Atmos. Meas. Tech. Discuss., 6, 7099–7148, 2013
www.atmos-meas-tech-discuss.net/6/7099/2013/
doi:10.5194/amtd-6-7099-2013

Atmospheric Measurement Techniques Discussions
Open Access

Trajectory matching of ozonesondes and MOZAIC measurements in the UTLS – Part 2: Application to the global ozonesonde network

J. Staufer[1], J. Staehelin[1], R. Stübi[2], T. Peter[1], F. Tummon[1], and V. Thouret[3]

[1]Institute for Atmospheric and Climate Science, ETH Zurich, Zurich, Switzerland
[2]Federal Office of Meteorology and Climatology MeteoSwiss, Aerological Station, Payerne, Switzerland
[3]Laboratoire d'Aérologie, Université de Toulouse, CNRS, Toulouse, France

Received: 4 June 2013 – Accepted: 8 June 2013 – Published: 1 August 2013

Correspondence to: J. Staufer (johannes.staufer@env.ethz.ch)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Ozone, an important greenhouse gas, has the largest climate forcing in the tropopause region, meaning that knowledge of long-term ozone changes in the upper troposphere/lower stratosphere (UTLS) is particularly important. Here, we perform a 16 yr comparison (1994–2009) of UTLS ozone measurements from balloon-borne ozonesondes and MOZAIC (measurements of ozone, water vapor, carbon monoxide and nitrogen oxides by in-service Airbus aircraft). The analysis uses trajectories computed from ERA-Interim wind fields to find matches between the two measurement platforms. Ozonesonde data quality is most critical in the UTLS, where natural variability is high, particularly close to the tropopause. On average, at the 28 launch sites considered, ozone mixing ratios measured by the sondes exceed MOZAIC data by 5–15 %, with differences being smaller in the LS than in the UT at many launch sites. For most sites, sondes and MOZAIC data are in close agreement after 1998. Before 1998 ozone mixing ratios measured by the Brewer–Mast (BM) sondes and Electrochemical Concentration Cell (ECC) sondes are systematically (up to 20 %) higher than the MOZAIC UV photometers. The reason for this large difference remains unclear. Results also show that after 1998 large background current signals may affect ozonesonde performance, limiting the determination of reliable ozone trends in the UTLS. Sonde measurements appear to be insensitive to changing the type of ECC ozonesonde, provided the cathode sensing solution strength remains unchanged. Only Scoresbysund (Greenland) showed systematically higher readings after changing from Science Pump Corporation sondes to ENSCI Corporation sondes, while keeping a 1.0 % KI cathode electrolyte. This suggests that ECC sondes, provided their background current and sensing solutions are properly monitored, are robust and reliable tools for ozone trend studies in the UTLS.

Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper |

AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Stauffer et al.

1 Introduction

Over the last 40 yr electrochemical ozonesondes have been widely used for measuring upper-air ozone (O_3), up to the burst of the balloon at altitudes of 30–35 km. Electronically coupled with a standard meteorological radiosonde for data transmission to the receiver at ground, they provide accurate measurements of O_3, with a typical vertical resolution of 100–200 m. Ozonesondes provide unique information that can be used to produce O_3 climatologies, validate satellite measurements, establish longterm atmospheric changes and trends, and for comparison with numerical model simulations.

Three main types of electrochemical ozonesondes have been developed since the 1960s: the Brewer Mast (BM, Brewer and Milford, 1960), the Electrochemical Concentration Cell (ECC, Komhyr, 1969) and the Japanese ozonesonde (KC, Kobayashi and Toyama, 1966). At present, most sites use ECC sondes, and, since 2010, KC ozonesondes are longer used operationally. The principle of operation is based on the titration of O_3, either in a potassium iodide (KI) sensing solution (ECC and BM sondes) or in a potassium bromide solution (KC sondes) (Smit et al., 2011). For each molecule of O_3 entering the solution, two iodide ions (I^-) are oxidized to form iodine (I_2), which is subsequently reduced back to I^- at the electrodes, generating an electric current of a few microamperes. This current is measured, and by assuming a 100 % reaction yield, can directly be related to the atmospheric O_3 partial pressure. Uncertainties may change during flight as the pump efficiency degrades with increasing altitude, or due to inaccurate pump temperature measurements or the presence of a background current that is subtracted from the measured current (Smit et al., 2007). The background current has largest influence on the overall accuracy at low O_3 concentrations and therefore becomes particularly important in the tropical troposphere and below the mid-latitude tropopause (e.g. Smit et al., 2011). Conversely, the pump efficency becomes the predominant uncertainty in the stratosphere (e.g. Stübi et al., 2008).

Although the primary principle of operation has not changed, ozonesondes have undergone several modifications, including changes to manufacturing, preparation, solution concentration and data processing, all of which may have affected the accuracy of the various sonde types and in turn the long-term trends estimated using these data (Smit et al., 2007). Over the past decades various research groups have put considerable effort into quantifying the precision and accuracy of ozonesondes, including balloon experiments using a multiple-instrument gondola (e.g. Hilsenrath et al., 1986; Deshler et al., 2008), dual flights (De Backer et al., 1998; Stübi et al., 2008) and environmental chamber simulations (Smit et al., 2007). A quantitative assessment of ozonesonde data quality is currently under way, following guide lines prepared by the ozonesonde data quality assessment panel (part of the SPARC-IO$_3$C/IGACO-O$_3$/UV-NDACC Assessment of "Past changes in the vertical distribution of ozone").

Comparison with continuous records from other instruments, for example, space-borne, ground-based or other aircraft-borne in-situ measurements, can also provide information about potential long-term changes in the performance of ozonesondes. The quality of tropospheric data from earlier European BM sondes has been questioned by Schnadt Poberaj et al. (2009) and recently also by Logan et al. (2012). Ozonesonde data quality is most critical in the UTLS, where O_3 concentrations are particularly low and where changes in O_3 distributions have the largest climate forcing (e.g. Forster and Shine, 1997; Forster and Tourpali, 2001).

Commercial aircraft have also been used to provide high quality UTLS O_3 measurements, for example, as part of the MOZAIC aircraft program (Measurements of ozone, water vapor, carbon monoxide and nitrogen oxides by in-service Airbus aircraft, Marenco et al., 1998), European long-range airliners were equipped with accurate UV photometers to measure O_3 and other traces gases. These data are available from August 1994. In a companion paper (Staufer et al., 2013) we use MOZAIC O_3 measurements to analyze ozonesonde data from Payerne (Switzerland). Using three-dimensional trajectories to find commonly sampled air masses, we found discrepancies of up to 20 % between 1994–1997. However, after 1998 the deviations of the mean values between the two types of instruments decreased to < 5 %.

AMTD

6, 7099–7148, 2013

In this paper, the analysis of Staufer et al. (2013) is extended to various other soundings sites in Europe, America, Japan and Africa to provide an insight of the longterm performance of UTLS ozonesonde measurements around the globe.

2 Data and methodology

2.1 Ozonesondes

The meteorological observatory at Hohenpeißenberg (MOHp), Germany, is the only ozonesonde station that continues to use BM sondes (manufactured by Mast Keystone Corporation, Reno, NV, USA), whereas Uccle and Payerne switched to ECC sondes in April 1997 and September 2002, respectively. KC sondes have only ever been flown at Japanese sites. ECC sondes are manufactured either by Science Pump Corporation (SP; model type 5A and 6A) or, since the early nineties, by the Environmental Science Corporation (ES; model type Z). In 2011 ES was taken over by Droplet Measurement Technologies. Originally, ES sondes were operated with a 1.0 % fully buffered KI cathode sensing solution, but after the environmental chamber tests of JOSIE (Smit et al., 2007), the manufacturer recommended diluting the solution by half, to 0.5 % KI. Unfortunately, this somehow led to some confusion in the community and only some observation sites changed the solution strength (see Table 1). Additionally, National Oceanic and Atmospheric Administration (NOAA) sites (in this study Boulder and Huntsville) experimented with different solution strengths and buffers (2.0 % KI unbuffered solution Johnson et al., 2002, and recently with a 1.0 % KI 1/10th buffered solution). In this study we analyze 11 stations (Alert, Churchill, Edmonton, Eureka, Goose Bay, Lerwick, Natal, Observatory Haute Provence (OHP), Resolute, Scoresbysund and Sodankylä) that switched from ES to SP (or vice versa) and/or operated both sonde types with or without changing the solution strength. These sites include Canadian stations from which both SP and ES-Z sondes were flown before 2004. After 2004 mainly ES sondes were launched but keeping the 1.0 % KI solution strength.

AMTD
6, 7099–7148, 2013

The standard operating procedures (SOP) for the BM sondes are defined by Claude et al. (1987) and have been followed by MOHp and Payerne. Payerne has higher correction or scaling factors (CF) than MOHp, probably because the pump temperature was assumed constant at 280 K instead of 300 K. The CF is determined as the ratio between the total O_3 column measured by the ozonesonde and a nearby independent column measurement, such as from Dobson or Brewer photospectrometers. Uccle data, as used here, are normalized following De Backer (1999) rather than utilizing the SOPs. The SOP for the BM sondes does not call for correction of the background current.

For ECC sondes the conventional correction is to assume that the background current is proportional to the oxygen partial pressure and thus declines with altitude (Komhyr, 1986). This, however, is neither supported by lab studies (e.g. Thornton and Niazy, 1982), nor by the the study of Reid et al. (1996), who found better agreement between ECC ozonesondes and an UV photometer for tropospheric O_3 concentrations when a constant background current was assumed. The recent assessment of ECC SOPs calls for a constant background current (Smit et al., 2011). The background current (ib) is measured three times during the pre-launch procedure: once the sondes are exposed to purified (ozone-free) air (ib_1), once after exposure to O_3 (ib_2), and just prior to flight (ib_3).

Ozonesonde data can be downloaded from several archives: ftp servers at the WOUDC (World Ozone and Ultraviolet Radiation Data Center), NDACC (Network for the Detection of Atmospheric Composition Change), SHADOZ (Southern Hemisphere ADditional OZonesondes), NILU (Norwegian Institute for Air Research) and NOAA (National Oceanic and Atmospheric Administration). Records from most stations can be found either on the WOUDC or NDACC homepages, or on both. Most tropical stations are now part of the SHADOZ network (Thompson et al., 2003a,b). NILU offers campaign data, for example, measurements from the VINTERSOL campaign, and high latitude station data.

