%), and instrument precision (2 ppbv), the characteristics of the ozone measurements performed on board the five MOZAIC aircraft are detection limit 2 ppbv, and uncertainties for individual (4 s) measurements ±[2 ppbv + 2%] (i.e., 2 ppbv at O_3 = 10 ppbv, 4 ppbv at O_3 = 100 ppbv, and 6 ppbv at O_3 = 200 ppbv).

2.3. Data Collection and Validation

The MOZAIC system uses the aircraft power supply and is computer-controlled. Through software held on the storage disk, an electronic interface drives the auxiliary devices (pumps, Teflon electrovalves, mass flowmeters, etc.). The measurements are taken every 4 s, starting after takeoff and continuing up to landing. MOZAIC data (O_3, H_2O, T, and status control parameters), together with the aircraft parameters from the Air Data Computer, are stored on removable high-capacity disks exchanged after about every 500 flight hours.

The Laboratoire d'Aérologie performs the processing and validation of the ozone and aircraft data by numerical and visual checks of their temporal recording and status of control parameters. Erratic values are filtered, and critical phases, for example, icing of the inlet in rare cases resulting in a drop of the airflow rate in the ozone measurement system, are detected and eliminated. As mentioned above, the performances of the analyzer are checked but not corrected at this step, resulting in the formation of provisional ozone data files. The ozone data files are definitively validated once the laboratory recalibration step of the ozone analyzer has been performed, taking into account the few cases of calibration drift.

2.4. In-Flight Performances

After the careful procedure of intercalibration and control of the ozone devices, it was important to check the quality, homogeneity, and repeatability of the measurements performed during the MOZAIC flights. As mentioned previously, several factors can perturb in-flight ozone measurements, and we have investigated any indication of deficiency in the data. The difficulty comes from the identification of conditions suitable for the intercomparison of ozone data. The ozone distributions are far from being homogeneous in the atmosphere; they often vary dramatically, and any difference between the measurements, in time and space, can have important consequences, preventing convenient comparisons. Thanks to the considerable amount of data already collected (54,000 flight hours), we have been able to select a sufficiently high number of cases, in close correspondence in time and space, suitable for these validations.

First, we can exclude any influence of the flight phases (made at ascent and descent) and atmospheric conditions (turbulence). Indeed, many examples of ozone profiles, established in favorable conditions, 2 hour intervals and identical flight tracks, during the descent and the ascent following the stopover at airports, have shown very similar vertical distributions, well within the range of the uncertainty of the method. This demonstrates that the angle of attack of the aircraft, different during ascent and descent which therefore might modify the aircraft boundary layer, has no detectable influence upon the measurements. Similarly, no perturbation in the functioning of the ozone system has been detected when entering clouds or in strong turbulence, except for some ozone peaks detected in the tropics close to strong convective systems and which have been related to stratosphere/troposphere exchange phenomena [*Suhre et al.*, 1997; *Cammas et al.*, this issue].

2.4.1. Vertical distributions. The regular operation by Lufthansa of their two MOZAIC aircraft from Frankfurt gives an opportunity to make close comparisons in the vicinity of this city. Numerous examples of ozone vertical profiles, established within the troposphere between 1995 and 1997, for different combinations of ozone devices (SN 01, SN 03, SN 04) and aircraft (AC 03, AC 04), flying along the same trajectory and only 5 to 18 min apart, show an excellent agreement both in the shape of the profiles and in the ozone concentrations recorded. Figure 3 presents an example of ozone profiles for an ascent from Frankfurt, and Table 1 gives a numerical data comparison in four cases, by integrating the profiles over different altitude ranges. The discrepancy between devices (fourth column) is found much lower (0.1 to 3 ppbv) than the maximum uncertainty (5 to 6 ppbv; fifth column), based on the precision of 4 s resolution measurements evaluated in subsection 3.2.3 (2 sigma between two different ozone devices). This is due partly to averaging the data over 3 to 5 min intervals which results in better instrument precision. The average discrepancy of these four examples amounts to 0.9 ppbv. The 4.2 ppbv observed for Figure 3, between individual measurements at 7.5/9 km altitude, is due to the divergence of aircraft trajectories, 15 km at 7.5 km, 60 km at 9 km, and is therefore not significant. These comparisons confirm that there is no bias between the different ozone devices.

2.4.2. Cruise recordings. Examples of the good correspondence between recordings at cruise altitude by different

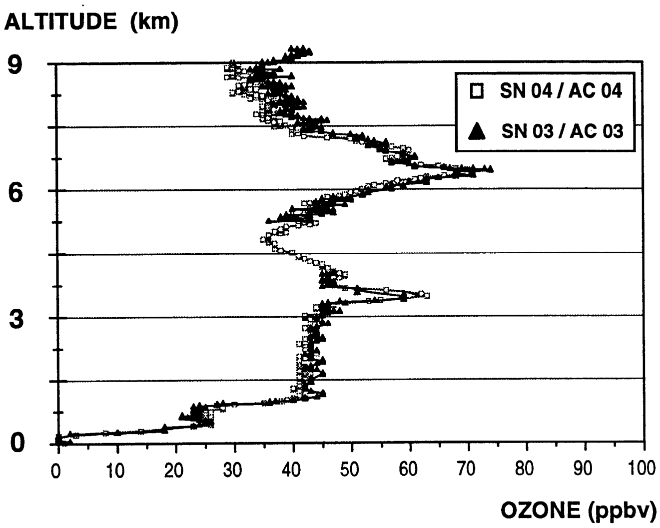

Figure 3. Ozone vertical profiles obtained by two ozone devices (SN 03 and SN 04) installed on board two MOZAIC aircraft (AC 03 and AC 04) during their ascent from Frankfurt on February 3, 1995, at a 5 min interval on the same trajectory.

MOZAIC aircraft are given in Figure 4 (zonal flights between New York and Frankfurt or Dusseldorf) and Figure 5 (meridional flights between Sao Paulo and Paris or Frankfurt) for different combinations of ozone devices (SN 01, SN 05, SN 06) and aircraft (AC 02, AC 03, AC 04), flying along the same trajectory (no more than 10 km horizontal spacing) and at a short time interval (1/2 hour). The ozone variations are in very good agreement: 0.2 to 3.4 ppbv for tropospheric conditions (Table 2; Figures 4 and 5), and 17 to 27 ppbv for stratospheric conditions (Table 2; Figure 4). Some high values are not representative, for example: (1) -43.6 ppbv at -60°/-50° longitude (example from Figure 4), due to the distinct climbs of the two aircraft just at the tropopause level (see the altitude plots in Figure 4); (2) 8.2 ppbv at -10°/0° longitude (same example), due to the divergence of horizontal trajectories, 30 km at -5°, 85 km at 0°.

These two cases correspond to large ozone variations (tropospheric, stratospheric) similarly caught by the two aircraft. The rather large discrepancy (17-27 ppbv) observed at high stratospheric concentrations (400-750 ppbv; Figure 4 and Table 2) is probably due to the existence of strong vertical and temporal ozone gradients (in a jet stream region), which makes the comparison difficult between the two aircraft flying at some vertical spacing and at a 1/2 hour interval. A rough estimation,

Table 1. Comparison of the Average Ozone Concentrations From the Integration, for Different Altitude Ranges, of Ozone Vertical Profiles Obtained by Two Ozone Devices (SN) Installed on Board Two MOZAIC Aircraft (AC) (Columns 2 and 3), With Discrepancy ΔO_3 (Columns 2-3) and Estimated Maximum Uncertainty (2 Sigma)

Altitude Range, km	O_3, ppbv	O_3, ppbv	ΔO_3, ppbv	2 Sigma O_3, ppbv
		Ascent From Frankfurt[a]		
0-1.5	25.8	25.7	0.1	5
1.5-3	43.7	42.2	1.5	5.8
3-4	47.3	49.2	-1.9	5.8
5-6	45.3	43.9	1.4	5.8
6-7.5	57.7	56.3	1.4	6.2
7.5-9	37.8	33.6	4.2	5.4
Mean	42.9	41.8	1.1	5.6
		Ascent From Frankfurt[b]		
0-1.5	26.8	25.2	1.6	5
1.5-3	40.2	40.7	- 0.5	5.6
3-4.5	39.3	38.3	1.0	5.6
4.5-5.1	40.4	39.7	0.7	5.6
Mean	36.7	36.0	0.7	5.4
		Ascent From Frankfurt[c]		
0-1.5	27.8	26.0	1.8	5.1
1.5-3	47.1	46.1	1	5.8
3-4.5	52.6	50.2	2.4	6
4.5-6	50.2	47.1	3.1	6
6-7.5	40.2	39.1	1.1	5.6
7.5-8.4	35.8	35.3	0.5	5.4
Mean	42.3	40.6	1.7	5.6
		Descent to Frankfurt[d]		
0.4-1.2	18.3	15.8	2.5	4.6
1.5-3	35.5	34.4	1.1	5.4
3-4.5	46.7	47.0	-0.3	5.8
4.5-6	37.7	37.8	-0.1	5.5
6-7.5	37.4	39.1	-1.7	5.5
7.5-9	38.4	40.7	-2.3	5.5
9-1.5	43.7	44.0	-0.3	5.7
Mean	36.8	37	-0.2	5.5

[a] February 2, 1995, at 5 min interval; Figure 3 example; column 2: SN 03 on AC 03; column 3: SN 04 on AC 04.

[b] December 28, 1996, at 13 min interval; column 2: SN 01 on AC 03; column 3: SN 03 on AC 04.

[c] March 13, 1997, at 18 min interval; column 2: SN 01 on AC 03; column 3: SN 04 on AC 04.

[d] March 18, 1997, at 9 min interval. column 2: SN 01 on AC 03; column 3: SN 04 on AC 04.

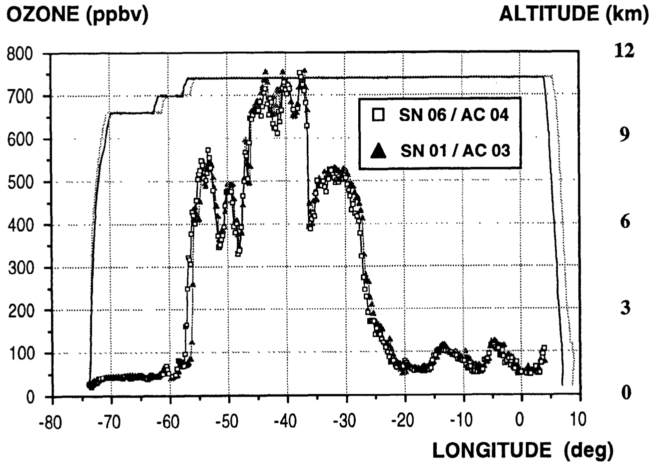

Figure 4. Ozone temporal recordings obtained by two ozone devices (SN 01 and SN 06) installed on board two MOZAIC aircraft (AC 03 and AC 04) during their travel from New York to Frankfurt or Dusseldorf on March 14, 1997, at a 30 min interval on the same trajectory.

based on the positive vertical ozone gradient observed in the lower stratosphere (400 ppbv over 600 m, during the climb at -57° longitude; Figure 4), suggests a 20 ppbv ozone excess for the aircraft (AC4) flying only 30 m above the other (AC 03). If this was the case, the real discrepancy would be therefore reduced to only a few ppbv.

An idea of the capability of the MOZAIC system to record similarly fast ozone variations is given in Figure 6 (an enlargement of Figure 5 between 13°-17°N latitude). Sharp ozone peaks, with a horizontal extension of 6 km, are recorded by the two MOZAIC aircraft flying from Sao Paulo to Paris and Frankfurt (flight paths: 15 m vertical spacing; 1-6 km horizontal spacing) at an interval of 28 min: 115 ppbv at 15.05°N/26.08°W for AC 02, and 108 ppbv at 15.12°N/26.15°W for AC 02. These peaks correspond certainly to the same stratosphere/troposphere exchange event, as previously indicated for the tropics by *Suhre et al.* [1997], using the MOZAIC data.

2.4.3. Comparison with ozone sonde data. Lastly, comparisons can be made between ozone profiles established near Frankfurt (MOZAIC) and from the station of Hohenpeissenberg (ozone sonde) in Germany. An interesting case is presented in Figure 19 (case 2) for January 2, 1995. The similarity of the ozone vertical distributions is excellent (Table 3; 0.4 to 5.1 ppbv discrepancy; 1 ppbv on average), despite the rather distinct locations of the measurements, 250 km between Frankfurt and Hohenpeissenberg and 400 km distance between sampling at 200 hPa, certainly because the troposphere was rather homogeneous and well mixed. The discrepancies in the boundary layer (250 km distance; Hohenpeissenberg at 900 hPa altitude) and in the upper layers (too far distant) are not considered significant. The removal of the scaling factor (1.09) against the Dobson column applied to Hohenpeissenberg ozone soundings (see discussion in subsections 4.4 and 7.3.2) does not improve in this case the agreement between the two distributions.

3. Ozone Sonde Measurement Techniques

3.1. Ozone Sondes

The two types of ozone sondes in common use in the OSN are the Brewer Mast (BM) bubbler [*Brewer and Milford*, 1960], and the electrochemical concentration cell (ECC) [*Komhyr*, 1969; *Komhyr and Harris*, 1971] which are both based on chemical detection of ozone after pumping the air into an iodine/iodide solution. The fundamental difference between the sonde types is a function of the type of anode used: a silver anode in the same reaction chamber for BM sondes, a platinum anode in a separate chamber for ECC sondes.

Similar ECC sondes were used at Wallops Island, Palestine, Pretoria, and Goose Bay, and BM sondes were employed at Hohenpeissenberg and Biscarosse. Soundings at Poona were performed with a BM Indian version, and those at Tateno were performed with a Japanese ECC version (KC79). Several of these sounding programs are described by the *World Meteorological Organization (WMO)* [1998], which also describes the sonde technique in detail, and associated errors.

3.2. Instrumental Uncertainties

Various studies using a standard UV photometer have shown that concentrations of tropospheric ozone are overestimated with ECC sondes, but underestimated by BM sondes [*Barnes et al.*, 1985; *Hilsenrath et al.*, 1986; *Beekmann et al.*, 1995; *Komhyr et al.*, 1995; *Reid et al.*, 1996].

The two error sources mainly affecting the tropospheric part of the profile are the contamination of the sonde sensor with reducing agents, causing a negative bias especially for BM sondes, and the estimation of the background current, causing presumably a positive bias for ECC sondes and a negative bias for BM sondes [*Beekmann et al.*, 1994].

Measurements of the tropospheric profiles are sensitive to the methods of background/offset signals processing. There are also uncertainties from the pump efficiency and solution concentration. The sensitivity of the ECC sonde increases during the flight, due to changes of concentrations caused by evaporation in the sensing solution [*Smit et al.*, 1998]. Applying the full background current correction leads to an underestimation in stratospheric ozone of around 2-6% [*Smit et al.*, 1994], possibly indicating a need to separate corrections for tropospheric and stratospheric data. Sharp changes in ozone during the ascent phase of an ozone sounding are smoothed by the effect of hysteresis. The descent portion of

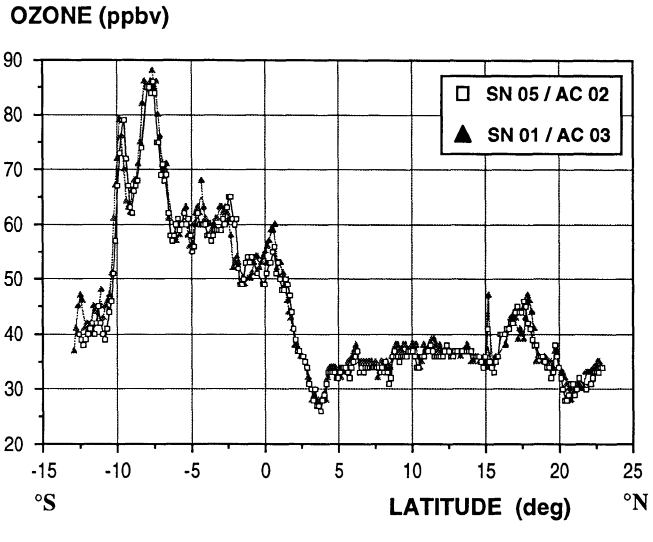

Figure 5. Ozone temporal recordings between 12°S and 22°N, obtained by two ozone devices (SN 01 and SN 05) installed on board two MOZAIC aircraft (AC 03 and AC 02) during flights Sao Paulo to Paris or Frankfurt on November 13, 1996, at 28 min interval on the same trajectory (data were not compared north of 22°N because the divergence of horizontal trajectories became rapidly superior to 60 km).

Table 2. Comparison of the Average Ozone Concentrations From the Integration, for Different Longitude or Latitude Ranges, of Ozone Recordings Obtained by Two Ozone Devices (SN) Installed on Board Two MOZAIC Aircraft (AC) (Columns 2 and 3), With Discrepancy ΔO_3 (Columns 2-3) and Estimated Maximum Uncertainty (2 Sigma)

Longitude or Latitude Band	O_3, ppbv	O_3, ppbv	ΔO_3, ppbv	2 Sigma O_3, ppbv
	Flights From New York to Frankfurt or Dusseldorf[a]			
73°W to 70°W	37.5	34.1	3.4	5.4
70°W to 60°W	44.7	46.3	-1.6	5.8
60°W to 50°W	299.4	344.1	-43.6	17.0
50°W to 40°W	603.9	586.3	17.6	28.0
40°W to 30°W	575.8	549.0	26.8	26.0
30°W to 20°W	243.2	216.0	27.2	12.8
20°W to 10°W	79.4	77.6	1.8	7.2
10°W to 0°	83.9	75.7	8.2	7.2
Mean	246	241	5	13.6
	Flights From Sao Paulo to Paris or Frankfurt[b]			
13°S to 10°S	46.0	42.6	3.4	5.8
10°S to 5°S	71.5	68.8	2.7	6.8
5°S to 0°	57.7	57.2	0.5	6.2
0° to 5°N	40.8	39.1	1.7	5.6
5°N to 10°N	35.4	34.4	1.0	5.4
10°N to 15°N	36.8	36.3	0.5	5.4
15°N to 20°N	38.8	38.6	0.2	5.5
20°N to 23°N	32.0	31.1	0.9	5.2
Mean	44.9	43.5	1.4	5.8

[a] March 14, 1997, at 30 min interval; Figure 4 example; column 2: SN 01 on AC 03; column 3: SN 06 on AC 04.

[b] November 14, 1997, at 28 min interval; Figure 5 example; column 2: SN 01 on AC 03; column 3: SN 05 on AC 02.

the profile is unreliable, especially in the troposphere, due to recent exposure to stratospheric O_3 concentrations [*Reid et al.*, 1996]. It is standard practice to use the ascent portion of the sounding. The use of revised corrections, for example, measurement of the background current before exposure to ozone in the preparation procedure, in place of the standard procedure recommended by the manufacturer, results in a significant reduction of the bias on the ECC sondes [*Beekmann et al.*, 1995; *Reid et al.*, 1996]. All of the sonde data used here, however, used the standard procedures for the particular type of sonde.

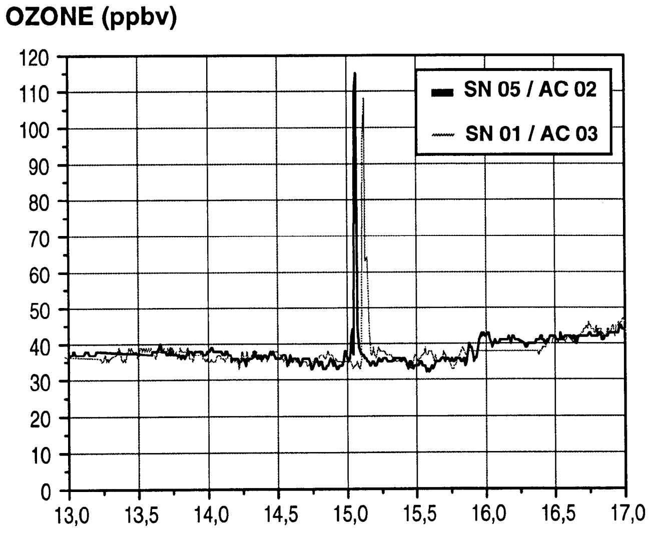

Figure 6. Enlargement of the 13°-17°N latitude band of Figure 5 example (Sao Paulo to Paris/Frankfurt on November 13, 1996).

3.3. Intercomparisons Campaigns

Various ozonesonde intercomparison campaigns have been performed since 1970. Results are summarized in Table 4. All the intercomparisons except *WMO* [1991] showed that Brewer Mast sondes measure less ozone in the troposphere than ECC sondes, by 10-25%; the intercomparison in Canada in 1991 showed that BM sondes measured 15% higher ozone values than ECC sondes [*WMO*, 1991]. Comparisons between ECC and Japanese sondes showed that KC-68 sondes measured about 13% less tropospheric ozone than the ECC sondes in 1970 and 1978 [*Attmannspacher and Dütsch*, 1970, 1981], while the comparison with KC-79 sondes in 1991 showed that they measured about the same amount of tropospheric ozone as ECC sondes [*WMO*, 1991]. All the Japanese sonde data used here were obtained with KC-79 sondes. Table 4 summarizes field measurements of the relative differences between BM and ECC sondes (with respect to ECC sondes), and Table 5 summarizes estimates of the bias of sonde measurements.

Estimates of the precision of ozone sondes in the troposphere are in the range 4-12% [*Barnes et al.*, 1985; *Beekmann et al.*, 1994; *Komhyr et al.*, 1995] with higher values shown for BM sondes in the 1991 intercomparison, 8-18% [*WMO*, 1991].

An international Jülich Ozone Sonde Intercomparison Experiment (JOSIE) was conducted in 1996 at Jülich to assess the performance of the different ozone sondes used within the ozone sounding network. Eight ozone sounding laboratories from seven countries participated, including the four sonde types used in this study, ECC, BM, BM/Indian, and Japanese KC-79 [*Smit et al.*, 1998]. These were compared with an accurate UV photometer in a simulation chamber that mimics the environment (pressure. temperature, and ozone) of typical ozone profiles in midlatitudes and the tropics. In general, ECC sondes provided much more consistent results than the other types of sondes. The precision was found to be better for the ECC sondes (5%) than for the non-ECC sondes (10-15%), particularly in the troposphere. Table 6 lists the variability (standard deviation) of different sonde types,

Table 3. Comparison of the Average Ozone Concentrations From the Integration, for Different Altitude Ranges, of Ozone Vertical Profiles Obtained on January 2, 1995, by a MOZAIC Aircraft Near Frankfurt and an Ozone Sonde Launched From Hohenpeissenberg Station, With Discrepancy ΔO_3 (Columns 2-3) and Estimated Maximum Uncertainty (2 Sigma)

Altitude Range, hPa	O_3, ppbv Frankfurt[a]	O_3, ppbv Hohenpeissenberg[b]	ΔO_3, ppbv	2 Sigma O_3, ppbv
900-700	34.6	29.7	4.9	5.4
700-600	34.6	35	-0.4	5.4
600-500	38.8	38	0.8	5.5
500-400	44.2	39.5	4.7	5.7
400-300	130	124.9	5.1	9.2
300-230	320	371	-51	16.8
mean (700-300)	61.9	59.3	1.6	6.4

[a] Aircraft ascent from Frankfurt, 0530 to 0600 UT; ozone device SN03 installed on board MOZAIC aircraft AC03.

[b] Ballon ascent from Hohenpeissenberg, 0700 to 0800 UT.

for 12 individual simulation runs during JOSIE, which is an indication of the accuracy and reproducibility of measurements.

These various results show that the ECC sondes perform better than other sondes with regards to precision and accuracy. The observed differences are mostly due to differences in the preparation and correction procedures applied by the different laboratories. In tropospheric conditions, biases of 3% for ECC sondes and no larger than ±5% for BM, BM/India, and KC79 are observed [*WMO*, 1998]. These biases can generally be attributed to an offset effect resulting from the evaluation of the background current [*Smit et al.*, 1998].

3.4. Dobson Normalization

In practice, individual ozone soundings are generally scaled to a concurrent measurement of the ozone column made at the same place with a Dobson spectrophotometer, by introducing a correction factor (CF). This procedure requires an estimate of the amount of ozone above the altitude reached by the sonde, about 30 km [*Dutsch et al.*, 1970]. The overall precision of the Dobson normalization was evaluated to about 5% [*Beekmann et al.*, 1994]. The scaling procedure introduces errors caused by the uncertainty in the ozone content above 30 km, the errors associated with the Dobson technique, and probably the bias resulting from variation of sonde efficiency with altitude. Correction factors are typically about 1.25 for BM sondes and 1.0 for ECC soundings [*Logan*, 1985, 1994].

The application of this correction to the tropospheric part of the ozone profile has recently become a subject of discussion [*Smit et al.*, 1994; *Beekmann et al.*, 1994, 1995; *Smit et al.*, 1998]. The CF depends primarily on the amount of ozone in the lower and middle stratosphere, where most of the column resides. Some stations, like Wallops Island (United States), Natal (Brazil), and Observatoire de Haute Provence (France), do not use the scaling procedure. From the discussion in the literature, it is unclear if the removal of the Dobson normalization should apply to all sonde types [*Smit et al.*, 1994; *Beekmann et al.*, 1994; *Smit et al.*, 1998] or only to BM sondes, as was recommended by *Beckmann et al.* [1995], following the 1991 Observatoire de Haute Provence (OHP) intercomparison. This issue is discussed by *WMO* [1998], where it is concluded that more research on the topic is required.

4. Ozone Data

The two sets of data used in this study to provide ozone climatologies differ in their sampling statistics as well as in the technique used. MOZAIC measurements are for 2 years (September 1994 to August 1996), with high density and extensive geographic coverage (Figure 7). The data used here are primarily those obtained during ascents and descents into and out of airports. The geographic and seasonal variation of ozone in the upper troposphere/lower stratosphere derived from cruise data is presented by *Thouret et al.* [this issue]. The sonde data are from a limited number of stations selected for their proximity to MOZAIC measurements; they are located primarily in northern midlatitudes. The measurement frequency for most of the sonde stations is weekly, and the data used here are several year averages, taken from J.A. Logan (manuscript in preparation, 1998).

Eight sonde stations were used in this study (see Table 7): (1) five stations, based on the criterion of proximity to a major MOZAIC airport (Hohenpeissenberg/Frankfurt; Wallops

Table 4. Differences of Brewer-Mast (BM) Sondes With Respect to ECC Sondes, With and Without Application of Correction Factor (CF)

Campaign[a]	Period	Reference	Troposphere, Without CF	Troposphere, With CF	Stratosphere, Without CF	Stratosphere, With CF
HOH	1970	1	-37	-25	-15	2
HOH	1978	2	-21	-10	-15	-2
BOIC	1983-84	3	-29	-20	-9	2
OHP	1989	4	-15	-14	1	4
OHP	1991	5	-19	-12[b]	N.D.	N.D.
WMO	1991	6	N.D.	15	N.D.	3

N.D., no data; differences in percent; references: 1, *Attmannspacher and Dütsch* [1970]; 2, *Attmannspacher and Dütsch* [1981]; 3, *Hilsenrath et al.* [1986]; 4, *Beekmann et al.* [1994]; 5, *Beekmann et al.*, [1995]; 6, *WMO* [1991].

[a] Hohenpeissenberg, HOH; Balloon Ozone Intercomparison Campaign, BOIC; Observatoire de Haute Provence, OHP; World Meteorological Organization, WMO.

[b] Correction factor (CF) applied on ECC sondes only.

Table 5. Absolute Bias of Brewer-Mast (BM) and ECC Sondes

Campaign[a]	Reference	ECC		BM	
		Troposphere	UT/LS[b]	Troposphere	Stratosphere
WALL	1	8 to 14	3 to 5	N.D.	N.D.
OHP	2	3.5	6	-9.5	6
STOIC	3	3	N.D.	N.D.	N.D.
WAL	4	lower than 4	N.D.	N.D.	N.D.
OHP	5	4.5	N.D.	-11	N.D.

N.D., no data; absolute bias in percent; references: 1, *Barnes et al.* [1985]; 2, *Beekmann et al.* [1994]; 3, *Komhyr et al.* [1995]; 4, *Reid et al.* [1996]; 5, *Beekmann et al.* [1995].

[a] Wallops Island, WALL; Observatoire de Haute Provence, OHP; Stratospheric Ozone Intercomparison Campaign, STOIC; Wales, WAL.

[b] Upper troposphere/lower stratosphere, UT/LS.

Island/New York; Tateno/Tokyo-Osaka; Palestine/Houston-Dallas; Pretoria/Johannesburg); (2) three stations below MOZAIC routes and well sampled by this program at cruise levels (Goose Bay; Biscarosse; Poona).

4.1. MOZAIC Data

The MOZAIC climatology presented here corresponds to ozone monthly mean values at 10 pressure levels (1000, 900, 800, 700, 600, 500, 400, 300, 250, and 200 hPa) averaged over September 1994 to August 1996. These values are the arithmetic means of all data recorded for a given month (e.g., if 10 flights are available for January 1995 and only two are available for January 1996, the January mean value represents the 12 flight average; this means that interannual variation is not taken into account).