AMTD
6, 7099–7148, 2013

For some stations we needed to switch between the archives to obtain the highest number of soundings. We found that the archives do not necessarily contain the same number of soundings for the same period. Some years are missing at one archive but available at another. Some years are also missing in all archives. Data from Boulder, DeBilt, Izaña, MOHp, OHP, Sodankylä were obtained from the NDACC archive in March and April 2010. Sodankylä data from 2004 onwards were obtained from NILU's NADIR database in May 2010. Data from the Canadian sites (Alert, Churchill, Edmonton, Eureka, Goose Bay, Resolute), as well as for Huntsville, Legionowo, Lerwick, Lindenberg, Madrid, Naha, Nairobi, Payerne, Uccle, Sapporo, and Tsukuba were also obtained from the WOUDC in March and April 2010. Uccle data from 2007 onwards were obtained in February 2011. Huntsville data for 2008 and 2009 were obtained from a NOAA ftp-server in February 2011. Irene, Natal and Paramaribo data were obtained from the SHADOZ database in April 2010, with the exception of data from the Natal site for 1997, which was obtained from the WOUDC. Wallops Island data for 1994, 1995, 2008, and 2009 were obtained from the WOUDC in March and April 2010, while all remaining data were obtained from the NDACC website in April 2010.

MOHp, Payerne and Uccle typically launch two to three ozonesondes per week, whereas most other sites typically launch one sonde per week. It is important to note, that not all sites flew ozonesondes for the entire MOZAIC period, with some stations starting later, particularly tropical ones. Consequently, the total number of launches for the entire MOZAIC period (August 1994–March 2009) is quite different from station to station, ranging from 300–400 (e.g. Paramaribo, Irene) to more than 2000 launches (e.g. MOHp, Payerne, Uccle) (cf. Table 1).

2.2 MOZAIC ozone observations

The MOZAIC program and data from it are described and analyzed in detail by Thouret et al. (1998). Here, only the main characteristics are summarized. Dual-beam UV absorption models from Thermo Environment were installed on several commercial aircraft participating in the MOZAIC project. These UV photometers have a response time

AMTD
6, 7099–7148, 2013

of 4 s, a detection limit of 2 ppbv, and an uncertainty of ±[2 ppbv + 2 %]. For example, for O_3 = 100 ppbv this results in an uncertainty of ±4 ppbv. The quality assurance and control procedures have not changed since the project started in 1994. MOZAIC analyzers are periodically calibrated (about every 12 months) with a reference analyzer at the French National Institute of Standards and Technology. Additionally, the analyzers are checked in-flight with a built-in ozone generator to detect any drift in instrument efficiency.

MOZAIC's main flight route is the North Atlantic flight corridor, but aircraft also fly to airports in South America, East Asia and Southern Africa. The flight distribution of the aircraft is shown in Fig. 1. The sounding sites investigated in this work are chosen according to these flight routes. In total, 31 494 flights were available when we downloaded the data (March 2010), covering the period from August 1994 to March 2009. We use 1 min averaged MOZAIC data, which correspond to a horizontal resolution of 10 to 15 km at cruise altitude.

2.3 Comparison methodology

For the comparison between routinely flown ozonesondes and ozone measurements from MOZAIC aircraft we use trajectories to ensure both instrument platforms observe the same air mass. In a companion paper (Staufer et al., 2013), we test and apply this method to comparisons between aircraft measurements from both MOZAIC and NOXAR (Nitrogen Oxides and Ozone along Air Routes project) aircraft and ozonesonde data from Payerne, Switzerland. Here, we summarize just the main points of this method, which is similar to the "trajectory match technique" used by Rex et al. (1998) or the "trajectory hunting technique" described by Danilin et al. (2002). After reconstructing the sonde's flight path using wind data (speed and direction) from the radiosonde, the trajectory tool LAGRANTO (Wernli and Davies, 1997) is used to calculate 6 day forward and backward trajectories for each sounding. LAGRANTO is forced with six-hourly wind fields from ECMWF's ERA-Interim reanalysis (1° horizontal resolution, 61 vertical levels). Because of chemical processing, O_3 cannot be assumed constant along each trajectory, but Staufer et al. (2013) show that the simultaneous use of both forward and backward matches alleviates this problem. Trajectories ascending or descending by more than 450 hPa during the six days simulated are excluded to avoid air masses that transport polluted boundary layer air or air from deep stratospheric intrusions. For each trajectory all MOZAIC measurements matching the trajectory within $r \leq 75$ km and $\Delta\Theta \leq 0.6$ K are collected, then a weighted mean of the aircraft observations is calculated and compared to the ozonesonde measurements at initialization of the trajectories. For the weighting a time lag compared to the soundings is used to account for the reduced accuracy of trajectories further away in time. To assess the uncertainty of this technique Staufer et al. (2013) checked the method for comparison of one instrument type with itself, i.e. MOZAIC–MOZAIC self-matches, and found mean differences of $\pm \lesssim 2$ %.

As shown by Staufer et al. (2013), the combination of forward and backward trajectories can be used to account for the potential effects of chemistry and mixing along the trajectory paths. However, for the data considered, very few trajectories were matched in both directions and Staufer et al. (2013) needed to analyze forward and backward trajectories separately. They found sonde biases of up to 10 % for the forward-only and backward-only trajectories, but showed that by combining forward and backward trajectories and by surrounding each trajectory with four additional trajectories, each displaced by 0.5° latitude and longitude from the central trajectory, the biases could be reduced by half. Furthermore, the sonde bias at Payerne was found to be largely insensitive to the trajectory duration (one or six day). Due to this robustness, and because some sites do not allow reconstructing the balloon flight path since no wind direction or speed data are available, we use all trajectories (the central plus the displaced trajectories), referred to as the combined trajectory set, unless otherwise mentioned.

For tropical stations ($\phi < 30°$ N), trajectories were determined every 1 K in potential temperature at altitudes between 5–15 km. This is in contrast to the mid- and high latitude stations, where similar to Staufer et al. (2013), the trajectories originate every 5 hPa within the UTLS, which is defined as ±125 hPa around the local (lapse-rate de-

AMTD
6, 7099–7148, 2013

fined) tropopause. Without this adaptation, no matches with MOZAIC would have been obtained because the tropical tropopause is much higher than the typical MOZAIC cruise altitude (8–12 km).

3 Ozonesonde comparisons with MOZAIC

Results are first presented as averages over the entire MOZAIC period, thereafter the differences between sonde and MOZAIC, $\Delta O_3 = 2(\text{Sonde} - \text{MOZAIC})/(\text{Sonde} + \text{MOZAIC})$, are analyzed in more detail by separately discussing the behavior and changes of ΔO_3 in both the upper troposphere (UT) and lower stratosphere (LS). We first focus on the mid-latitudes ($30^\circ\,\text{N} \leq \phi \leq 60^\circ\,\text{N}$) where most stations are located, then show results for stations at high northern latitudes ($\phi > 60^\circ$ N) and for tropical and southern latitudes ($\phi < 30^\circ$ N).

Results of the comparisons are limited by the number of matches, which in turn depends on the number of ascents, the location of the station and the MOZAIC flight paths (see Fig. 1, and Tables 1 and 2). The number of matches per launch site varies considerably. Stations that are either closer to highly frequented MOZAIC airports or whose balloon trajectories cross the main MOZAIC flight path over the North Atlantic are favored. Previous results for Payerne show that the temporal distribution of matches (and the appropriate weighting) is an important factor for comparison (Staufer et al., 2013).

3.1 Results for mid-latitude stations

3.1.1 MOHp/Payerne/Uccle

During the MOZAIC period (1994–2009) only three stations flew BM ozonesondes. Uccle and Payerne switched to ECC sondes (ES-Z type operated with 0.5 % KI half buffered sensing solution) in April 1997 and September 2002, respectively, Hohenpeißenberg (MOHp) still launches BM sondes, and thus is the only remaining BM station worldwide. Sample sizes from these three stations are largest because 2–3 sondes are flown per week at each of these sites, and the stations are located close to the main MOZAIC airports and flight routes. The percentage of matched ozonesondes is largest at Uccle (≈ 90 %) and smallest at MOHp (71 %). This disparity is related partly to the location of Uccle, which lies close to Brussels airport and the main flight route of aircraft over North and Western Europe (see Fig. 1). In addition, MOHp data from the WOUDC are reported on fewer pressure levels than at Payerne and Uccle.

The 16 yr mean O_3 concentrations from sondes and MOZAIC are shown in Fig. 2a. They show a striking agreement. The height of the lapse-rate defined tropopause is derived only from the sondes used in this comparison, resulting in a mean tropopause pressure of 250 hPa. Sonde–MOZAIC differences obtained from the unidirectional trajectories are negligibly small at all altitudes for both MOHP and Uccle, but range up to 5 % at Payerne (see also Staufer et al., 2013), although the differences in absolute concentrations are on the order of a few ppb in the troposphere. For all stations, the O_3 concentrations obtained from forward-only trajectories are systematically higher than from backward-only trajectories. This difference is possibly because of the chemistry along the trajectory path and is further discussed in Staufer et al. (2013).

Figure 2b shows the mean relative differences between sonde and MOZAIC split into three different periods. The first period, 1994–1997, is characterized by large differences between BM sondes and early MOZAIC observations. At MOHp the sondes exceed MOZAIC by 10–15 %, while at Payerne and Uccle even higher offsets are found (up to 25 % in the vicinity of the tropopause). Mean 1994–1997 differences are lowest (5 % at MOHp and 10 % at Payerne, Uccle) at the 175 hPa level, where mean O_3 concentrations are on the order of 200 ppb. After 1997/1998 the mean differences drop to less than 10 % at all three stations at all altitudes.

Figure 2c–e shows the time series of 13 month central moving average monthly mean differences. The lower stratosphere (Fig. 2c) includes only trajectories where the difference in pressure between the trajectory at initialization ($p(t = 0)$) and the tropopause pressure (p_{TP}) is smaller than 15 hPa. Figure 2d contains a narrow

AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.

tropopause band with $|p(t=0) - p_{TP}| < 15$ hPa. The values chosen are similar to those of Thouret et al. (2006), who also considered a 30 hPa thick tropopause zone. The time series of the tropopause differences is more uncertain and more variable because the strongest O_3 gradients are typically found in the vicinity of the tropopause. Figure 2e comprises the upper troposphere and includes all trajectories satisfying $p(t=0) - p_{TP} \geq 15$ hPa. All calculations follow the methodology laid out in Staufer et al. (2013), however 50 hPa pressure intervals are used here instead of 1 km altitude bins.