4.1.1. Cities. The data used here correspond to the vertical profiles (ascent and descent) obtained near the airports and are available for practically all standard pressure levels. The MOZAIC measurements are initially referred to pressure, the parameter directly measured by the aircraft, and pressure is converted to altitude using a standard atmosphere. For this analysis, pressure was used, as the sonde data are also referred to pressure. Data are averaged in layers 50 hPa thick and centered on the standard pressure levels (i.e., the value at 500 hPa is the average of all data between 475 and 525 hPa). Table 8 presents the monthly frequency of profiles available at these cities for 900 to 300 hPa. Since vertical profiles are defined as the part of the flight between ground level and the first stabilized level, for example, the 300 hPa level for ascents and 200 hPa for descents, data available above 300 hPa are less numerous, about half the values in Table 7, since they are obtained only during descents. Figure 8 describes the distribution of profiles as a function of the time of day, a useful parameter to consider when interpreting results in lower layers. This time depends on the city and on the airline strategy.

4.1.2. Cruise locations. Only three standard pressure levels (300, 250, and 200 hPa) are documented at the five cruise locations and they correspond to the integration of MOZAIC data obtained at five main cruise levels (Figure 9). Depending on the air traffic density over these areas, these locations are sampled much less than the cities (see Table 8, bottom). The ozone monthly mean values were averaged in square 5° (latitude) x 5° (longitude) centered on the ground coordinates of the station.

Table 6. Standard Deviation (s.d.) of Ozone Sondes Observed During the JOSIE Intercomparison

Sonde Type	Origin		s.d.[a]
	Country	Location	
BM	Germany	Hohenpeissenberg	0.07
	Switzerland	Payerne	0.10
ECC	USA	Boulder	0.02
	Canada	Ontario	0.02
	Germany	Jülich	0.02
	France	OHP	0.07
BM/Ind	India	Poona	0.08
KC79	Japan	Tokyo	0.10

[a] In a scale 0 to 1; informations taken from *Smit et al.* [1998].

4.2. Ozone Sonde Data

The ozone sounding climatology presented here was developed by J.A. Logan (manuscript in preparation, 1998), in part for the evaluation of 3-D models [*Friedl et al.*, 1997; *Wang et al.*, 1998]. Monthly statistics are provided at 22 pressure levels from 1000 to 10 hPa for about 40 stations. The base period of the analysis was 1980 to 1993, but not all stations operated during this period. Earlier data were included to increase the geographic coverage, and later data were included where needed to provide an adequate number of soundings. Soundings with anomalously low or high correction factors were excluded from the analysis [*Logan*, 1994]. Ozone values were interpolated to the selected pressure levels for each sounding, and mean monthly values were formed. These weight each sounding equally, in a similar manner to the climatology for the MOZAIC data. The dates for the stations used in this comparison are given in Table 7. They are all prior to those for the MOZAIC data for 1994-1996, some by as much as 15 years. Sonde data for 1995-1996 for Hohenpeissenberg and Tateno were used to determine whether a trend in ozone is influencing the comparisons made here. The number of ozone soundings retained at each station is given in Table 9, as an indication of the confidence in the corresponding climatologies. The time of launch of the sondes was also considered in making the comparisons near the surface as discussed below. For most of the stations considered here the tropopause heights are available, based on an analysis of the temperature data obtained with the ozone data (J.A. Logan, manuscript in preparation, 1998).

5. Presentation of MOZAIC and Ozone Sonde Climatologies

5.1. Comparisons Near Cities

5.1.1. Hohenpeissenberg/Frankfurt. This location corresponds to the best statistics for both soundings (2-3/week) and MOZAIC data (2-3 profiles/day) (see Tables 7, 8, and 9). BM sondes were used at Hohenpeissenberg. Ozone seasonal variations show good agreement between the two climatologies, both in phase and mixing ratios, despite the quite different statistics and sampling periods (Figure 10). The seasonal variations are characteristic of the mid northern latitudes with a summer maximum in the troposphere (1000-300 hPa) related to photochemistry, and a spring maximum above the 300 hPa level, in the region of the tropopause, where both upper tropospheric and lower stratospheric air may be sampled. The mean tropopause

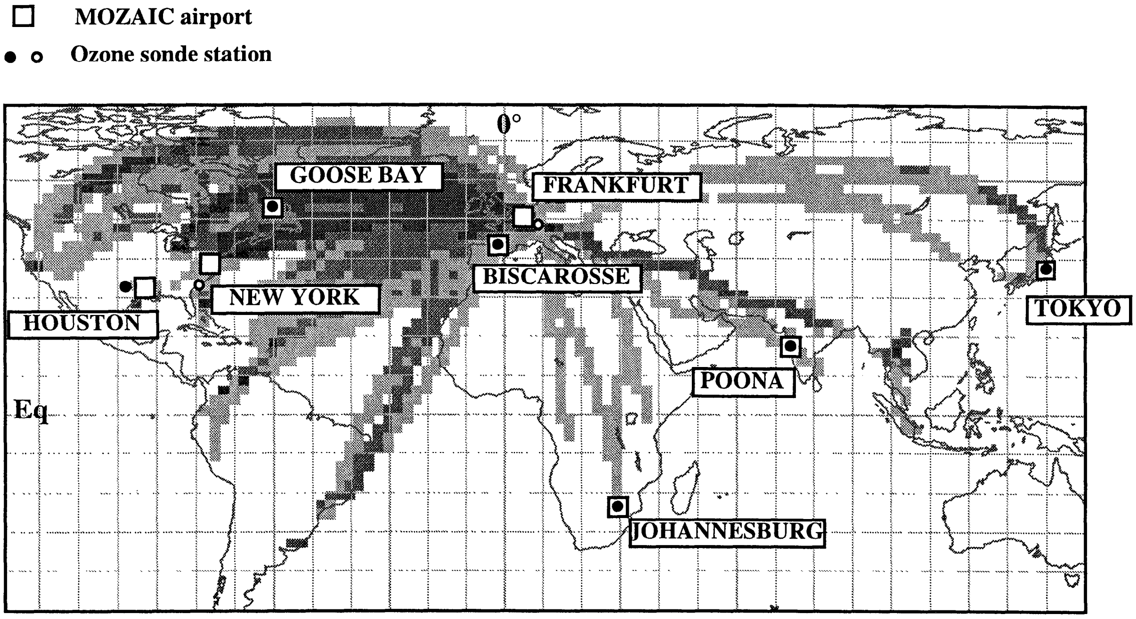

Figure 7. Example of the geographical coverage of MOZAIC flights with indications of data density (low, light gray; medium, gray; high, dark gray) at the 262 hPa level in winter (1995; 1996) and position of MOZAIC airports/altitude locations (squares) and ozone sonde stations (circles).

is ~250 hPa in spring and ~200 hPa in autumn at Hohenpeissenberg. Ozone concentrations (70-400 ppbv) and standard deviations are much higher in this region than in the middle and lower troposphere. The best agreement between the two data sets is obtained at the 800, 300, and 250 hPa levels.

Some differences are observed, however (see also Plate 1): (1) in the boundary layer (900 hPa), where MOZAIC data exceed OSD in spring and summer (see discussion in section 7); (2) in the free troposphere (700-400 hPa), where the O_3 mixing ratio is 40-80 ppbv, OSD exceed MOZAIC data all year by 5- 35%; and (3) in the lower stratosphere (200 hPa) in spring where OSD exceed MOZAIC data (by a factor of 2 in April).

5.1.2. Wallops Island/New York. The statistics are lower at this location for both soundings (2-3/month) and MOZAIC data (1-2 profiles/day). An ECC sonde was used at Wallops Island, without correction to the total ozone column.

Here again, the two seasonal variations are in agreement and are similar to those obtained at Hohenpeissenberg (summer maximum in the troposphere; spring maximum in the upper layers) (Figure 11).

OSD exceed MOZAIC data by 5 to 45% at each level between the 1000 and 400 hPa levels (Plate 1). The tropopause at Wallops Island is ~240 hPa in late winter/early spring and near 150 hPa in late summer. The discrepancy between the two data sets varies

Table 7. Ozone Sounding Stations and Corresponding MOZAIC Airports (or Areas) Considered in This Comparison, With Indications of Distances Between Ozone Soundings and Average MOZAIC Profiles, Data Frequencies, Ozone Sonde Types, and Series Periods

Ozone Sounding Station[a]	MOZAIC Cities or Cruise Location	Position	Distance, km	Frequency	Sonde Type	Period
Hohenpeissenberg		48°N, 11°E	350	2-3 / week	BM	1980-1993
	Frankfurt	50°N, 9°E		2-3 / day		1994-1996
Wallops Island[b]		38°N, 76°W	500	2-3 / month	ECC	1980-1993
	New York	41°N, 74°W		1-2 / day		1994-1996
Tateno		36°N, 140°E	200	2-3 / month	ECC/KC79	1980-1995
	Tokyo-Osaka	36°N, 140°E		2-3 / week		1995-1996
Palestine		32°N, 96°W	300	1-2 / month	ECC	1975-1985
	Houston-Dallas	29°N, 95°W		1-2 / week		1994-1996
Pretoria		26°S, 28°E	200	1 / week	ECC	1990-1993
	Johannesburg	26°S, 28°E		2 / week		1995-1996
Goose Bay		53°N, 60°W	0	3-4 / month	ECC	1980-1993
	cruise	53°N, 60°W ± 2.5°		2-3 / week		1994-1996
Biscarosse		44°N, 1°W	0	1 / week	BM	1976-1982
	cruise	44°N, 1°W ± 2.5°		2-3 / week		1994-1996
Poona		19°N, 74°E	0	0-1 / month	BM/Ind	1966-1986
	cruise	19°N, 74°E ± 2.5°		2-3 / month		1994-1996

[a] Data before 1992 were corrected by [x 0.9743], at stations applying Dobson scaling, following the recommendations of WOUDC for change in absorption coefficients.

[b] No Dobson scaling applied.

Table 8. Number of Vertical Profiles Used to Build the MOZAIC Ozone Climatology at Cities and Number of Flights Available at Cruise Locations

Location	Jan.	Feb.	March	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
					Cities[a]							
Frankfurt	171	177	137	160	188	191	176	184	118	81	126	134
New York	74	54	60	103	97	85	95	106	56	40	50	53
Tokyo-Osaka	17	22	11	40	35	57	35	40	20	12	10	15
Houston-Dallas	10	12	16	33	55	60	66	52	20	19	16	7
Johannesbourg	12	8	11	21	19	12	13	12	2	8	10	12
					Cruise Locations[b]							
Goose Bay	66	64	40	64	76	57	36	46	31	17	25	25
Biscarosse	20	25	27	23	14	11	8	9	9	1	6	8
Poona	8	9	1	9	5	6	0	6	1	2	5	5

Integration of data between September 1994 and August 1996.

[a] Statistics at the 900 hPa level.

[b] Statistics at the 200 hPa level.

with season between 200 and 300 hPa: OSD are lower than MOZAIC in winter and spring, near the tropopause, while there is better agreement in autumn, and OSD are higher than MOZAIC in summer, in the upper troposphere.

5.1.3. Palestine/Houston-Dallas. To increase the data density, the MOZAIC profiles obtained at Dallas and Houston have been merged because of the proximity of the cities. The sonde data (ECC) are from periods of balloon campaigns and are irregularly spaced through the year; the MOZAIC data are sparser in winter than the rest of the year. The sets of data are about 15 years apart in the mean.

At this latitude (32°N), O_3 variations are more typical of the subtropics (Figure 12): (1) lower concentrations; (2) a higher tropopause, with a suggestion of a stratospheric signature only in March at the 250-200 hPa levels; and (3) a weak summer maximum at all levels. The agreement between the two data sets is good, with OSD exceeding MOZAIC data by 5 to 45% above the 700 hPa level (see also Plate 1).

5.1.4. Tateno/Tokyo-Osaka. The MOZAIC data from Tokyo and Osaka have been merged for the same reasons as Houston/Dallas. They correspond to 2-3 profiles /week over a shorter 17 month period because regular flights to Japan started in April 1995.

Ozone distributions observed over Japan (Figure 13) are strongly influenced in the mid and lower troposphere (1000-600 hPa) by the summer monsoon which induces a summer minimum

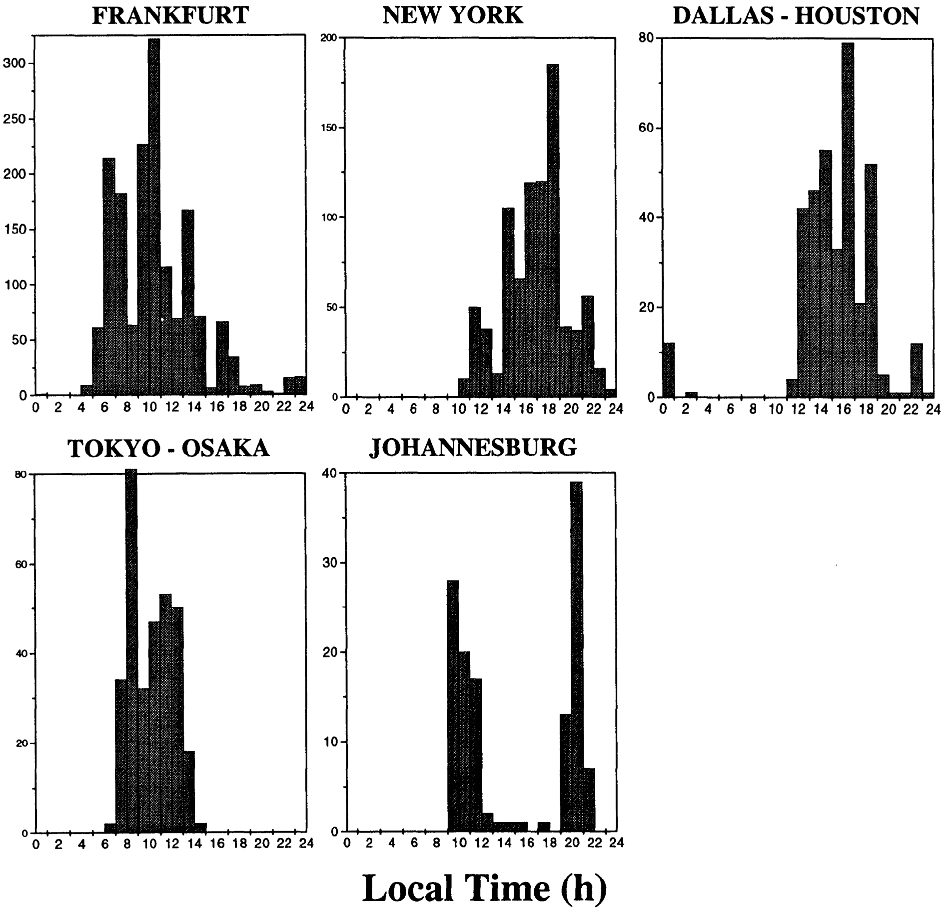

Figure 8. Distribution of the number of vertical profiles, used to build the MOZAIC ozone climatology, as a function of the time of day near MOZAIC airports.

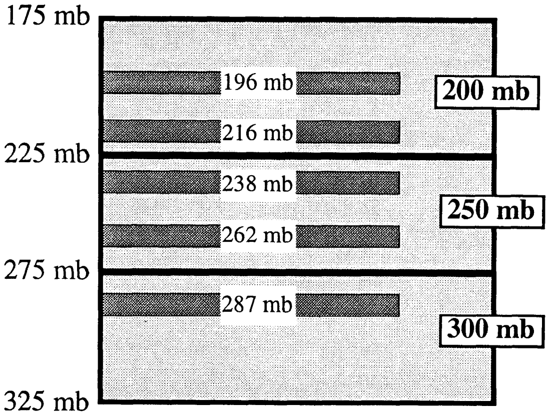

Figure 9. Position of the MOZAIC cruise levels (dark gray bars) with respect to standard pressure levels 50 hPa thick (light gray).

when the airflow from the Asian continent is replaced by the ozone-depleted air from the tropical Pacific [*Logan*, 1985; *Fukui*, 1977]. This feature appears clearly in both data sets, except for OSD in the boundary layer.

The two climatologies are very similar. Most of the time, OSD exceed MOZAIC data, by only 0-20% between the 1000 and 400 hPa levels except in summer; the large discrepancies observed in the lower troposphere in summer are attributable to the influence of the Tokyo pollution plume on the sonde measurements at Tateno (see discussion in section 7). In addition, as the airport is located northeast of Tokyo, and the aircraft land from over the Pacific, MOZAIC measurements in lower layers correspond to the sampling of cleaner oceanic air. Above the 300 hPa level there are some differences in winter/spring (see also Plate 1); the tropopause is between 215 and 290 hPa from December to April. The ozone minimum (50 ppbv) in March for MOZAIC data is not significant, since there are only five flights above 300 hPa in this month. In summer and autumn, when the tropopause is above 200 hPa (115 to 150 hPa), the agreement for 300-200 hPa is similar to that in the middle troposphere.

5.1.5. Pretoria/Johannesburg. There is only a short period of sonde data for Pretoria (1990-1993; 1 sounding/week), and MOZAIC flights to Johannesburg started in April 1995 (2 profiles/week). The measurements are all in the troposphere.

Ozone shows a spring maximum throughout the troposphere (Figure 14), which results from biomass burning combined with circulation patterns [*Diab et al.*, 1996; *Thompson et al.*, 1996]. Concentrations increase from 20-40 ppbv at the 800 hPa level (the surface is at 1500 m) to 40-80 ppbv at the 200-250 hPa levels.

The two climatologies are similar, particularly between the 800 and 600 hPa levels. Above the 400 hPa level, OSD exceed MOZAIC data by 10 to more than 45% (see also Plate 1).

5.2. Comparisons at Cruise Locations

The comparisons OSN/MOZAIC at cruise locations were made for upper levels only (300 to 200 hPa). The amount of data is lower than for cities and depends on location (see Tables 7, 8, and 9). Statistics are low at Poona, but better at Goose Bay, and Biscarosse.

5.2.1. Goose Bay. Considering the high latitude and altitude, the air sampled is mostly of stratospheric origin (100-500 ppbv ozone); the tropopause is ~290 hPa in winter and ~230 hPa in late summer above Goose Bay (Figure 15). As at the other northern hemisphere stations, a broad spring maximum is found. The sonde data exceed in the MOZAIC data by 5 to 50%. Mean values are within one standard deviation of the mean for the other data set.

5.2.2. Biscarosse. This station (44°N; 1°W) is not far from Hohenpeissenberg (48°N; 11°E), but the data are from an earlier period (1976-1982). Despite the difference in time period, and the sparse sampling of the MOZAIC data in the second half of the year (Table 8), there is reasonable agreement between the data sets at 300 hPa, which is in the troposphere year-round (Figure 16). There are differences as high as 100% in spring above 300 hPa, but the mean values agree within their error bars; agreement is better in other seasons.

5.2.3. Poona. Soundings are made with the Indian version of the BM sonde. The agreement between OSD and MOZAIC is reasonable considering the data scarcity for both climatologies (Tables 8 and 9), the 10/20 year lags between the two series, and the lower precision of the Indian sonde (see subsection 3.3). The levels shown here are in the troposphere (Figure 17). The sonde data generally exceed the MOZAIC data, but both show high variability and have poor statistics.

5.3. Summary

The similarity between the ozone variations obtained from the two data sets is striking, in terms of both ozone average concentrations and seasonal cycles. The variations of ozone, as a function of season, altitude, and latitude, in both the troposphere and the lower stratosphere, are captured rather well by both data sets. The mean differences between the data sets in the troposphere are between 3 and 13%, with the sonde data systematically exceeding the MOZAIC data; discrepancies are larger in the boundary layer and in the upper troposphere/lower stratosphere, and are not systematic in sign (Table 10).

The variability of MOZAIC data appears somewhat higher than the sonde data, which can probably be attributed to higher statistics and much better time resolution for MOZAIC. The sonde data archived at the World Ozone and Ultraviolet Radiation Data Center (WOUDC) generally have vertical resolution of about 300 m or less.

The reasons for these discrepancies must be identified, in order to understand if they correspond to differences in data

Table 9. Number of Ozone Soundings Used to Make the Ozone Climatology at the Eight Stations of the Ozone Sounding Network

Stations[a]	Jan.	Feb.	March	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
Hohenpeissenberg	169	158	165	153	112	100	109	105	115	106	159	164
Wallops Island	34	36	34	32	45	24	28	33	26	34	34	28
Tateno	52	42	40	40	38	20	23	18	34	36	41	43
Palestine	0	0	7	4	25	39	9	5	32	51	12	0
Pretoria	10	11	7	7	13	13	8	6	15	23	11	12
Goose Bay	46	43	50	52	48	42	41	44	51	39	43	48
Biscarosse	22	24	33	27	33	33	29	19	32	33	31	20
Poona	9	9	8	11	5	6	13	10	16	16	17	12

[a] See periods of the series in Table 7.

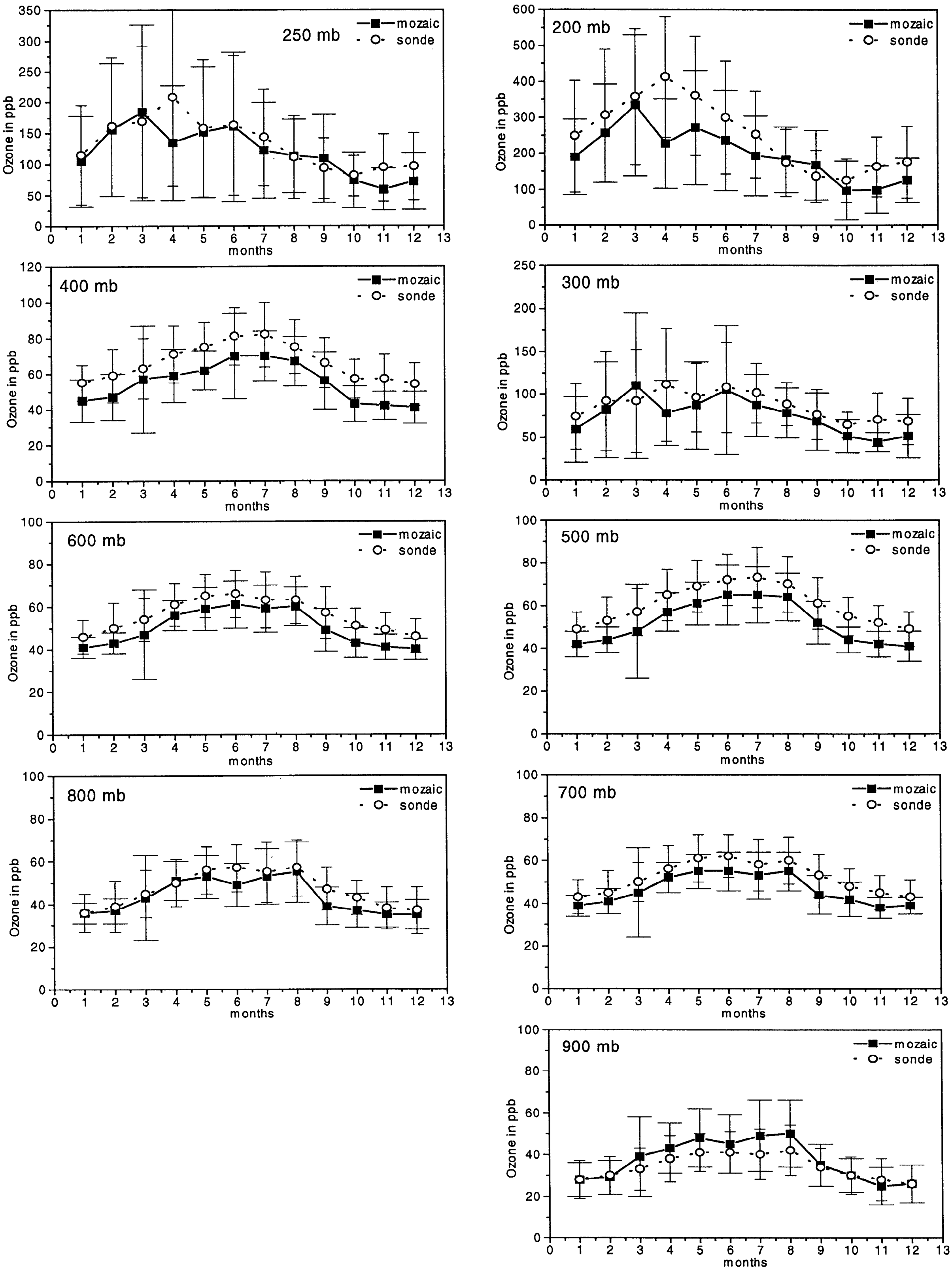

Figure 10. Seasonal variations in monthly ozone averages, at each standard pressure level, for the Hohenpeissenberg sounding station (26°N; 28°E) (1980-1993) compared with MOZAIC data over Frankfurt (September 1994 to August 1996). Standard deviations are plotted as error bars with large cap for MOZAIC and small cap for OSD (as Hohenpeissenberg is at 975 m altitude, there is no plot for 1000 hPa).

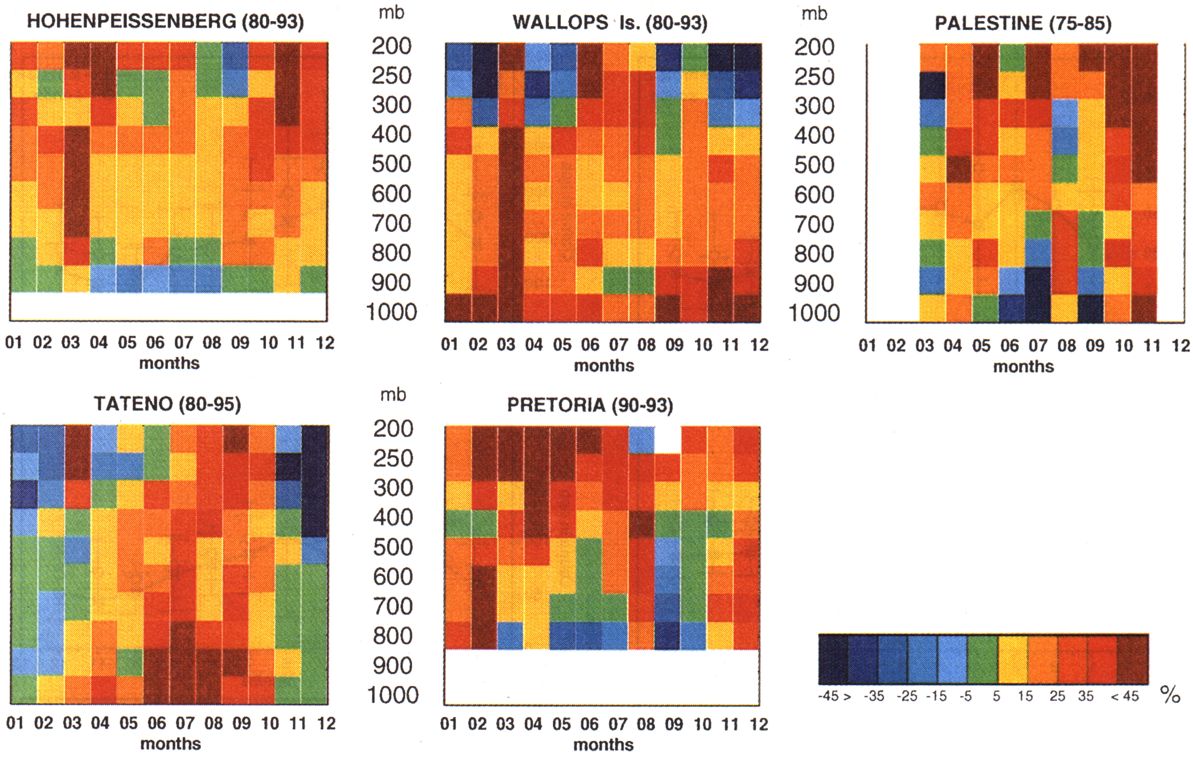

Plate 1. Comparison between ozone sounding data (1975 to 1995) and MOZAIC data (September 1994 to August 1996) at five locations. The percent difference between OSD and MOZAIC is calculated from the largest of the OSD/MOZAIC or MOZAIC/OSD ratios; that is, positive values correspond to OSD higher than MOZAIC, and negative ones correspond to MOZAIC higher than OSD.

collection (period, frequency, local conditions) or if they represent real biases between the techniques. Data collected by both methods at two locations are discussed in the next section.

6. Comparison at Hohenpeissenberg/Frankfurt and Wallops Island/New York for 1994-1995

Comparisons of sonde/MOZAIC data were made for September 1994 to October 1995 for Hohenpeissenberg, and for September 1994 to May 1995 for Wallops Island; these were the most recent data at WOUDC when the study was initiated (and still are for Wallops Island). Hohenpeissenberg is about 250 km east of Frankfurt, and Wallops Island is about 350 km south of New York. The aircraft ascents and descents extend horizontally over 300-400 km, and the balloon soundings are generally obtained within 60 km of the station. This means that the MOZAIC profiles are typically 350 km northeast of Hohenpeissenberg and 500 km north of Wallops Island, increasing to 400-550 km and 700-800 km, respectively, for the upper part of the profile.

Two types of comparisons were made. The first involved profiles with an overlap of less than 2 hours: 75 cases for Hohenpeissenberg/Frankfurt, and nine cases for Wallops Island/New York. The second used all the data for the selected period: 125/1021 profiles for Hohenpeissenberg/Frankfurt, respectively, and 16/234 profiles for Wallops Island/New York.