At MOHp, the CF corrects for the low BM sonde bias in the LS, except for the 1994–1997 period when the application of the CF results in a high bias compared to MOZAIC. For the UT, application of the CF is counterproductive for almost all periods. In contrast, at Payerne the agreement with MOZAIC in both the UT and LS is better when no scaling is applied. Whereas Stübi et al. (2008) recommended scaling both sonde types to column O_3, Staufer et al. (2013) suggest that the transition from BM to ECC sondes is smoother when the BM sondes remain unscaled, at least for the LS as defined here. The homogenized Uccle and MOZAIC data show differences of less than 5 % in the LS (Fig. 2c), but the homogenization does not remove the high offset in the mid-1990s in the troposphere (Fig. 2e). However, their CF, whose calculation differs from the usual approach, reduces the bias to MOZAIC to 5 % in the UT after 1996. Our analysis qualitatively confirms results from Schnadt Poberaj et al. (2009) for the European ozonesonde stations. Our analysis furthermore shows that the mean discrepancies at Uccle from 1994–2001 can be traced back to the use of BM sondes. Our results for the free troposphere ($p > 430$ hPa) also qualitatively agree with the recent study of Logan et al. (2012), who found that the tropospheric portion of BM sonde data before 1998 should be discarded for trend analysis due to the mismatch with MOZAIC. The anomalous peak in the Uccle tropospheric data in 2007 is present in our analysis, although in 2002 a peak of similar magnitude is also found. However, note that the peak in 2002 is not present in the UT, when trajectories satisfying $p(t=0) - p_{TP} \geq 30$ hPa are used instead (not shown). The high bias in UT O_3 compared to MOZAIC observed at all three BM sonde sites for the 1994–1997 period remains unexplained.

AMTD
6, 7099–7148, 2013

3.1.2 DeBilt

In contrast to Payerne and Uccle, at DeBilt SP sondes operated with a 1.0 % KI cathode electrolyte have been used during the entire MOZAIC period. A smaller number of ozone soundings is available from this launch site compared to MOHp, Payerne and Uccle because typically just one sounding is launched per week. Despite this, 90 % of the soundings could be matched with MOZAIC (Table 1). Similar to MOHP, Payerne and Uccle, the 16 yr mean O_3 concentrations from both DeBilt and MOZAIC agree to within 10 % at all altitudes (Fig. 3a). However, the difference between ozonesondes and MOZAIC shows a distinct time dependence: ΔO_3 amounts to 15 % in the LS and 20 % in the UT from 1995–1996, then slowly decreases to below 0 % by the end of the 1999, and then increases again to up to 10 % after 2003 (Fig. 3c and d). This is likely related to the background signal, the main source of uncertainty in calculating tropospheric O_3 partial pressures, which shows a significant trend over this period (Fig. 3e). Prior to 2003 the background current was high (0.10–0.16 µA) and highly variable. The reduced background current after 2003 is a consequence of a change in pre-launch procedure. Chemicals are renewed more often and the signal is measured outdoors just prior to launch instead of being measured indoors (Ankie Piters, personal communication, 2012). With typical values of < 0.06 µA, the background current at DeBilt now agrees well with the signals measured at Payerne and Uccle (0.03–0.04 µA). The drop of ΔO_3 to below 0 % from 1998–2002 can be explained by two factors, the large background current values and the change in background current treatment. Since November 1998 a constant background current has been used to process the data instead of a background current that declines with altitude. In the case of having large background current values, when a constant value is subtracted from the measured cell current much lower O_3 partial pressures are obtained than when an altitude dependent background current is subtracted. This feature is more pronounced in the upper troposphere than in the lower stratosphere since smaller O_3 partial pressures are measured. The hypothesis that the background current values and the changing

AMTD
6, 7099–7148, 2013

background current treatment are responsible for the low O_3 partial pressures measured by sondes is further supported by the fact that the differences between sonde and MOZAIC are more stable (i.e. ΔO_3 do not drop to below 0 %) before 2003 if the data are processed with an altitude dependent background current, especially in the LS (Fig. 3c). After 2003, when the mean background values drop below 0.06 µA, the spread in the results resulting from different correction schemes (altitude dependent or constant) is significantly smaller. During this period, the agreement with MOZAIC in the LS is better than 5 % when a constant background is subtracted, and better than 10 % when an altitude-declining background current is subtracted. In the UT, the agreement is better than 10 % with a constant background current and better than 15 % with an altitude-declining background current.

3.1.3 Legionowo

At Legionowo, Poland, SP sondes (1.0 % KI) were also flown for the entire period considered here. In terms of data treatment, but an altitude- declining background current with altitude is applied. The number of ozonesondes launched is similar to that at DeBilt, and 89 % of the launched sondes can be matched with MOZAIC. Mean differences between the Legionowo sondes and MOZAIC are substantial in the troposphere (10–15 %), but smaller in the stratosphere (< 10 %) (see Fig. A1a and b). The differences remain relatively constant in time, similar to the background current values (Fig. A2). The only exception is 1995, when the sondes exceed MOZAIC in the troposphere by up to 20 % (Fig. A1e), as observed at Payerne, Uccle, MOHp, and DeBilt. In the LS, differences between sonde and MOZAIC are large from 1994–1997 (15–20 %), then decrease to below 10 % from 1998–2001, and remain around 10 % thereafter. The LS time series of the ΔO_3 at both Legionowo and DeBilt show similar patterns if both data are processed assuming an altitude-declining background signal.

AMTD
6, 7099–7148, 2013

3.1.4 Lindenberg/Madrid/OHP

The comparison between ozonesonde data from Lindenberg, Madrid and the Observatory Haute Provence (OHP) with MOZAIC observations is presented in Fig. A3. The Lindenberg and Madrid sites followed the ECC flight instructions of Komhyr (1986) and Komhyr et al. (1995) for SP sondes (1.0 % KI unbuffered cathode solution, processed assuming an altitude-declining background current signal). At OHP they changed from using the SP sondes (flown with 1.0 % KI) to using ES sondes (1.0 % KI) in March 1997, and the data are post-processed assuming a background current that is constant with altitude. Between 400–500 ozonesondes are matched at these three sites, substantially less compared to DeBilt and Legionowo (700–800), or MOHp, Uccle, Payerne (Table 1). About 80 % of the sondes flown from Madrid and OHP can be matched with MOZAIC, while at Lindenberg less than 70 % are matched, partly because fewer trajectories are initialized. Similar to MOHp, the other DWD (German Weather Service) station, data from Lindenberg are reported on fewer pressure levels than at Madrid, OHP and several other sites. The tropopause height at Lindenberg calculated using the soundings available for comparison is 20–30 hPa higher than several stations, including Madrid, OHP, and Legionowo (Fig. A3a).

Between 300–400 hPa there are some sonde–MOZAIC differences (up to 10 %) between the backward- and forward-only trajectories, with the backward-only trajectories yield larger sonde biases (Fig. A3a). As mentioned previously, and as described in detail by Staufer et al. (2013), this may be due to chemical processing along the trajectories.

The lower stratospheric sonde–MOZAIC differences at Lindenberg, Madrid and OHP range from −5 % (OHP) to 5–10 % (Lindenberg, Madrid), while in the troposphere they are somewhat larger (Fig. A3b). Large discrepancies between sondes and MOZAIC are found in the stratosphere at both Lindenberg and Madrid from 1994–1996 (up to 15 %) (Fig. A3c). In the troposphere the discrepancies increase in the 1990s at Madrid, while at Lindenberg the sonde data are 15–30 % larger than MOZAIC from 1994–1998.

AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.

Our analysis shows that there is no obvious break in the ΔO_3 time series over OHP resulting from the switch of ECC sonde manufacturer in March 1997. There is, however, a decrease in tropospheric bias after 1997, although this cannot be attributed to the change in ECC sonde type since several other stations show similar deviations during this period as well. The times series in the LS is too noisy to draw any firm conclusions.

3.1.5 Churchill/Edmonton/Goose Bay

The sonde–MOZAIC comparison at the Canadian mid-latitude stations Churchill, Edmonton and Goose Bay is presented in Fig. A4. Because of the MOZAIC flight distribution, most matches are obtained from forward-trajectories (Fig. A4a), with most trajectories originating from the lower stratosphere. In total 350–500 ozonesondes can be matched with MOZAIC at these stations, the equivalent of only one third the sample size of the European BM stations.

In the lower stratosphere, the ΔO_3 time series are qualitatively similar, especially for Edmonton and Goose Bay. The sondes overestimate MOZAIC by up to 5 % at Goose Bay, and by up to 15–35 % at Edmonton and Churchill from 1994–1996, but then underestimate O_3 compared to MOZAIC from 1997–1999 (Fig. A4c). Thereafter, the sonde–MOZAIC bias becomes positive again, ranging between 5–15 %, depending on the station. The results suggest no statistically significant differences in the mean lower stratospheric deviations (at the 90 % confidence level; see Fig. A4b). Although there are few tropospheric matches, the discrepancies at Goose Bay from 1995–1996 (ranging between 15–20 %) are similar to those observed at European stations (Fig. A4e).

3.1.6 Boulder/Huntsville/Wallops Island

Matches for the United States stations are obtained mostly from forward trajectories originating in the upper troposphere, particularly at Huntsville and Wallops Island (see Fig. A5a). Note, that as for the Canadian stations, the number of matched ozone soundings is much lower than at MOHp, Payerne or Uccle. At Boulder, the sensing solution

AMTD
6, 7099–7148, 2013

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◂◂ | ▸▸
◂ | ▸
Back | Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

strength changed twice, in August 1997 and November 2005 (Table 1), but no obvious change in the sonde–MOZAIC agreement is found in the UTLS (Fig. A5b). This is likely in part due to the low measurement frequency and the position of these stations. The NOAA team (responsible for the Boulder and Huntsville stations) also found only very small tropospheric differences when using different sensing solutions (B. J. Johnson, personal communication, 2012). At Boulder, the sondes exceed MOZAIC observations by < 5 % above 300 hPa for most of the time period considered, but show a positive offset of up to 20 % in the lower stratosphere from 1995–1996 (Fig. A5c). This provides further evidence that MOZAIC perhaps underestimates O_3 compared to the sondes from 1994–1997.