6.1. Individual Profiles

Figure 18 presents some examples of simultaneous MOZAIC and Hohenpeissenberg profiles. Sonde data with the vertical resolution available at WOUDC and ozone reduced data (150 m vertical integration) for MOZAIC flights are shown as function of altitude (0-200 hPa). There is in general good similarity between pairs of vertical profiles, with some discrepancies, mainly in upper layers, caused by the sampling of distinct air masses. This is not surprising because no attempt was made to select data on the basis of air origin. Larger differences correspond to cases where there are sharp gradients in the tropopause field. For example, in Figure 19 (case 2), the aircraft and the balloon encountered the tropopause near 400 hPa, and the ozone profiles agree; in Figure 19 (case 3) the aircraft encountered the tropopause at 220 hPa, and the balloon at 300 hPa, and there is a large discrepancy in ozone between these levels. There is usually a dramatic change in gradient in ozone near the thermal tropopause. Significant discrepancies (Hohenpeissenberg lower than MOZAIC) are also found in lower layers, due to different boundary layer air composition and altitudes: Hohenpeissenberg and Frankfurt airport are distant from 250 km and located at 975 m and 111 m altitude, respectively. Excluding the discrepancies in upper and lower layers, the agreement between the sonde and MOZAIC data looks good in most of the free troposphere and lower stratosphere when reliable comparisons can be made.

6.2. Seasonal Variations

To quantify the discrepancy between the data sets, we compare average values obtained by data integration over the 14 month period for two cases: (1) all data and (2) profiles matched in time. Results are shown for 500 hPa for Hohenpeissenberg in Figure 19. The left panel shows all data, and the right panel shows the selected data. Sonde data are shown with and without the scaling to the correction factor for each case. For each case there is better agreement between the sonde and MOZAIC data if the sonde data are not scaled to the ozone column.

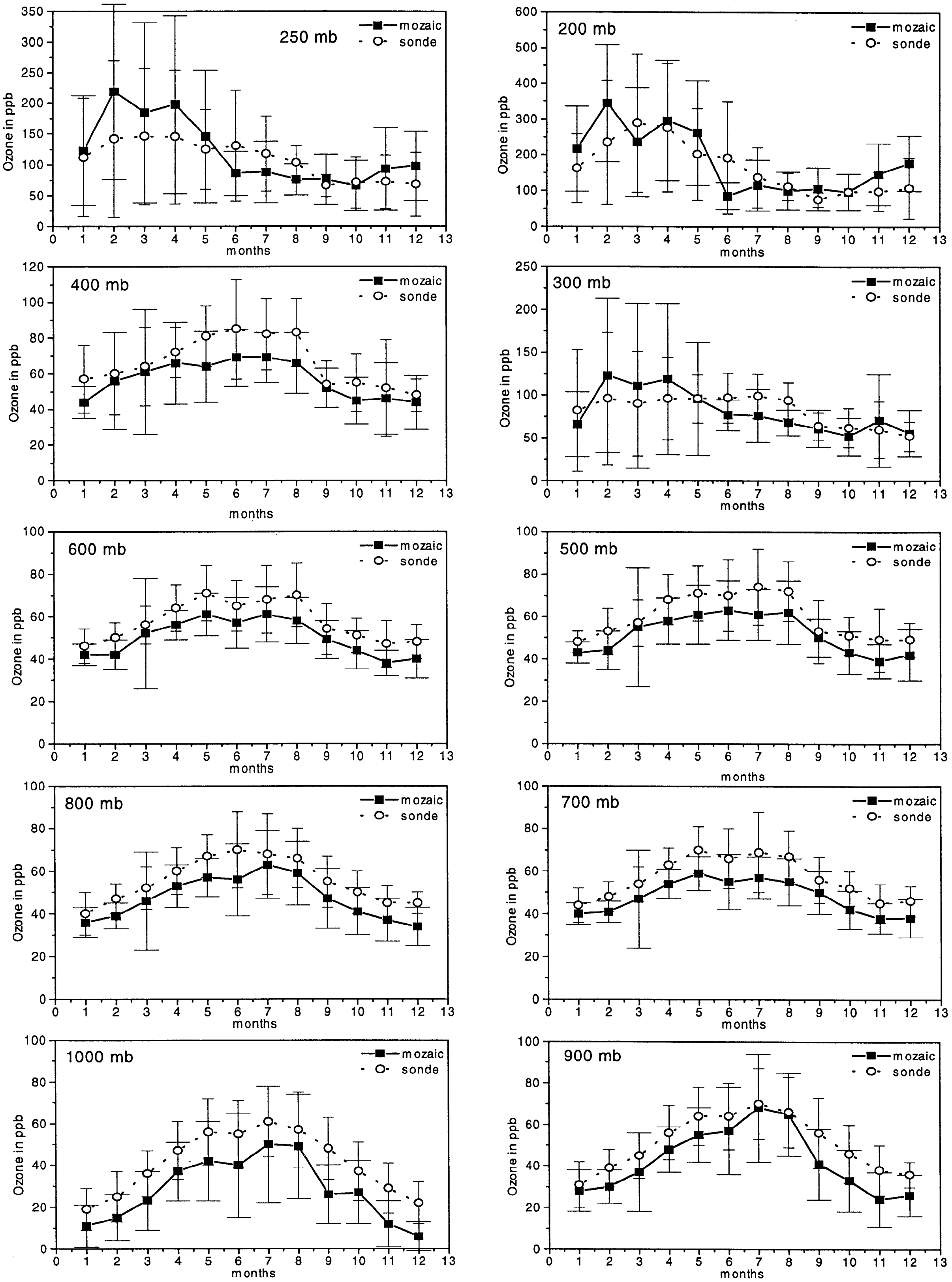

Figure 11. Seasonal variations in monthly ozone averages, at each standard pressure level, for the Wallops Island sounding station (38°N; 76°W) (1980-1993) compared with MOZAIC data over New York (September 1994 to August 1996).

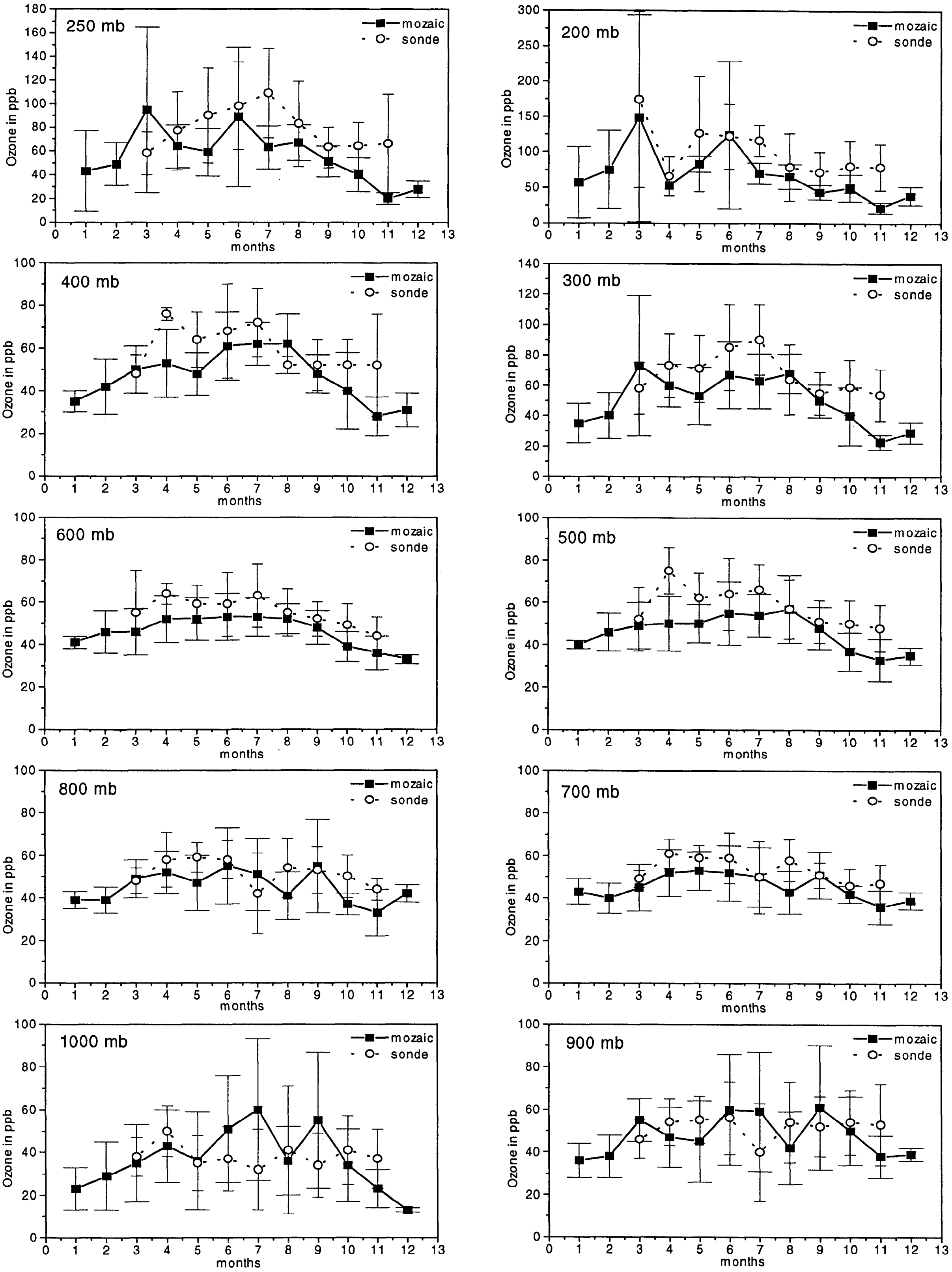

Figure 12. Seasonal variations in monthly ozone averages, at each standard pressure level, for the Palestine sounding station (32°N; 96°W) (1975-1985) compared with MOZAIC data over Dallas/Houston (September 1994 to August 1996).

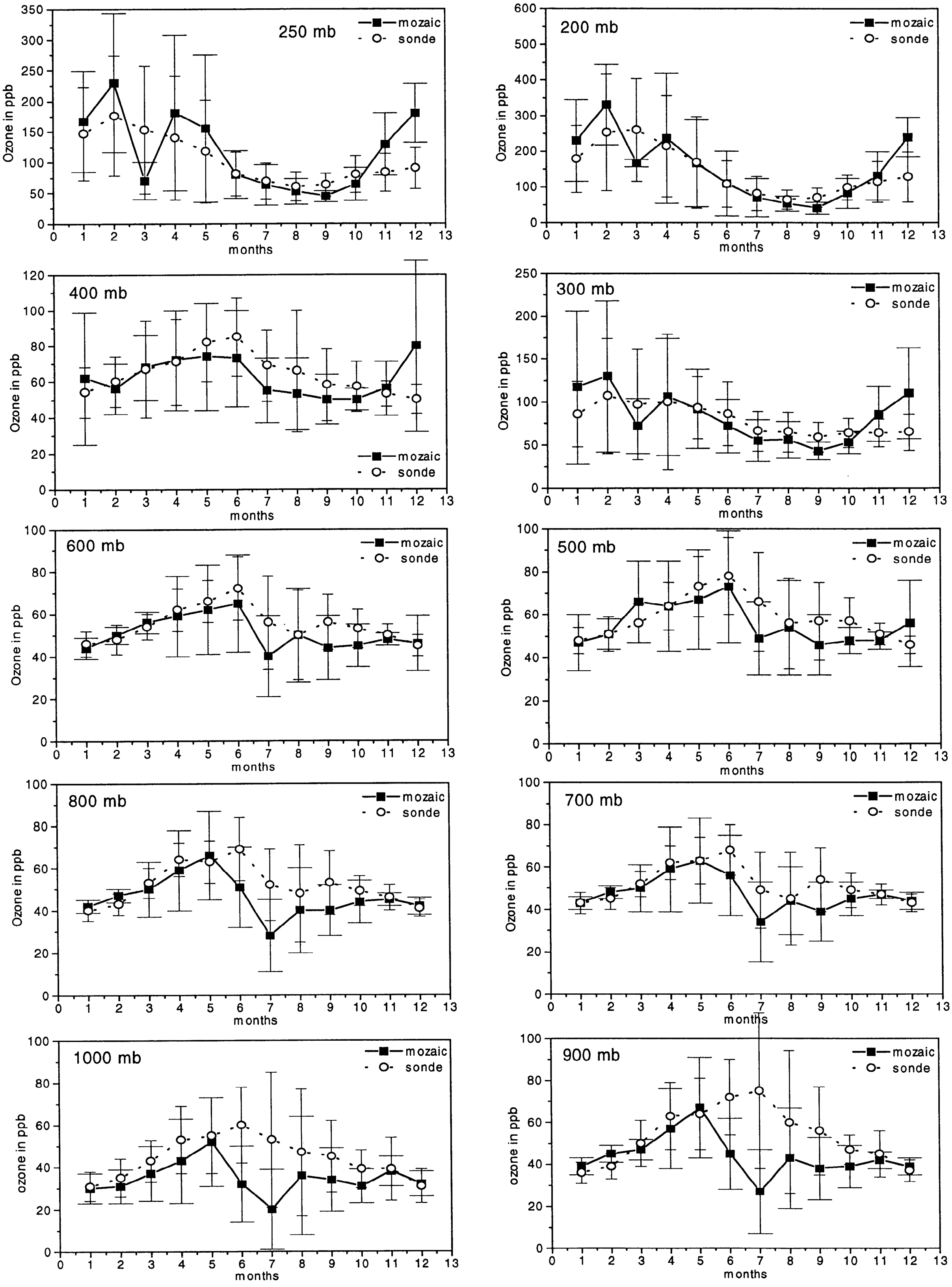

Figure 13. Seasonal variations in monthly ozone averages, at each standard pressure level, for the Tateno sounding station (36°N; 140°E) (1980-1995) compared with MOZAIC data over Tokyo/Osaka (April 1995 to August 1996).