Ozone soundings began at Huntsville in 1999. From all observations considered here, only very few backward trajectories are matched so the O_3 time series is almost entirely determined from forward trajectories. Sondes exceed MOZAIC by up to 10 %, depending on altitude (Fig. A5b), with slightly higher values being observed after the change from a 2.0 % KI unbuffered solution to a 1.0 % KI 1/10th buffered solution in March 2006.

Above 350 hPa ozonesondes flown from Wallops Island show similar results from 1994–2009 with sonde measurements exceeding MOZAIC by 5–15 % (Fig. A5b). These values agree well with results from Schnadt Poberaj et al. (2009), who also show a positive sonde bias of 5–20 % compared to MOZAIC from 1994–2001. Below 350 hPa the sondes tend to measure more O_3 from 2005–2009 than in previous periods, in particular between 350–450 hPa. Such a trend is not visible at other sites.

3.1.7 Sapporo/Tsukuba

The 16 yr mean O_3 concentrations obtained from sondes flown at Tsukuba and Sapporo, and from MOZAIC show encouraging results in the troposphere (Fig. 4a). However, there are differences in the stratospheric performance (typically above 250 hPa) from forward-only and backward-only trajectories at Tsukuba. The agreement of sondes with MOZAIC also tends to evolve differently at Sapporo and Tsukuba (Fig. 4b).

AMTD
6, 7099–7148, 2013

Although both stations used the same sonde type (KC-79 until summer 1997, KC-96 from summer 1997 to December 2009), from 2005–2009 ΔO_3 is positive (10–20 %) above 350 hPa at Tsukuba but negative (–10 %) at Sapporo.

As already mentioned, the time lag between sonde–MOZAIC matches can be an important factor for the comparison. The temporal distribution of the individual matches is provided in Fig. 5, however, this distribution might be biased since it does not account for the averaging of the matches along each trajectory – some trajectories contain more individual matches than others – nor for the weighting of matches along the trajectories. It does, though, provide an idea of the mean time lag between MOZAIC measurements and the soundings. In contrast to most of the European stations included here, the majority of stratospheric matches at non-European stations are not found within the first 50 h of air parcel travel time. Rather, many matches occur at the end of the trajectories where they have already traveled more than 100 h. In our companion paper (Staufer et al., 2013), a 2 % uncertainty was found when testing this matching technique using MOZAIC–MOZAIC self-matches. This comparison found that most matches (50 %) occurred within the first two days (48 h) of trajectories. Thus, in the case of the Japanese stations with almost no matches in the first two days of the trajectories, this uncertainty is likely to be higher than 2 % due to accumulated trajectory errors and may explains the discrepancies observed. The results for Sapporo and Tsukuba therefore should be treated with caution.

3.2 High-latitude stations

3.2.1 Lerwick/Scoresbysund/Sodankylä

Results for the high latitude stations included in this study are presented in Fig. 6. At these sites the tropopause is located above 300 hPa except for at Lerwick, where it is located above 250 hPa. A larger sample is obtained for the LS compared to the UT because the height of the tropopause is lower at high-latitudes than at mid-latitudes while MOZAIC's cruise altitude remains constant (8–12 km) independent of latitude.

AMTD
6, 7099–7148, 2013

For all three high latitude stations the sondes exceed MOZAIC by 5–10 % in the stratosphere and by 10–15 % in the troposphere. For Lerwick and Sodankylä the differences between sondes and MOZAIC obtained from backward-only trajectories are systematically larger than for forward-only trajectories (5 %) (Fig. 6a). No statistically significant change in the sonde performance is observed at these three launch sites, although Sodankylä and Scoresbysund reported changes in the type of sondes used (Fig. 6b). At Sodankylä both SP and ES sondes were flown with an 1.0 % KI cathode electrolyte before February 2006. The majority of the sondes during this period were SP sondes, although for short periods, ranging from several weeks to 3 months, ES sondes were used. From February 2006 onwards only ES sondes with a 0.5 % KI solution have been flown. During both periods the sondes were operated following recommendations of the scientific community and manufacturers, and therefore only small differences of a few percent are to be expected (e.g. Smit et al., 2007; Deshler et al., 2008). Note that since February 2006 data are processed with a constant background current instead of with an altitude-dependent background current. In contrast to DeBilt, the trend in background current values measured as part of pre-flight preparations is small, and therefore this change is not likely to influence the sonde performance at Sodankylä.

At Scoresbysund, after the change from SP to ES sondes, measured O_3 concentrations are systematically higher at all altitudes (Fig. 6b). This is in accordance with the JOSIE experiments (Smit et al., 2007), which showed that ES sondes had systematic high bias compared to SP sondes when both are operated with a 1.0 % KI cathode sensing solution.

At Lerwick sondes exceed MOZAIC, in particular in the upper troposphere in the early years (> 20 % from 1994–1996, see Fig. 6c). ΔO_3 in the LS is < 10 % for most of 1994–2009, similar to most ECC stations. Lerwick frequently changed between SP and ES sondes, but it seems this does not influence the agreement with MOZAIC data.

AMTD
6, 7099–7148, 2013

3.2.2 Alert/Eureka/Resolute

Only one third of all ozone soundings from these sites are available for comparison with MOZAIC (see Table 1). Tropospheric data from these stations are particularly scarce. Because of the very small sample size, only the 16 yr 1994–2009 average is provided (see Fig. A6). Most sonde–MOZAIC matches are found at altitudes between 200–300 hPa, and indicate that the sondes exceed MOZAIC measurements by 5–10 %.

3.3 Tropical stations

3.3.1 Izaña

Results for the troposphere using only forward trajectories reveal a sonde–MOZAIC bias similar to that observed at European sites such as Legionowo or Madrid. In contrast, results using only backward trajectories reveal a large offset between sondes and MOZAIC (> 20 %, see Fig. 7a), which is 2–3 times larger than what was shown in the JOSIE experiments for SP sondes operated with a 1.0 % KI cathode solution (Smit et al., 2007). The geographical distribution of matches (Fig. 8) shows two major peaks in the backward direction, one over the Canary Islands and one over the East Coast of the United States, while most matches in the forward direction are found over the Mediterranean sea, with no pronounced peaks. The spatial distribution is also reflected in the temporal distribution of matches, since no pronounced peaks are observed (Fig. 5). Most matches with MOZAIC observations therefore occur after 3 days travel along the trajectory paths, where the trajectories are expected to be less accurate. It may also be possible that O_3 production takes place over the course of the 12 day trajectories given that the photochemical lifetime of tropospheric O_3 is expected to be shorter in the subtropics and tropics than in the mid- and high latitudes (e.g. Logan et al., 1981).

Differences between the two data sets are less than 5 % in the stratosphere, and no trend in performance is found (Fig. 7b). No statistically significant changes in bias are

AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Stauffer et al.

found in the troposphere either, but again note that the the sonde data have a greater positive bias from 1994–1995 compared to the subsequent four to five years (Fig. 7e).

3.3.2 Nairobi/Irene/Naha/Paramaribo/Natal

As a result of the distribution of MOZAIC flights, only very few matches with ozonesondes in the tropics and Southern Hemisphere were found (less than one third of all sondes flown sondes). Furthermore, comparison was only possible in the troposphere because the aircraft cruise altitude is usually below the height of the tropopause. Because of the very small sample size, only the 16 yr (1994–2009) average results are provided (see Fig. A7). In addition, the rather poor temporal distribution of matches adds to the uncertainty (Fig. 5). Most stations agree with MOZAIC to within 10 % below 400 hPa and within 20 % (10–20 ppbv) above 400 hPa. The bias found at Paramaribo is considerably larger than the other stations, ranging between 30–40 % higher than MOZAIC below 300 hPa.

4 Summary and conclusions

In this study 16 yr (1994–2009) of O_3 observations from MOZAIC aircraft were compared with measurements from balloon-borne ozonesondes using 6-day, three-dimensional trajectories. Match criteria of 75 km maximum horizontal distance and 0.6 K maximum potential temperature difference ($\approx \pm 20$ m) were chosen to ensure that measurements from both platforms sampled the same air mass. This methods relies on 14 859 balloon ascents that are matched with observations from MOZAIC flights, yielding a total of 129 340 independent match trajectories. Results are encouraging for mid- and high latitude stations after 1998, where ozonesondes typically differ from MOZAIC by less than 5 %, in agreement with previous studies (for example, the JOSIE and BESOS experiments; Smit et al., 2007; Deshler et al., 2008). Our results also confirm that, at least during the MOZAIC period, ozonesondes provide a reliable tool for investigating atmospheric O_3 climatologies, even in proximity to the tropopause, where O_3 partial pressures are low and measurement uncertainty high. The differences between ozonesondes and MOZAIC are typically smaller in the lower stratosphere than in the upper troposphere, where the uncertainty of ozonesondes is higher.

From 1994–1997 ozonesonde data quality is an issue, particularly in the UTLS, where O_3 partial pressures are lowest. These results also agree well with previous studies (Logan et al., 2012; Schnadt Poberaj et al., 2009). The BM sondes flown operationally at MOHp, Payerne and Uccle during this period overestimate O_3 by up to 25 % in the upper troposphere compared to MOZAIC. Due to the more favorable signal-to-noise ratio, measurements in the lower stratosphere show a smaller offset during this period, especially at Uccle. After 1998, the sonde–MOZAIC deviations decrease in both the UT and LS to values below 10 %. By 1998 most stations had switched from using BM to ECC sondes, with, for example, Uccle having switched in 1997 and Payerne in 2002. In comparison, MOHp continues to operate BM sondes. From 2000 onwards, the sondes flown at these three stations agree with MOZAIC to better than 5 %. Interestingly, larger discrepancies between sonde and MOZAIC measurements in the upper troposphere in the mid to late 1990s were also found at DeBilt, Legionowo, Lindenberg, Goose Bay, Lerwick, Sodankylä and Izaña, with the discrepancies decreasing to between 5–15 % from 1998 onwards (see Fig. 9). All these stations used ECC sondes, mainly SP sondes flown with a 1.0 % KI fully buffered cathode sensing solution strength. In the lower stratosphere, at many ECC-only stations, an increasing agreement with MOZAIC was found between the 1990s and 2000s, possibly pointing to an improved sonde performance over time. Typically ECC sondes exceed MOZAIC by 10–20 % before 1998 but from 1998–2002 they agree to within 5–10 %, or, as found for DeBilt, Goose Bay, Edmonton, the measured O_3 concentrations are even lower than those measured by MOZAIC. Since we have no evidence for changes in preparation procedures or data processing at these stations, the change in agreement over time may indicate some dependence on the manufacturing batch. Differences in the characteristics of the same ECC sonde types, even when operated under the same conditions, were also identified by Smit et al. (2007) during different JOSIE campaigns. At DeBilt we found evidence for the effect of large background current signals: this was partly attributable to the large variability in background current signals, combined with changing from applying an altitude-dependent background current to using a constant background current.