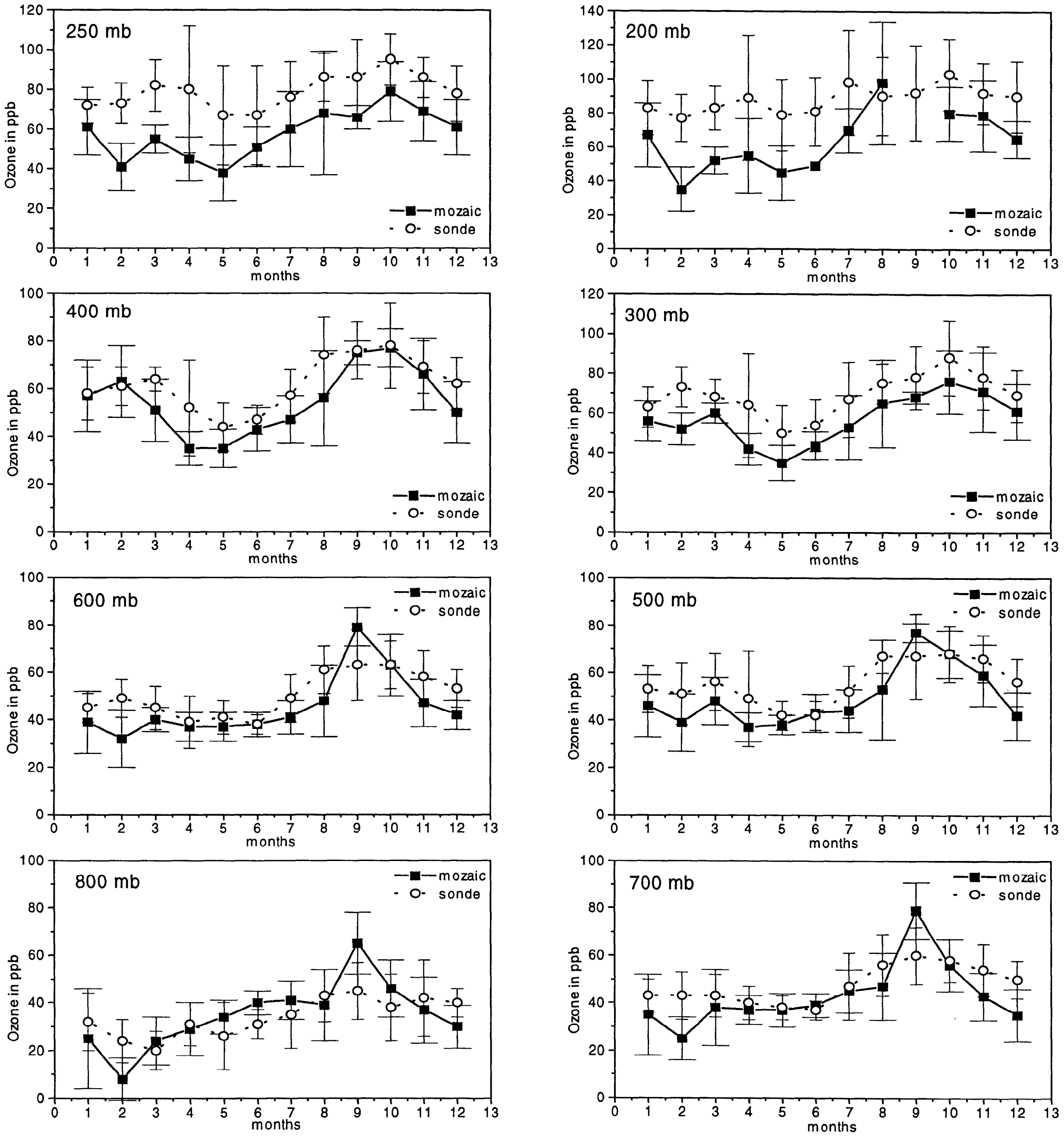

Figure 14. Seasonal variations in monthly ozone averages, at each standard pressure level, for the Pretoria sounding station (26°S; 28°E) (1990-1993) compared with MOZAIC data over Johannesburg (April 1994 to August 1996) (note that there are not 1000 and 900 hPa levels due to location of the station in altitude).

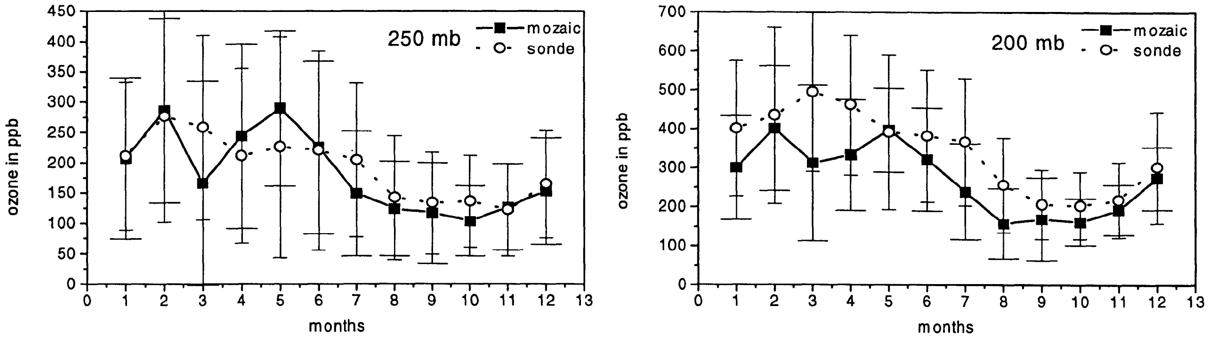

Figure 15. Seasonal variations in monthly ozone averages, at each standard pressure level, for the Goose Bay sounding station (53°N; 60°W) (1980-1993) compared with MOZAIC data at cruise levels (September 1994 to August 1996).

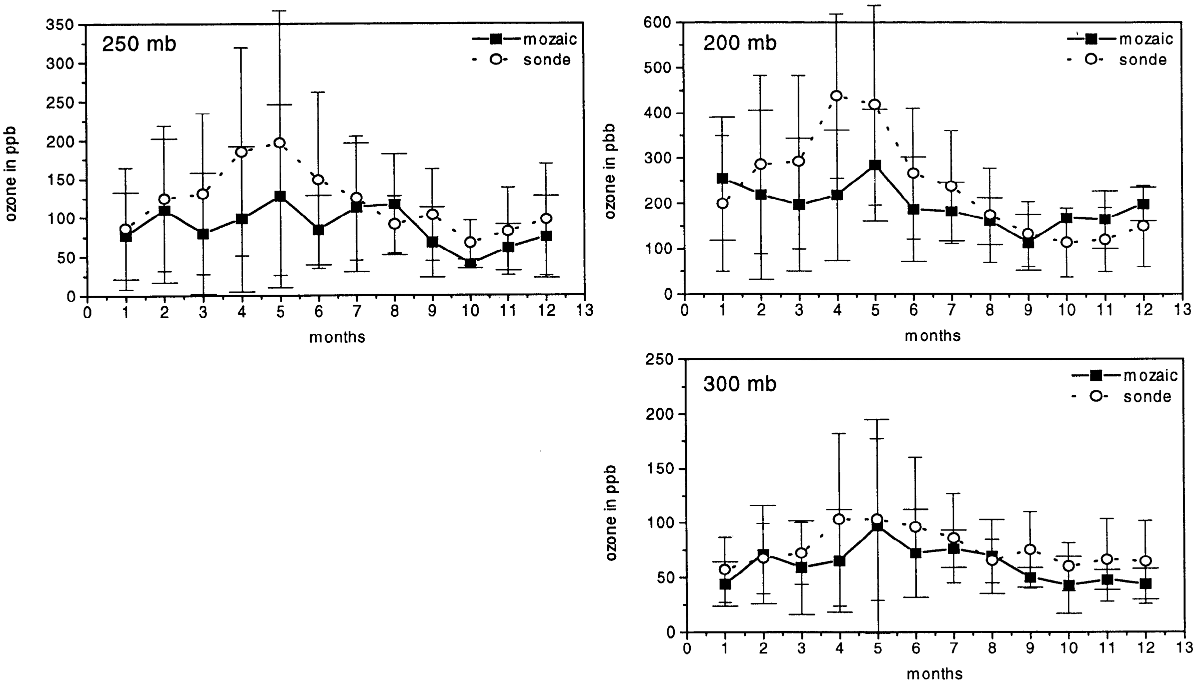

Figure 16. Seasonal variations in monthly ozone averages, at each standard pressure level, for the Biscarosse sounding station (44°N; 1°W) (1976-1982) compared with MOZAIC data at cruise levels (September 1994 to August 1996).

6.3. Average Ozone Profiles

Average ozone profiles are very similar for all data and for data matched in time, for both pairs of locations. Figure 20 shows the average MOZAIC and ozone sonde profiles (and standard deviations) for the data matched in time; the relative differences [OSD-MOZAIC] are shown in the left panels. Differences for Hohenpeissenberg are shown with and without application of the CF to the sonde data. Table 11 gives the mean relative deviation between the two data sets in the boundary layer, free troposphere, and upper troposphere (UT)/lower stratosphere (LS).

Reliable comparisons can best be made in the free troposphere, as discussed above. The discrepancies in the UT/LS at Wallops Island/New York are much larger than those found for

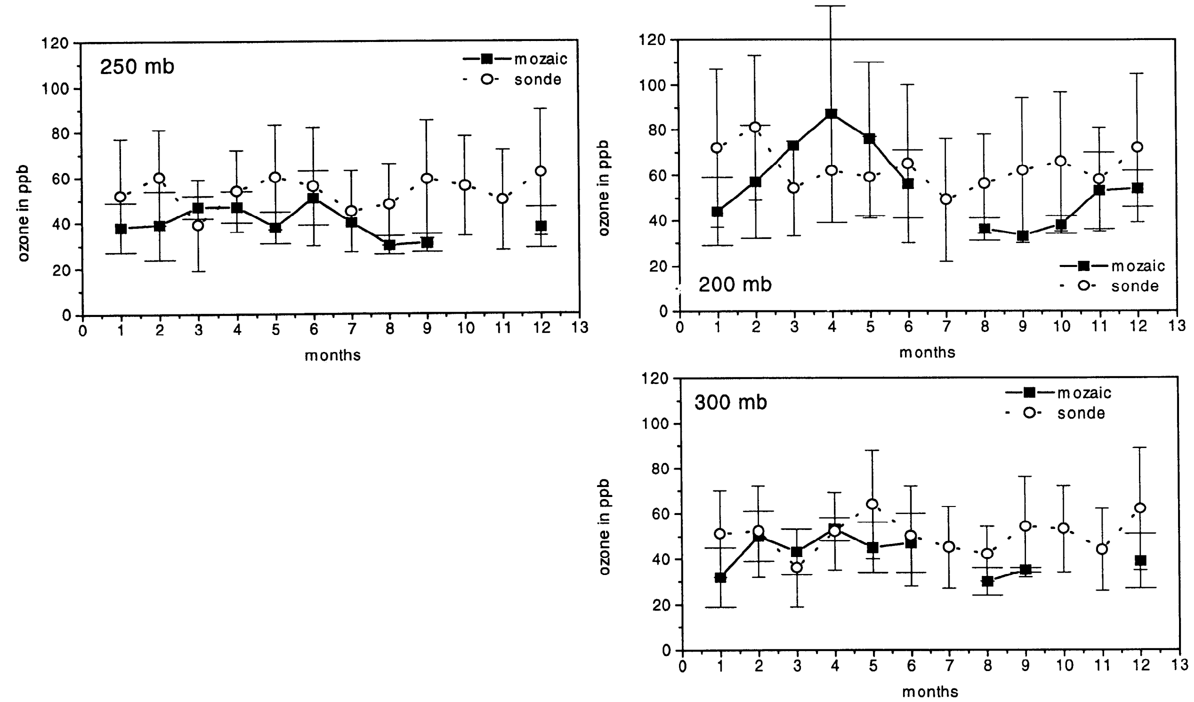

Figure 17. Seasonal variations in monthly ozone averages, at each standard pressure level, for the Poona (19°N; 74°E) (1966-1986) sounding station compared with MOZAIC data at cruise levels (September 1994 to August 1996).

Table 10. Percentages of the Difference Between Ozone Sonde and MOZAIC Data, and Mean Values of the Correction Factor (CF)

Ozone Sounding Station[a]		MOZAIC Cities or Cruise Location		Sonde Type	CF	BL[a]	FT[a]	UL[a]
Location	Period	Location	Period					
Hohenpeissenberg	1980-1993	Frankfurt	Sept. 1994 to Aug. 1996	BM	1.08	- 8	11	18
Wallops Island[b]	1980-1993	New York	Sept. 1994 to Aug. 1996	ECC		31	12	-11
Tateno	1980-1995	Tokyo-Osaka	April 1995 to Aug. 1996	ECC/KC79	0.99	26	3	-10
Palestine	1975-1985	Houston-Dallas	Sept. 1994 to Aug. 1996	ECC	1.01	-10	13	23
Pretoria	1990-1993	Johannesburg	April 1995 to Aug. 1996	ECC	1.05	N.D.	13	38
Goose Bay	1980-1993	Goose Bay	Sept. 1994 to Aug. 1996	ECC	1.01	N.D.	N.D.	15
Biscarosse	1976-1982	Biscarosse	Sept. 1994 to Aug. 1996	BM	1.17	N.D.	N.D.	22
Poona	1966-1986	Poona	Sept. 1994 to Aug. 1996	BM/Ind	1.12	N.D.	N.D.	18

N.D., no data; difference in percent between OSD and MOZAIC is calculated from the largest of the OSD/MOZAIC or MOZAIC/OSD ratios: that is, positive values correspond to OSD higher than MOZAIC, and negative ones correspond to MOZAIC higher than OSD.

[a] Boundary layer, BL: 1000-900 hPa; free troposphere, FT: 800-300 hPa; upper layers, UL: 250-200 hPa.

[b] No correction factor applied.

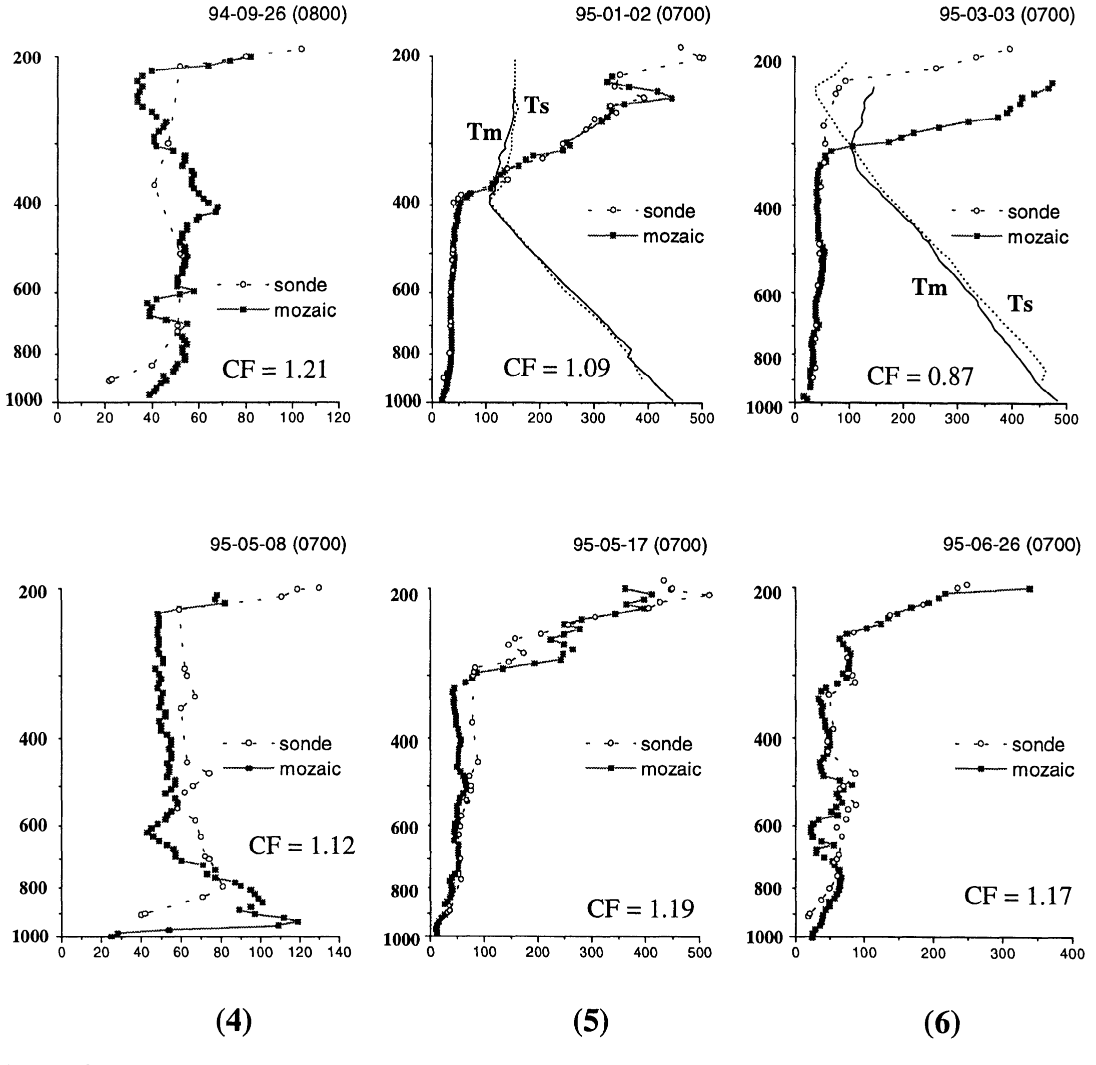

Figure 18. Examples of ozone vertical distributions at Hohenpeissenberg (48°N; 11°E) and ozone MOZAIC profiles near Frankfurt (50°N; 9°E), with values of ozone sonde Dobson correction factor CF (the vertical profiles of temperature for sonde Ts and MOZAIC Tm were plotted for cases 2 and 3).

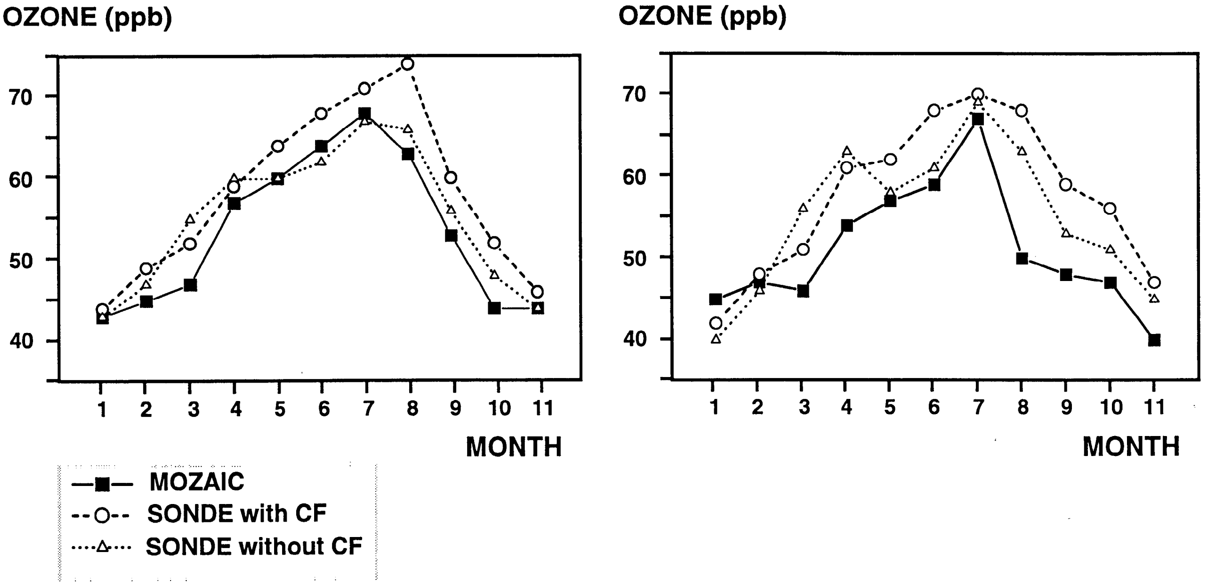

Figure 19. Seasonal variations in monthly ozone averages at the 500 hPa level for MOZAIC data (UV photometer) in the area of Frankfurt (50°N; 9°E) and ozone sonde data (Brewer-Mast) at Hohenpeissenberg (48°N; 11°E) for the period August 1994 to October 1995. (left) All data available considered; (right) selection of 75 cases of simultaneous profiles, with and without application of Dobson correction factor CF.

Hohenpeissenberg/Frankfurt, which can be explained by the larger distances involved (see Table 7) and by the existence of a sharp southward gradient of ozone at the tropopause level over the eastern coast of the United States (see discussion in subsection 7.2.2), linked to the vicinity of polar front boundary [*Thouret et al.*, this issue].

The difference between the Hohenpeissenberg and MOZAIC (Frankfurt) data in the free troposphere is reduced from 8 to 4% by removal of the Dobson normalization. Differences for Wallops Island are somewhat larger, 14% for all data, and 10% for selected data; the number of profiles available for the latter pair of sites is much lower than for the former.

Analysis of individual pairs for Hohenpeissenberg confirms that agreement is better when the data are not scaled to the ozone column. There is better agreement with MOZAIC data for 62% of the cases in the free troposphere (winter 58%, spring 33%, summer 72%, and autumn 83%) but only 36% in the upper troposphere/lower stratosphere, due certainly to the vicinity and variability of the tropopause at this level.

6.4. Discussion

A possible evolution of the background current, during the ascent of the balloon, was mentioned (see subsection 4.2) as a cause of uncertainty in ozone sonde measurements. If so, this should produce an evolution with time, and thus with altitude, in the discrepancy between MOZAIC and ozone sonde data. Figure 21 highlights the ozone differences Δ (in ppbv) between Hohenpeissenberg (OSD) and Frankfurt (MOZAIC) for 800-300 hPa. An increase of Δ with altitude is observed, the absolute discrepancy being reduced if the CF is not applied.

Another reason for systematic discrepancies between the sonde and MOZAIC measurements is a spatial gradient in the ozone field. This was examined by comparing MOZAIC data from Frankfurt and Paris (Figure 21, top). The increase with altitude for Δ[Frankfurt - Paris] is similar to that for Δ[Hohenpeissenberg-Frankfurt]. If the zonal gradient southeast of Frankfurt is of similar magnitude, the bias between Hohenpeissenberg and Frankfurt could be due entirely to spatial variations in ozone. The same explanation could apply to the 10-14% discrepancy observed between Wallops Island and New York, and this will be examined with more recent MOZAIC data for the eastern United States.

This study confirms the good agreement between the two methods, well within the range of uncertainty, when taking into consideration the various causes of differences: uncertainties in both methods, Dobson normalization, and geographical variations in ozone.

7. General Discussion

The comparison between the two climatologies shows strong similarities between the sonde and MOZAIC data, with the sonde data being 3-13% higher in the middle troposphere. Discrepancies are larger in the boundary layer and in the upper layers (Table 10). Here we discuss potential causes for these discrepancies.

7.1. Boundary Layer

The time at which measurements are made could influence the discrepancies between the sonde and MOZAIC data. Ozone concentrations are in general lower during the night and in the early morning because of the thinness of the boundary layer (physical and chemical losses). Concentrations increase during the morning after the breakup of the nighttime inversion layer because ozone is produced photochemically and the region which was isolated above the inversion mixes down. This results in a diurnal variation in surface ozone, with highest values in the afternoon.

Times at which ozone soundings and MOZAIC profiles are made are given in Table 12. The times for the sondes tends to be earlier for Hohenpeissenberg and Palestine, which is consistent with the sondes measuring lower values for ozone, and later for Tateno, consistent with the sondes measuring more ozone. The time of aircraft data at Johannesburg is bimodal, with the morning flights coinciding with the launch time of the sondes. For Wallops Island, most sondes are launched in late morning or early afternoon, but there is a second peak of launches in the evening. The aircraft data tend to be later than the majority of sonde

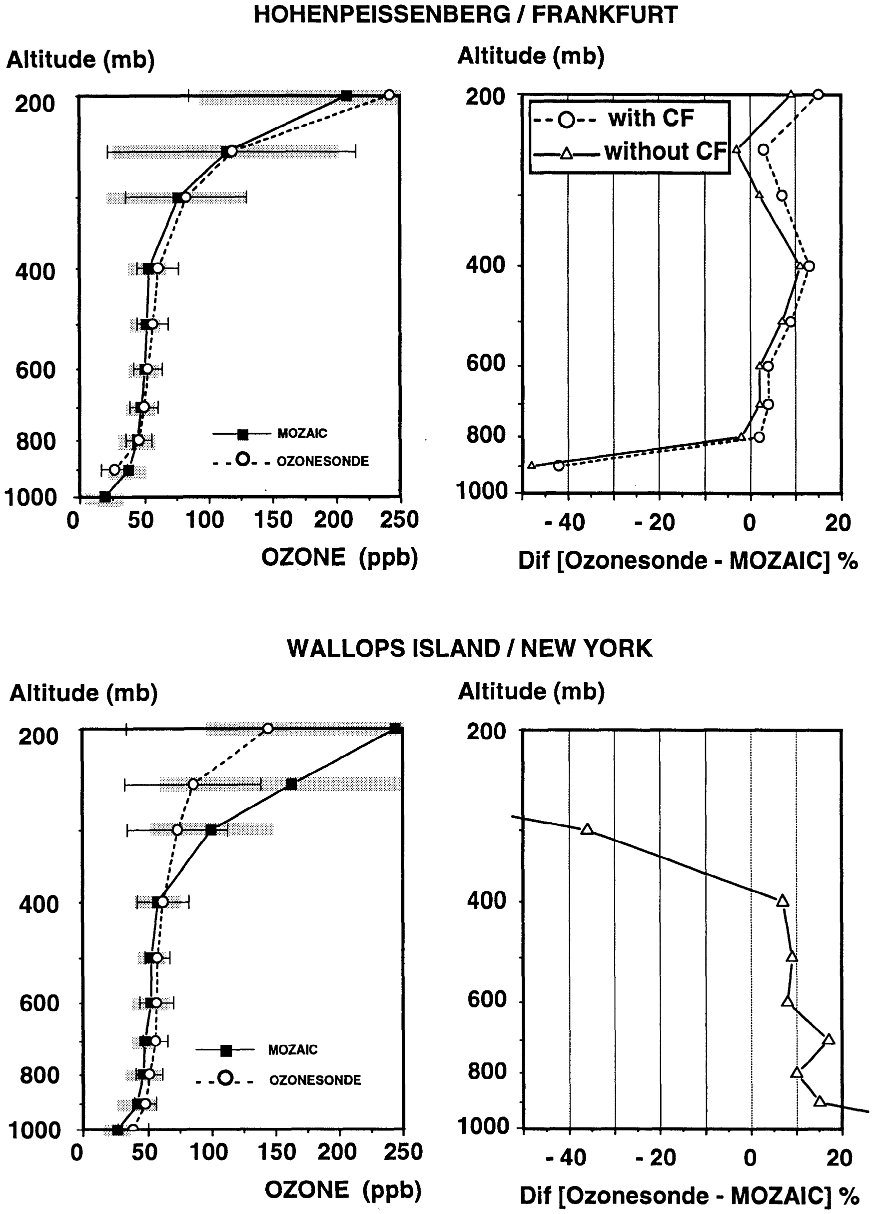

Figure 20. Comparison for the period September 1994 to October 1995 between (top) ozonesonde data at Hohenpeissenberg (48°N; 11°E) and MOZAIC ozone data in the area of Frankfurt (50°N; 9°E), and (bottom) ozonesonde data at Wallops Island (38°N; 76°W) and MOZAIC ozone data in the area of New York (41°N; 74°W); (left) average profiles, (right) relative difference (ozonesonde - MOZAIC) with and without application of the Dobson correction factor CF (selection of simultaneous profiles: 75 cases for Hohenpeissenberg/Frankfurt, nine cases for Wallops/New York) (standard deviations: ozone sonde, line; MOZAIC, gray bar).

Table 11. Percentages of the Mean Relative Deviation (Excess of Ozone Sonde Data (OSD) With Respect to MOZAIC Data) at Two Locations, for Different Options of Data Integration, Different Layers, and With/Without Dobson Normalization (CF)

Locations	Option[a]	Boundary Layer		Free Troposphere		UT/LS	
		With CF	Without CF	With CF	Without CF	With CF	Without CF
Hohenpeissenberg / Frankfurt	1	-37	-42	8	4	13	9
	2	-42	-48	7	4	11	5
Wallops Island[b] / New York	1		26		14		-56
	2		26		10		-69

Mean relative deviation in percent; boundary layer: 1000-900 hPa; free troposphere: 800-400 hPa; upper troposphere/lower stratosphere, UT/LS: 400-200 hPa.

[a] Option: 1, all data; 2, selected data.

[b] No correction factor applied.

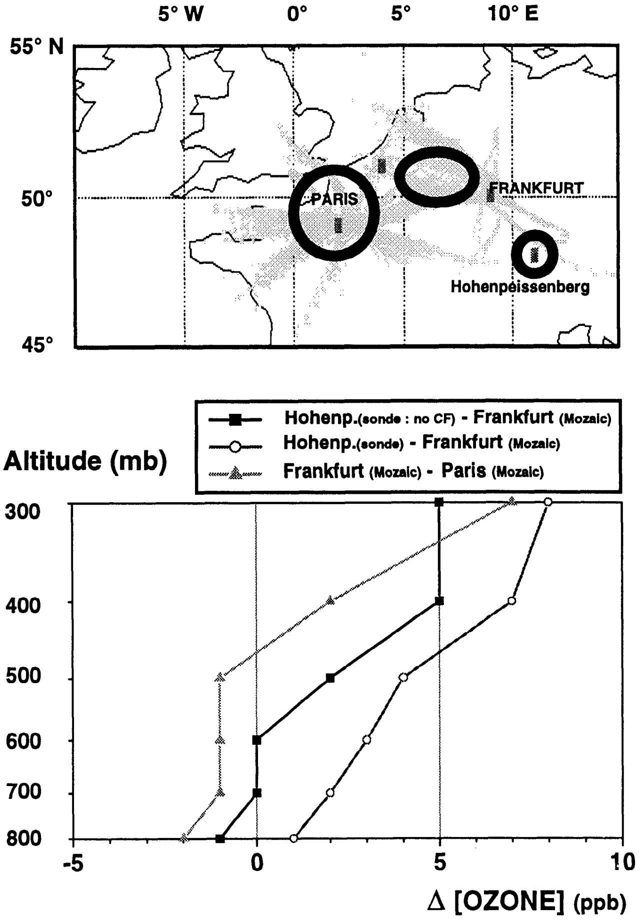

Figure 21. (top) Locations of Hohenpeissenberg station and MOZAIC airports (Frankfurt, Paris) with indications of MOZAIC flight tracks (grey) and average positions of data (solid circles) between 0 and 300 hPa. (bottom) Differences (Δ) of ozone concentrations (ppbv) between ozone profiles obtained at Hohenpeissenberg (ozone sondes, applying or not the Dobson correction CF), and MOZAIC airports Frankfurt, and Paris.

launches, which would lead to higher ozone values, all other things being equal; the opposite is observed.