The only launch station at which we find a systematic increase in the sonde O_3 measurements resulting from a change from SP to ES sondes is Scoresbysund (and the solution strength being kept constant). These findings are in agreement with conclusions from the JOSIE 2000 experiments (Smit et al., 2007), which reported a higher bias for ES sondes compared to SP sondes when both are operated with a 1.0 % KI sensing solution.

For all other stations, in particular at Boulder, where the sensing solution was changed twice, our analysis reveals no obvious break in the ΔO_3 times series, meaning that either the uncertainty of the method applied in this study is too low to detect these changes, or that the differences introduced by these changes are negligibly small under UTLS conditions.

There is an ongoing debate on the application of a correction factor (CF) to normalize sonde profiles to a nearby column O_3 measurement (Dobson or Brewer), in particular concerning the application of a CF to the tropospheric fraction of the measurements (e.g. SPARC/IOC/GAW, 1998; Thouret et al., 1998; Stübi et al., 2008; Schnadt Poberaj et al., 2009). Since the CF largely depends on stratospheric O_3 levels, doubts have been raised with respect to its application to the tropospheric parts of profiles. The application of a CF implies making assumptions about the O_3 content above burst altitude, which can introduce biases originating from the independent column measurements used. We find no systematic behavior of sondes to the application of a CF, but rather annual differences, for example, at MOHp, soundings in the lower stratosphere show better agreement with MOZAIC before 1998 if not normalized, but after 1998 the normalization decreases the sonde–MOZAIC differences. In the troposphere, however, better agreement is obtained without the normalization over the

AMTD
6, 7099–7148, 2013

entire time period. The only two exceptions are at Uccle and Legionowo where upper tropospheric soundings agree better with MOZAIC when the data are normalized using a CF.

The method developed in the companion paper (Staufer et al., 2013) and applied here provides the most reliable results if the ozonesonde launch site used lies within or near the main aircraft corridors. For the tropical and Japanese stations we find few matches with MOZAIC, and only an indication of sonde performance from 10 yr averages was obtained. At these sites, certain inconsistencies between the sondes and MOZAIC may be attributable to the inadequate temporal distribution of the matches, also in combination with the very low O_3 partial pressures found in the UTLS. Overall, results are more uncertain and less consistent when the majority of the aircraft measurements match with trajectories after they have traveled more than three days, which is the case, for example, for the Japanese and most of the tropical stations.

The above considerations have been used to argue that the ozonesondes should only be trusted after 1998. Furthermore, the UV photometry technology used by MOZAIC program is expected to be more precise, particularly at the low ozone concentrations typical of the UTLS. The MOZAIC instruments are also regularly calibrated so that errors can be detected after the flights, which is not the case for single-use balloon-borne ozonesondes. The fact, however, that 10 of the 20 time series shown in Fig. 9 indicate large positive differences compared to MOZAIC in the mid-1990s followed by a systematic tendency to smaller differences in subsequent years, casts doubts on the explanation that the differences are due solely to errors stemming from the ozonesondes. It is remarkable that various sonde types reveal similar behavior, namely BM (Brewer Mast) and ECC (electrochemical cells manufactured by either SP or ES). In view of the fact that three different manufacturers were involved in building these instruments, it is not straightforward to understand this behavior. Likewise, however, MOZAIC operated five aircraft instruments simultaneously, and it is also not clear how these instruments could explain the observed differences, even though they are identically constructed, maintained and calibrated. A comparison between BM sondes and O_3 measurements made during the NOXAR B747 project from 1995–1996 showed a smaller offset of around 15 % (scaled) compared to MOZAIC, which may indicate a small drift in the MOZAIC calibration (see Staufer et al., 2013). These results require further investigation, but it would appear premature to suggest that all pre-1998 discrepancies result solely from the ozonesonde measurements.

Acknowledgements. J. Staufer's research is funded by a MeteoSwiss GAW-CH grant. The authors would like to thank the European Commission, Airbus, CNRS-France, FZJ-Germany for their strong support of the MOZAIC project, as well as the airlines (Lufthansa, AirFrance, Austrian, Air Namibia, and former Sabena) involved in the project who have carried the MOZAIC instrumentation free of charge since 1994. MOZAIC is presently funded by INSU-CNRS (France), Météo-France, CNES, Université Paul Sabatier (Toulouse, France) and Research Center Jülich (FZJ, Jülich, Germany). IAGOS is additionally funded by the EU projects IAGOS-DS, IAGOS-ERI, and IGAS. MOZAIC-IAGOS data are available via the CNES/CNRS-INSU Ether web site http://www.pole-ether.fr. We gratefully acknowledge all staff at the soundings sites for carefully preparing and performing all launches. The ozone sonde data used in this publication were obtained from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC; data publicly available at http://www.woudc.org, the Network for the Detection of Atmospheric Composition Change (NDACC; data publicly available at http://www.ndsc.ncep.noaa.gov), SHADOZ (http://croc.gsfc.nasa.gov/shadoz/), a NOAA ftp server (ftp://ftp.cmdl.noaa.gov/ozwv/ozone/), and from NILU's NADIR database. Finally, we would also like to thank Bryan J. Johnson and Alberto Redondas for providing information regarding the sonde configuration used at the NOAA and Izaña sites, respectively.

References

Brewer, A. and Milford, J.: The Oxford Kew ozonesonde, P. Roy. Soc. Lond., 256, 470–495, doi:10.1098/rspa.1960.0120, 1960. 7101

Claude, H., Hartmannsgruber, R., and Köhler, U.: Measurement of Atmospheric Ozone Profiles Using the Brewer/Mast Sonde – Preparation, Procedure, Evaluation, WMO Global Ozone Research and Monitoring Project, World Meteorol. Organ., Geneva, Switzerland, 17, 1987. 7104

Danilin, M. Y., Ko, M. K. W., Froidevaux, L., Lyjak, L. V., Bevilacqua, R. M., Zawodny, J. M., Sasano, Y., Irie, H., Kondo, Y., Russell III, J. M., Scott, C. J., and Read, W. G.: Trajectory Hunting as an effective technique to validate multiplatform measurements: analysis of the MLS, HALOE, SAGE-II, ILAS and POAM-II data in October–November 1996, J. Geophys. Res., 107, 4420, doi:10.1029/2001JD002012, 2002. 7106

De Backer, H.: Homogenisation of ozone vertical profile measurements at Uccle, Wetenschappelijke en technische publicaties van het K. M. I., no. 7, 26, available at: ftp://ftp.kmi-irm.be/dist/meteo/hugo/publ/1999/o3prof.pdf (last access: 1 March 2010), 1999. 7104

De Backer, H., De Muer, D., and De Sadelaer, G.: Comparison of ozone profiles obtained with Brewer–Mast and Z-ECC sensors during simultaneous ascents, J. Geophys. Res., 103, 19641–19648, 1998. 7102

Deshler, T., Mercer, J. L., Smit, H. G. J., Stubi, R., Levrat, G., Johnson, B. J., Oltmans, S. J., Rigel, K., Thompson, A. M., Witte, J., Davies, J., Schmidlin, F. J, Brothers, G., and Sasaki, T.: Atmospheric comparison of electrochemical cell ozonesondes from different manufacturers and with different cathode solution strenghts: the Ballon Experiment on standards for ozonesondes, J. Geophys. Res., 113, D04307, doi:10.1029/2007JD008975, 2008. 7102, 7117, 7119

Forster, P. M. D. F. and Shine, K. P.: Radiative forcing and temperature trends from stratospheric ozone changes, J. Geophys. Res., 102, 10841–10855, 1997. 7102

Forster, P. M. D. F. and Tourpali, K.: Effect of tropopause height changes on the calculation of ozone trends and their radiative forcing, J. Geophys. Res., 106, 12241–12251, doi:10.1029/2000JD900813, 2001. 7102

Hilsenrath, E. W., Attmannspacher, W., Bass, A., Evans, W., Hagemeyer, R., Barnes, R. A., Komhyr, W., Mauersberger, K., Mentall, J., Proffitt, M., Robbins, D., Taylor, S., Torres, A., and Weinstock, E.: Results from the balloon ozone intercomparison campaign (BOIC), J. Geophys. Res., 91, 13137–13152, 1986. 7102

Johnson, B. J., Oltmans, S. J., Vömel, H., Smit, H. G. J., Deshler, T., and Kröger, C.: Electrochemical concentration cell (ECC) ozonesonde pump efficiency measurements and tests on the sensitivity to ozone of buffered and unbuffered ECC sensor cathode solutions, J. Geophys. Res., 107, 4393, doi:10.1029/2001JD000557, 2002. 7103

Kobayashi, J. and Toyama, Y.: On various methods of measuring the vertical distribution of atmospheric ozone (III) – carbon iodine type chemical ozonesonde, Pap. Meteorol. Geophys., 17, 113–126, 1966. 7101

Komhyr, W. D.: Electrochemical concentration cells for gas analysis, Ann. Geophys.-Italy, 25, 203–210, 1969. 7101
Komhyr, W. D.: Operations Handbook – Ozone Measurements to 40 km Altitude with 4A-ECC-Ozone Sondes, NOAA Techn. Memorandum ERL-ARL-149, Air Resources Laboratory, Boulder, CO, 1986. 7104, 7113
Komhyr, W. D., Barnes, R. A., Brothers, G. B., Lathrop, J. A., and Opperman, D. P.: Electrochemical concentration cell ozonesonde performance evaluation during STOIC 1989, J. Geophys. Res., 100, 9231–9244, 1995. 7113
Logan, J. A., Prather, M. J., Wofsy, S. C., and McElroy, M. B.: Tropospheric chemistry: a global perspective, J. Geophys. Res., 86, 7210–7254, 1981. 7118
Logan, J. A., Staehelin, J., Megretskaia, I. A., Cammas, J.-P., Thouret, V., Claude, H., De Backer, H., Steinbacher, M., Scheel, H.-E., Sübi, R., Frölich, M., and Derwent, R.: Changes in ozone over Europe: analysis of ozone measurements from sondes, regular aircraft (MOZAIC) and alpine surface sites, J. Geophys. Res., 117, D09301, doi:10.1029/2011JD016952, 2012. 7102, 7110, 7120, 7133
Marenco, A., Thouret, V., Nédélec, P., Helten, M., Kley, D., Karcher, F., Simon, P., Law, K., Pyle, J., Poschmann, G., Von Wrede, R., Hume, C., and Cook, T.: Measurement of ozone and water vapour by Airbus in-service aircraft: the MOZAIC airborne program, an overview, J. Geophys. Res., 103, 25631–25642, 1998. 7102
Reid, S. J., Vaughan, G., Marsh, A. R. W., and Smit, H. G. J.: Accuracy of ozonesondes measurements in the troposphere, J. Atmos. Chem., 25, 215–226, 1996. 7104
Rex, M., von der Gathen, P., Harris, N. R. P., Lucic, D., Knudsen, B. M., Braathen, G. O., Reid, S. J., De Backer, H., Claude, H., Fabian, R., Fast, H., Gil, M., Kyrö, E., Mikkelsen, I. S., Rummukainen, M., Smit, H. G., Stähelin, J., Varotsos, C., and Zaitcev, I.: In situ measurements of stratospheric ozone depletion rates in the Arctic winter 1991/1992: a Lagrangian approach, J. Geophys. Res., 103, 5843–5853, 1998. 7106
Schnadt Poberaj, C., Staehelin, J., Brunner, D., Thouret, V., De Backer, H., and Stübi, R.: Long-term changes in UT/LS ozone between the late 1970s and the 1990s deduced from the GASP and MOZAIC aircraft programs and from ozonesondes, Atmos. Chem. Phys., 9, 5343–5369, doi:10.5194/acp-9-5343-2009, 2009. 7102, 7110, 7115, 7120, 7121
Smit, H. G. J., Straeter, W., Johnson, B. J., Oltmans, S. J., Davies, J., Tarasick, D. W., Hoegger, B., Stubi, R., Schmidlin, F. J., Northam, T., Thompson, A. M., Witte, J. C., Boyd, I., and Posny, F.: Assessment of the performance of ECC-ozonesondes under quasi-flight conditions

in the environmental simulation chamber: insights from the Juelich Ozone Sonde Intercomparison Experiment (JOSIE), J. Geophys. Res., 112, D19306, doi:10.1029/2006JD007308, 2007. 7101, 7102, 7103, 7117, 7118, 7119, 7121

Smit, H. G. J. et al.: Quality Assurance and Quality Control for Ozonesonde Measurements in GAW, WMO Global Atmos. Watch Rep., No. 201, World Meteorol. Organ., Geneva, Switzerland, 2011. 7101, 7104

SPARC/IOC/GAW: Assessment of Trends in the Vertical Distribution of Ozone, edited by: Harris, N., Hudson, R., and Phillips, C., SPARC Report No. 1, WMO Ozone Research and Monitoring Project, Report No. 43, World Meteorol. Organ., Geneva, Switzerland, 1998. 7121

Staufer, J., Staehelin, J., Stübi, R., Peter, T., Tummon, F., and Thouret, V.: Trajectory matching of ozonesondes and MOZAIC measurements in the UTLS – Part 1: Method description and application at Payerne, Switzerland, Atmos. Meas. Tech. Discuss., 6, 7063–7098, doi:10.5194/amtd-6-7063-2013, 2013. 7102, 7103, 7106, 7107, 7108, 7109, 7110, 7113, 7116, 7122, 7123

Stübi, R., Levrat, G., Hoegger, B., Viatte, P., Staehelin, J., and Schmidlin, F. J.: In-flight comparison of Brewer–Mast and electrochemical concentration cell ozonesondes, J. Geophys. Res., 113, D13302, doi:10.1029/2007JD009091, 2008. 7101, 7102, 7110, 7121

Thompson, A. M., Witte, J. C., McPeters, R. D., Oltmans, S. J., Schmidlin, F. J., Logan, J. A., Fujiwara, M., Kirchhoff, V. W. J. H., Posny, F., Coetzee, G. J. R., Hoegger, B., Kawakami, S., Ogawa, T., Johnson, B. J., Vömel, H., and Labow, G.: Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology 1. Comparison with Total Ozone Mapping Spectrometer (TOMS) and ground-based measurements, J. Geophys. Res., 108, 8238, doi:10.1029/2001JD000967, 2003a. 7104

Thompson, A. M., Witte, J. C., Oltmans, S. J., Schmidlin, F. J., Logan, J. A., Fujiwara, M., Kirchhoff, V. W. J. H., Posny, F., Coetzee, G. J. R., Hoegger, B., Kawakami, S., Ogawa, T., Fortuin, J. P. F., and Kelder, H. M.: Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology 2. Tropospheric variability and the zonal wave-one, J. Geophys. Res., 108, 8241, doi:10.1029/2002JD002241, 2003b. 7104

Thornton, D. C. and Niazy, N.: Sources of background current in the ECC ozonesonde: implications for total ozone measurements, J. Geophys. Res., 87, 8943–8950, 1982. 7104

Thouret, V., Marenco, A., Logan, J. A., Nédélec, P., and Grouhel, C.: Comparisons of ozone measurements from the MOZAIC airborne program and the ozone sounding network at eight locations, J. Geophys. Res., 103, 25695–25720, 1998. 7105, 7121

Thouret, V., Cammas, J.-P., Sauvage, B., Athier, G., Zbinden, R., Nédélec, P., Simon, P., and Karcher, F.: Tropopause referenced ozone climatology and inter-annual variability (1994–2003) from the MOZAIC programme, Atmos. Chem. Phys., 6, 1033–1051, doi:10.5194/acp-6-1033-2006, 2006. 7110

Wernli, H. and Davies, H. C.: A Lagrangian-based analysis of extratropical cyclones, I: The method and some applications, Q. J. Roy. Meteorol. Soc., 123, 467–489, 1997. 7106

AMTD

6, 7099–7148, 2013

Table 1. Overview of the sonde types used, data processing methods applied for the MOZAIC period (August 1994–March 2009), and soundings available for comparison (see text). Fields are left blank when information is missing or redundant. SST denotes the cathode sensing solution strength of the ECC sondes, whereby SST 1.0 denotes the fully buffered 1.0 % KI solution, SST 0.5 the half-buffered 0.5 % KI, SST 2.0 the unbuffered 2.0 % KI and SST 1.0b the 1.0 % KI, 1/10th buffered solution. MOHp denotes the Meteorological Observatory at Hohenpeißenberg, Germany, OHP the Observatory Haute Provence, France. W denotes the WOUDC archive, N the NDACC archive, S the SHADOZ archive.

Station	Archive	lat	lon	Sonde Type	SST	Background Signal	Switch Date	Scaled to Column	Number of ascents total	used
Alert	W	82.5	−62.3	SP, ES	1.0	declining	2004	Yes	464	141 (30 %)
				ES					269	100 (37 %)
					1.0				160	101 (63 %)
Boulder	N	39.9	−105.2	ES	2.0	constant	21 Aug 1997	No	413	251 (61 %)
					1.0b		30 Nov 2005		210	107 (51 %)
Churchill	W	58.7	−94.1	SP, ES	1.0	declining	2004	Yes	419	209 (50 %)
				ES					209	132 (63 %)
DeBilt	N	52.1	5.2	SP	1.0	declining	1 Nov 1998	No	247	223 (90 %)
						constant			540	483 (89 %)
Edmonton	W	53.5	−114.1	SP, ES	1.0	declining	2004	Yes	461	318 (69 %)
				ES					256	171 (67 %)
Eureka	W	79.9	−85.9	SP, ES	1.0	declining	2004	Yes	670	151 (23 %)
				ES					356	130 (37 %)
Goose Bay	W	53.3	−60.3	SP, ES	1.0	declining	2004	Yes	424	313 (74 %)
				ES					247	176 (71 %)
Huntsville	W, NOAA	34.7	−86.7	ES	2.0	constant	1 Mar 2006	No	317	201 (63 %)
					1.0b				183	98 (54 %)
Irene	S	−25.9	28.2	SP	1.0				231	32 (14 %)
Izaña	N	28.5	−16.3	SP	1.0	constant		No	840	533 (63 %)
Legionowo	W	52.4	21.0	SP	1.0	declining		No	884	783 (89 %)

AMTD
6, 7099–7148, 2013

Table 1. Continued.

Station	Archive	lat	lon	Sonde Type	SST	Background Signal	Switch Date	Scaled to Column	Number of ascents total	used
Lerwick	W	60.1	−1.2	SP, ES	1.0	declining			719	592 (82 %)
Lindenberg	W	52.2	14.1	SP	1.0	declining		Yes	748	500 (67 %)
Madrid	W	40.8	−3.6	SP	1.0	declining			538	413 (77 %)
MOHp	W	47.8	11.0	BM				Yes	1889	1335 (71 %)
Naha	W	26.2	127.7	KC		declining	13 Nov 2008	Yes	579	159(27 %)
				ES	0.5				12	5 (42 %)
Nairobi	W	−1.3	36.8	ES	1.0	declining			519	103 (20 %)
Natal	S, W	−5.4	−35.4	SP	0.5		1 Apr 1999		46	16 (35 %)
					1.0				389	44 (11 %)
OHP	N	43.9	5.7	SP	1.0	declining	1 Mar 1997	No	51	47 (92 %)
				ES					541	424 (78 %)
Paramaribo	S	5.8	−55.2	SP	1.0	declining			380	82 (22 %)
Payerne	W	46.7	6.6	BM			1 Sep 2002	Yes	1288	1065 (83 %)
				ES	0.5	constant		Yes	1009	834 (83 %)
Resolute	W	74.7	−94.9	SP, ES	1.0	declining	2004	Yes	304	83 (27 %)
				ES					205	82 (40 %)
Sapporo	W	43.1	141.3	KC		declining		Yes	647	415 (64 %)
Scoresbysund	N	70.5	−22.0	SP	1.0	constant	13 Jul 2001	No	397	284 (72 %)
				ES					341	243 (71 %)
Sodankylä	N, NILU	67.4	26.6	SP, ES	1.0	declining	25 Jan 2006	No	796	514 (65 %)
				ES	0.5	constant			266	115 (43 %)
Tsukuba	W	36.1	140.1	KC		declining		Yes	793	374 (47 %)
Uccle	W	50.8	4.4	BM			1 Apr 1997	Yes	385	346 (90 %)
				ES	0.5	constant		Yes	1775	1536 (87 %)
Wallops Island	N, W	37.9	−75.5	SP	1.0	constant			874	594 (68 %)

Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper |

AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Stauffer et al.