The local environment may affect the regional ozone characteristics in a number of ways, and since the sonde locations and airports are a few hundred kilometers apart (Table 7), regional inhomogeneities could explain the differences. For example, Hohenpeissenberg is located at 975 m, so the 900 hPa level is closer to the surface and may be directly influenced by dry deposition, while corresponding MOZAIC profiles are obtained at higher elevations above the ground. Also, the Frankfurt location is closer to a major metropolitan area. In the case of New York and Wallops Island, both are coastal sites, but the former is within a major urban area, and the latter is downwind from one. The differences in summertime ozone at the surface are similar to those at 700 hPa. The reasons for the difference are unclear, but could reflect more efficient ozone production downwind from an urban area. In the case of Tateno and Tokyo, the sonde station is within the Tokyo urban plume, with mean concentrations of 70 ppb in summer at 900 hPa. The low concentrations measured by the aircraft may reflect the sampling of much cleaner marine air, if the planes land from over the ocean. The aircraft data are typical of values found at remote rural sites at the same latitudes in Japan [*Sunwoo and Carmichael*, 1994].

7.2. Upper Layers

OSD are higher than MOZAIC data by 15 to 38% (Table 10), except for Wallops Island and Tateno. As will be discussed in subsection 7.3, the sampling period can play an important role, but statistics and the distance between measurements should be considered.

MOZAIC data in upper layers at the seven northhemispheric stations show higher intermonthly fluctuations in winter and spring than OSD. This is probably caused by the smaller number of measurements where there are large vertical gradients in ozone at the tropopause. The mean values for these layers are an average over high ozone values in stratospheric air and lower values in tropospheric air, leading to greater variability in ozone, particularly in spring when the vertical gradients are steepest and the variability of stratospheric ozone is greatest. There is significant interannual variability in ozone in the upper troposphere/lower stratosphere [e.g., *Logan*, 1994, Figure 23], much more so than in the middle troposphere. It is not surprising that the sonde data, which correspond to averages over 10-15 years, show smoother month to month changes than the MOZAIC data, and different mean values, particularly for locations/months where the MOZAIC data are sparse.

Several sonde stations are a few hundred kilometers from the MOZAIC airports, and the distance between the measurements is even larger in the upper troposphere, as much as 300-800 km depending on the station, as illustrated in section 6. This is less of an issue for the three cruise locations, as the aircraft data were selected to be within 2.5° of the sonde station.

In the case of Tateno and Wallops Island, the MOZAIC seasonal ozone climatologies at cruise level (integration in cells 10° by 10° [*Thouret et al.*, this issue]) show the existence of a steep meridional gradient, decreasing southward (Figure 22), linked to the vicinity of the polar front. This explains why MOZAIC data are 30-40% higher than OSD in winter and spring at these stations only (Plate 1 and Figures 11 and 13). At Pretoria, a weaker but significant gradient, with ozone decreasing northward, is observed, contributing to the higher OSD values (38%). There is no systematic zonal gradient at Palestine, and gradients at Hohenpeissenberg are small as is discussed in section 6.

7.3. General Discrepancies

It is clear that OSD are generally higher than MOZAIC data in the free troposphere (0-40% for monthly discrepancies; 3-13% on average) and in the UT/LS (Table 10). Such discrepancies must be

Table 12. Discrepancies Between Ozone Sounding Data (OSD) and MOZAIC Data in the Boundary Layer, With Indications of Ozone Sounding Times at Stations and MOZAIC Flight Departure/Arrival at Airports

Ozone Sounding Station	Local Time	MOZAIC City	Local Time	Discrepancy	Period
Hohenpeissenberg	0600-0900	Frankfurt	0600-1400	OSD lower than MOZAIC	spring, summer
Wallops Island	1000-1500; 1900-2100	New York	1200-2200	OSD much higher than MOZAIC	E.S.
Palestine	0700-1300	Dallas-Houston	1200-1800	OSD lower or equal to MOZAIC	E.S.
Tateno	1500	Tokyo-Osaka	0800-1200	OSD much higher than MOZAIC	summer
Pretoria	1000-1200	Johannesburg	0900-1200; 1900-2100	OSD lower or equal to MOZAIC	E.S.

E.S., every season.

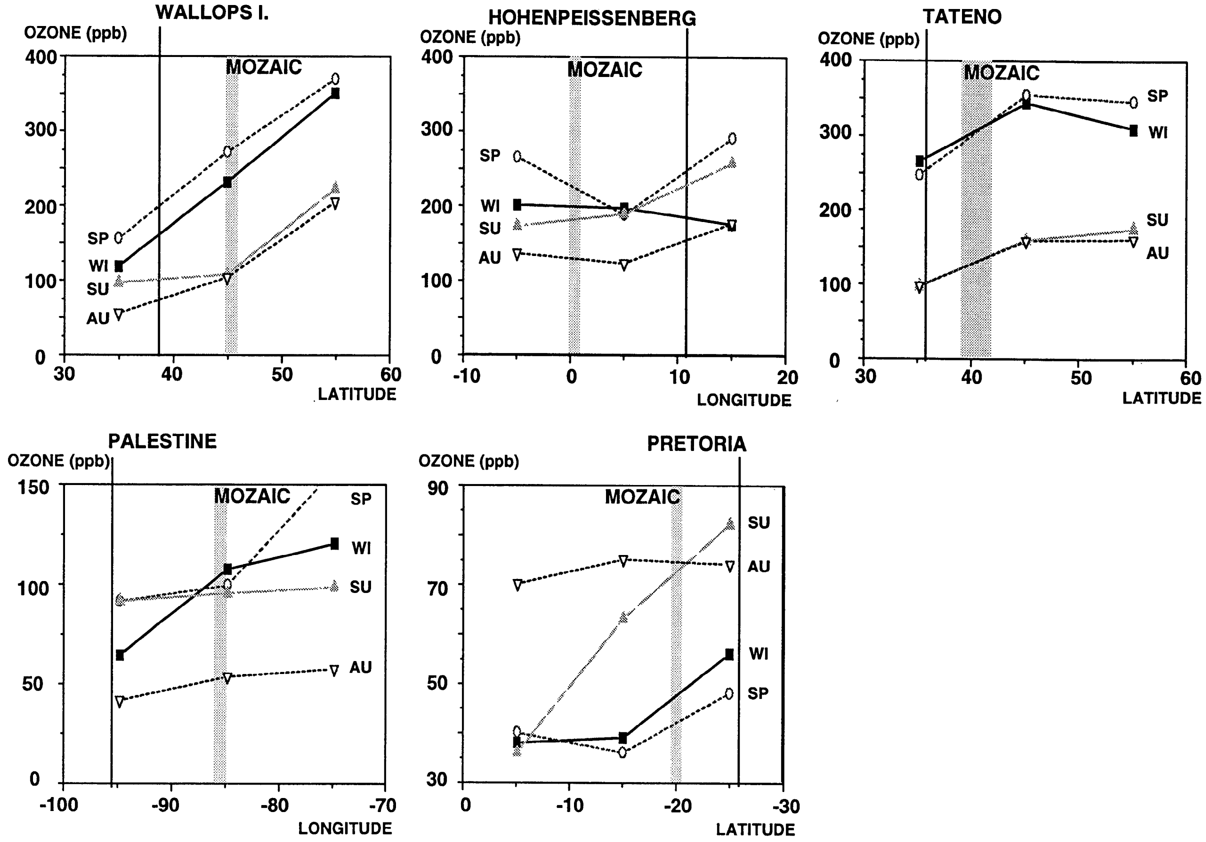

Figure 22. Variations with latitude or longitude in seasonal ozone concentrations from MOZAIC data at level 216 hPa between September 1994 and August 1996 for the five locations considered (see text) with indications of the respective locations of ozone soundings (line) and MOZAIC aircraft (gray bar) at this level (seasons refer to the northern hemisphere with WI, winter; SP, spring; SU, summer; AU, autumn).

explained in order to make reliable comparisons when evaluating 3-D models with different data sets. The differences (techniques, scaling, and time periods) in the OSN and MOZAIC climatologies have to be analyzed and their consequences evaluated.

7.3.1. Measurement techniques. Characteristics and precisions of the methods (see sections 3 and 4) show that the UV photometer used in MOZAIC is the most accurate, due to the technique itself and to the careful procedure of calibration, compared with the ECC and BM sondes.

The study at Hohenpeissenberg (BM) with data matched in time (September 1994 to October 1995) shows good agreement between the sonde and aircraft data, especially when taking into consideration possible causes of differences (Dobson normalization and geographical variations). Results for Wallops Island were also encouraging, although there were fewer contemporaneous data. Unfortunately, there are few locations with both current sonde measurements and MOZAIC airports at which such comparisons can be made; Tateno is the only other location, and there MOZAIC data start later, so inadequate statistics were available at the start of this study.

7.3.2. Correction factor. As discussed above, the scaling of the sonde data to the total ozone column may introduce a small bias in the BM tropospheric measurements. The study at Hohenpeissenberg/Frankfurt (section 6) supports such a hypothesis. Two other sonde stations, Biscarosse and Poona, also used BM sondes, but these data predate the MOZAIC data by over a decade and have larger correction factors (Table 10). BM sondes from that period clearly underestimated tropospheric ozone values compared to ECC sondes, and removing the CF would greatly increase this discrepancy. The Indian sondes did not perform particularly well in the sonde intercomparisons discussed above. At present, only a few stations in Europe still use BM sondes. Most ozonesonde stations operating today use ECC type sondes. For these the mean CF is typically 1.0 [*Logan*, 1994]. Consequently, the presence of the CF has little effect on mean values.

7.3.3. Trends in ozone concentrations. Any trend in ozone could contribute to the differences between the two climatologies, since the MOZAIC data are for a later period than the sonde data. Trends in the sonde data are reported by *Logan* [1994] for most long-term stations, by *De Muer et al.* [1994] for Uccle, and by *Ancellet and Beekmann* [1997] for Observatoire de Haute Provence (OHP). More recent results are given in a new assessment of trends in the vertical distribution of ozone [*WMO*, 1998]. Trends in tropospheric ozone since 1980 are small in most locations.

WMO [1998] shows that trends in tropospheric ozone calculated for Canadian stations are negative or near zero for the period from 1970 through 1996 and are in the range -2 to -8% per decade for 1980 to 1996; they are small and insignificant for Wallops Island for 1980 to 1995. Trends at three European stations are strongly positive for the period 1970 through 1996 but are essentially zero at Hohenpeissenberg and Uccle for 1980 to 1996. Trends at Japanese stations show a mixture of positive and insignificant trends for both time periods. For Tateno, in particular, the trend is near zero and insignificant throughout the troposphere for 1980-1996.

Ancellet and Beekmann [1997] compared results from Biscarosse with ECC sonde data from OHP, 600 km distant, for 1991 to 1995. They found a trend of 8% per decade based on annual mean values, but noted that there is no trend if they assume there is a 15% instrumental bias between the earlier BM sonde data and the ECC data.

We used the sonde data to estimate any effect of interannual variability of ozone on the results shown here. For Hohenpeissenberg and Tateno, a comparison was made between the sonde climatology and data overlapping with MOZAIC measurements. Differences in mean ozone values for 1980-1993 and 1994-1996 are less than 1% at Hohenpeissenberg between 800-300 hPa, and less than 6% at the 250-200 hPa levels. For Tateno, the differences are less than 5% between 800-300 hPa, with the later data higher in some layers and lower in others. The data overlapping with MOZAIC data are 9% higher than the data for 1980-1995 at 200 and 250 hPa; applying this factor to the results in Table 10 improves agreement for the upper layers at Tateno from -10 to -1%.

8. Conclusions

Intercomparisons for in-flight conditions do not show detectable differences between the six ozone devices operating in the MOZAIC program. A 1 ppbv agreement, on average, between the ozone measurements obtained from different devices was found experimentally at tropospheric concentrations, within the theoretical precision deduced from the addition of the individual uncertainties (5 ppbv). In the case of stratospheric measurements, a somewhat higher discrepancy is observed (17-27 ppbv), remaining, however, in the range of the evaluated precision (25 ppbv), but it might well result from the difficulty of making reliable comparisons in these regions of sharp ozone gradients. The precision and reproducibility of MOZAIC ozone data are thus suitable for building reliable ozone climatologies.

Very similar ozone seasonal variations are observed at different levels for the eight locations used in the comparison study. The agreement is best in the troposphere above the boundary layer, 3 to 13% (sonde higher), within the range of uncertainties of the two methods. Larger differences are found in the upper troposphere/lower stratosphere, with the sonde data generally higher, but these differences are not surprising, considering the different periods of observation and the large distances between measurements, particularly in cases where there are large spatial gradients in ozone (e.g., near the polar front). The comparisons are less reliable in general in the boundary layer, due to the importance of local influences.

The comparisons of contemporaneous data at Hohenpeissenberg and Wallops Island show good agreement in the free troposphere (800-300 hPa). The Hohenpeissenberg study supports the hypothesis that the Dobson scaling, applied to recent BM ozone sonde data, may not be appropriate when using the tropospheric measurements. Although other causes of discrepancies are obviously influencing the ozone profiles, such as the existence of geographical gradients in tropospheric ozone, the application of the CF contributes to an increase in the ozone concentration by about 3-6% in the case of Hohenpeissenberg.

What is the impact of the differences between the sonde and MOZAIC data in the context of using these data for model evaluation? We believe it to be small. The data agree well in the free troposphere. In the boundary layer, neither sonde data, nor data in the vicinity of major airports, are well suited for model evaluation; data from rural surface sites with 24 hour operation are generally used for this purpose. Considering the bimodal distribution of data in the UT/LS and the few number of annual cycles (2 years) for MOZAIC, it is not surprising that the data sets do not agree well in this region as discussed above.

The sonde data sets are complementary, in that the sonde data provide long-term averages, while the aircraft data give high-frequency data for a shorter time series, and in particular give better spatial coverage for altitudes of 9-12 km, a critical region for both S/T exchanges and aircraft emissions, and over remote oceanic areas which are not covered by the ozone sounding network. In this sense, the sonde data are suitable for evaluation of models based on winds from a general circulation model, while the aircraft data offer denser data for evaluation of models based on assimilated meteorological products for specific recent periods. However, given the scarcity of ozone data for much of the troposphere, it is likely that both types of data will be used to evaluate both types of models. As the MOZAIC program continues, the statistics for any one location will improve, and more tropical locations are now visited by the aircraft than at the start of the program.

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