Table 2. Total number of matches, matched trajectories and ascents used for the different launch sites included in this study.

Station	Total number of		
	matches	matched trajectories	matched sonde ascents
Alert	2703	1051	241
Boulder	5986	1816	459
Churchill	5271	1994	341
DeBilt	41 520	13 277	706
Edmonton	9301	3269	489
Eureka	2935	1077	281
Goose Bay	10 942	3979	489
Huntsville	5250	1543	299
Irene	743	219	32
Izaña	7450	2261	533
Legionowo	22 384	8118	783
Lerwick	16 048	5958	592
Lindenberg	5718	2080	500
Madrid	10 204	3381	413
MOHp	17 576	5885	1335
Naha	2589	693	164
Nairobi	1783	397	103
Natal	739	142	60
OHP	11 060	3702	471
Paramaribo	792	204	82
Payerne	70 336	21 439	1899
Resolute	1599	627	165
Sapporo	8198	2890	415
Scoresbysund	13 004	4699	572
Sodankylä	9559	3530	629
Tsukuba	3909	1468	374
Uccle	96 746	30 007	1882
Wallops	10 854	3631	594
total	395 203	129 340	14 859

AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.

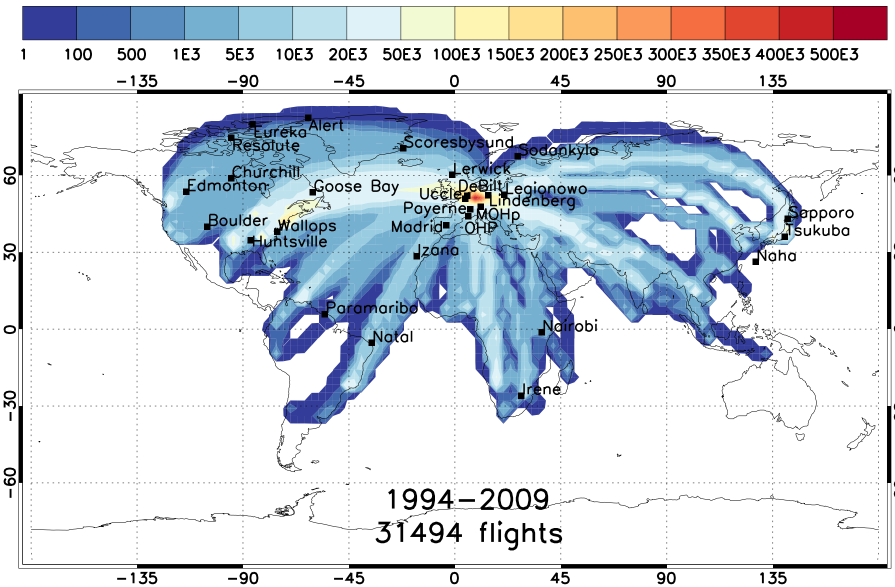
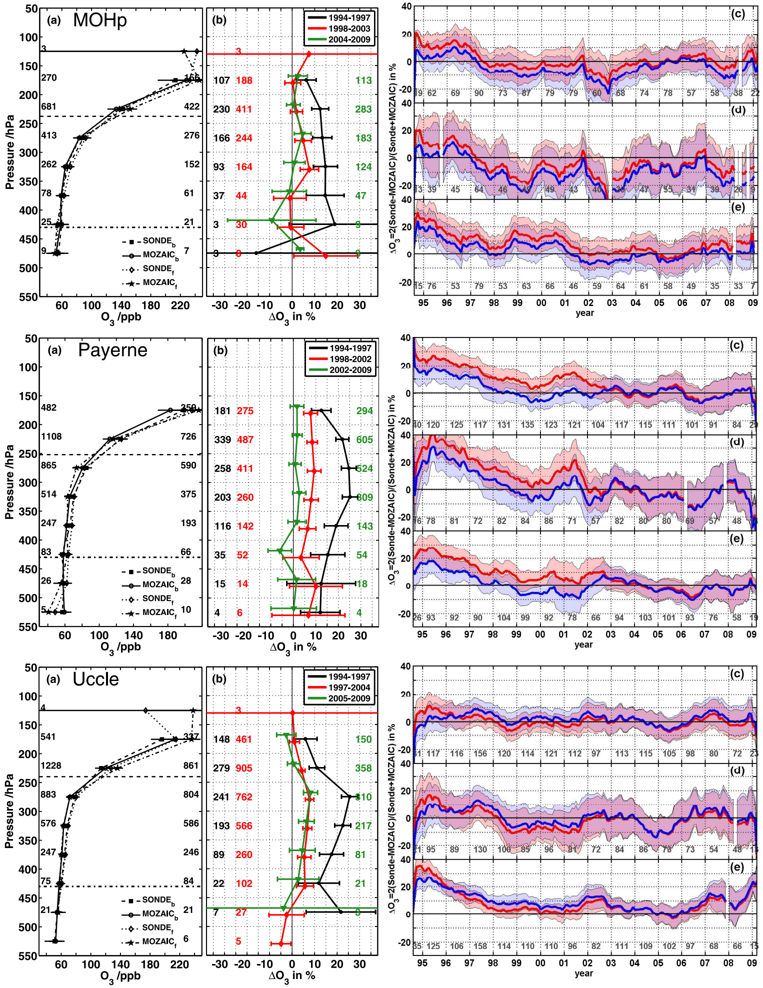


Fig. 1. Distribution of the ozonesonde stations considered in this work (black squares) and total number of measurements from the MOZAIC aircraft program (1994–2009), averaged over a 3° × 3° latitude × longitude grid. The color bar shows the total number of 1 min averaged MOZAIC measurements. MOHp denotes the Meteorological Observatory at Hohenpeißenberg, Germany, OHP the Observatory Haute Provence, France.

AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.



AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◀| |▶
◀ ▶
Back | Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

Fig. 2. Comparison between MOZAIC O_3 measurements and ozonesondes from MOHp, Payerne and Uccle. **(a)** 16 yr average O_3 profiles from sonde and MOZAIC binned into 50 hPa layers. Numbers on the left and right denote the number of soundings using 6 day backward-only and forward-only trajectories, respectively. The dashed horizontal line denotes the tropopause, the dash-dotted horizontal line the level up to which Logan et al. (2012) compared ozonesondes with MOZAIC. **(b)** Relative differences $\Delta O_3 = 2(\text{Sonde} - \text{MOZAIC})/(\text{Sonde} + \text{MOZAIC})$ split into three periods. The number of sondes available for comparison is displayed for each period on the sides. Time series of ΔO_3 with CF (red) and without CF (blue) are displayed for the LS **(c)**, a narrow tropopause band **(d)**, and the UT **(e)**. Numbers at the bottom indicate the number of soundings used for calculating monthly mean differences. The error bars in **(a)** and **(b)** denote the 90 % confidence of the median, while in **(c)**–**(e)** the shaded areas denote the standard error (68 % confidence). Overlapping areas are displayed in purple.

AMTD
6, 7099–7148, 2013

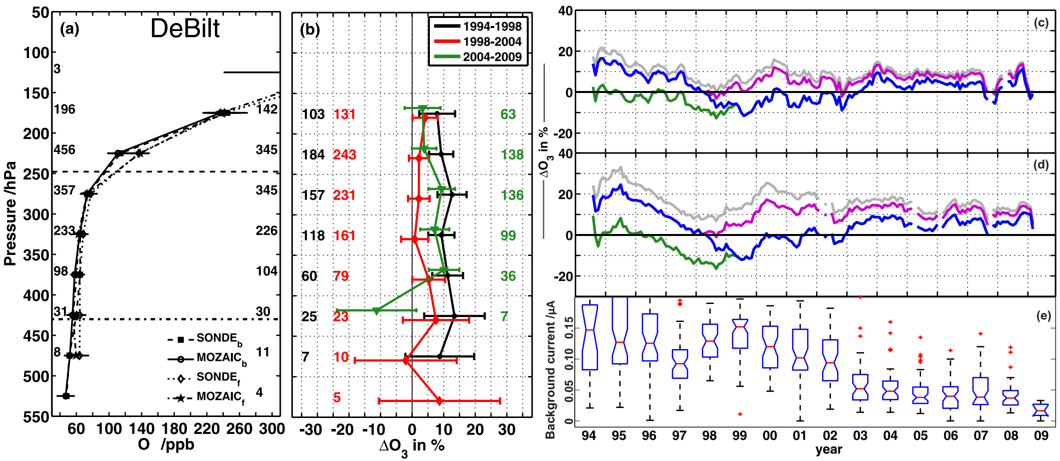


Fig. 3. Comparisons of DeBilt ozonesondes with MOZAIC measurements. **(a, b)** As for Fig. 2a and b. **(c, d)** Time series of ΔO_3 for the LS **(c)** and UT **(d)**, respectively. Time series are shown for the difference resulting from using the operating procedures described in Table 1 (blue line), with no correction for background signal (gray line), with a constant background current correction (green line), and with the application of an altitude-dependent background correction (purple line). **(e)** Annual statistical distribution of the background current used to process the ozonesonde data. The median is depicted as a horizontal solid red line, the 90 % confidence interval as the notches. The box limits correspond to the 75 and 25 % quartiles. The whiskers extend out to the maximum or minimum values, or to 1.5 times either the 75 or 25 % quartile if there are data beyond this range. Outliers are identified with red crosses.

AMTD
6, 7099–7148, 2013

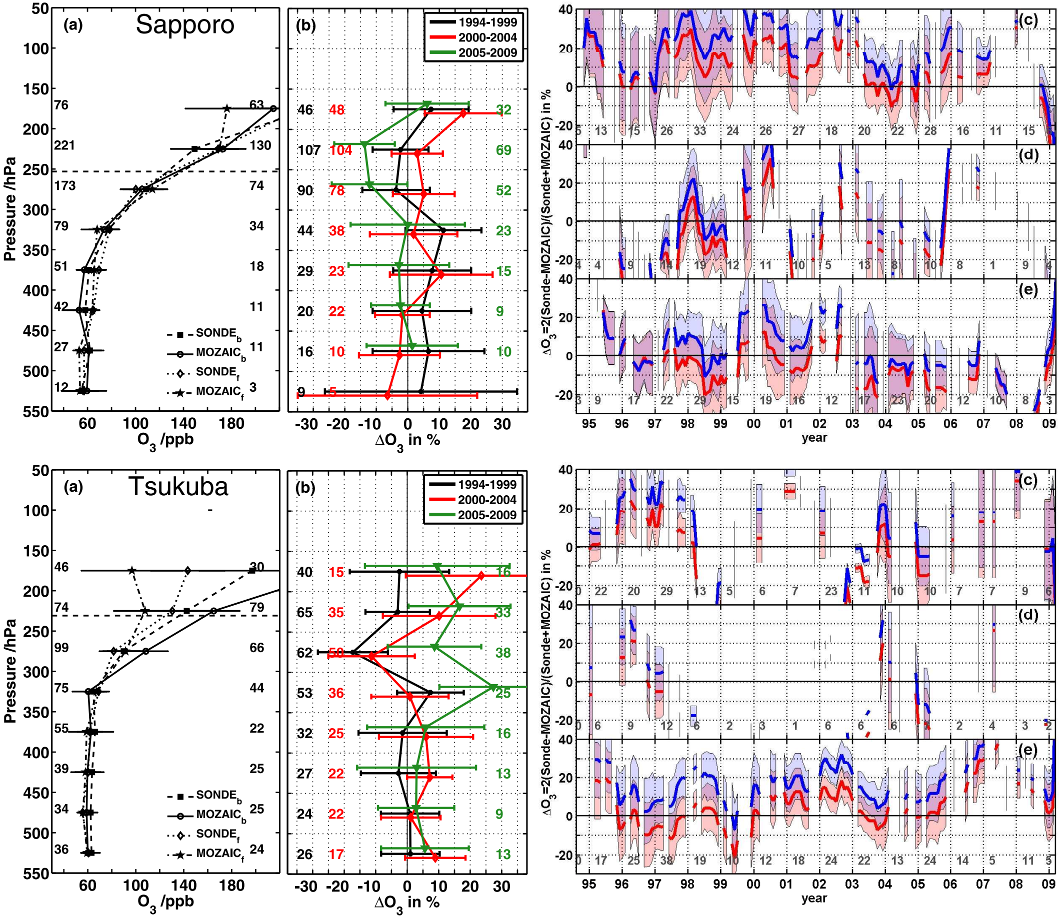


Fig. 4. Comparisons of ozonesondes and MOZAIC measurements. As for Fig. 2, but for Sapporo and Tsukuba.

AMTD

6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◀I | ▶I
◀ | ▶
Back | Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

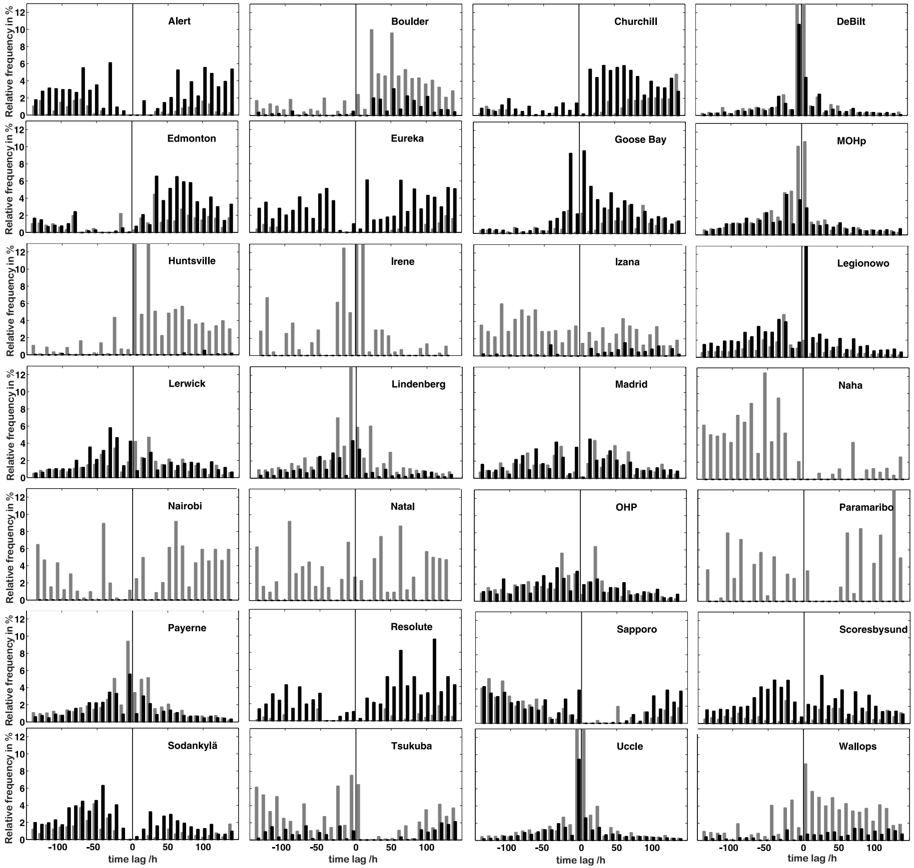


Fig. 5. Temporal distribution of the number of matches at different stations. The time lag is positive for forward-only trajectories and negative for backward-only trajectories. Matches in the stratosphere are shown in black, while matches in troposphere are shown in gray. The bin size is 10 h.

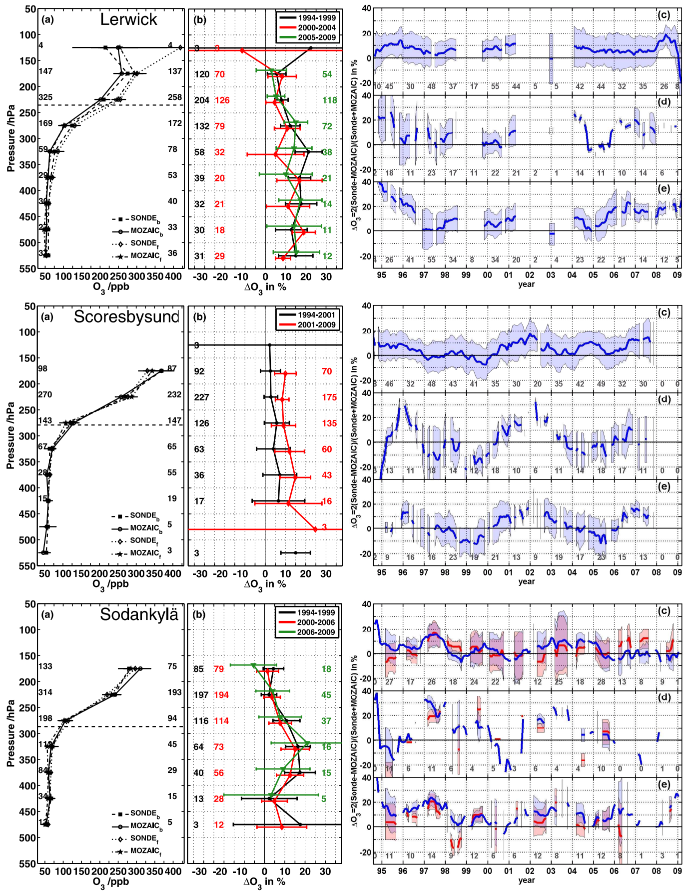


Fig. 6. Comparisons of ozonesondes and MOZAIC measurements. As for Fig. 2, but for Lerwick, Scoresbysund and Sodankylä.

AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.

Title Page
Abstract Introduction
Conclusions References
Tables Figures
◀ ▶
Back Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

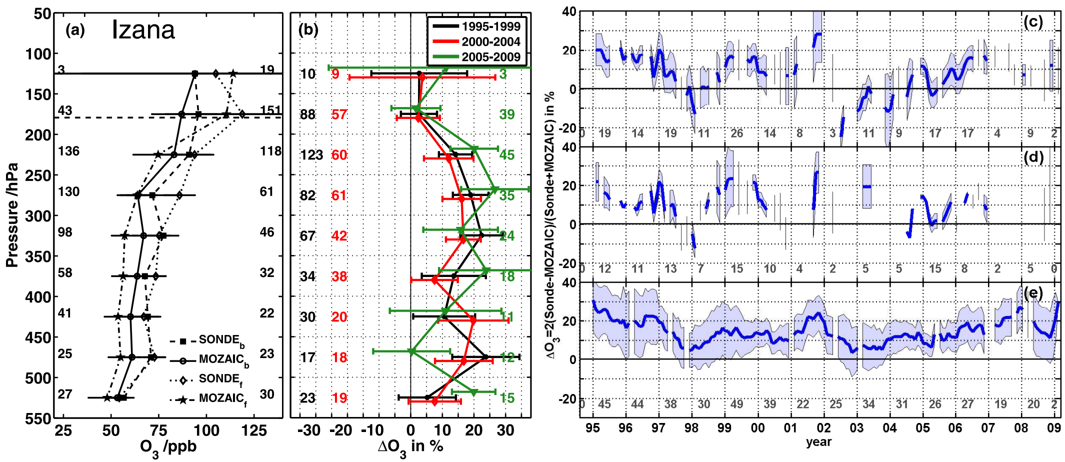


Fig. 7. Comparison of ozonesonde and MOZAIC ozone measurements. As for Fig. 2, but for Izaña.

AMTD
6, 7099–7148, 2013

J. Staufer et al.

Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper |

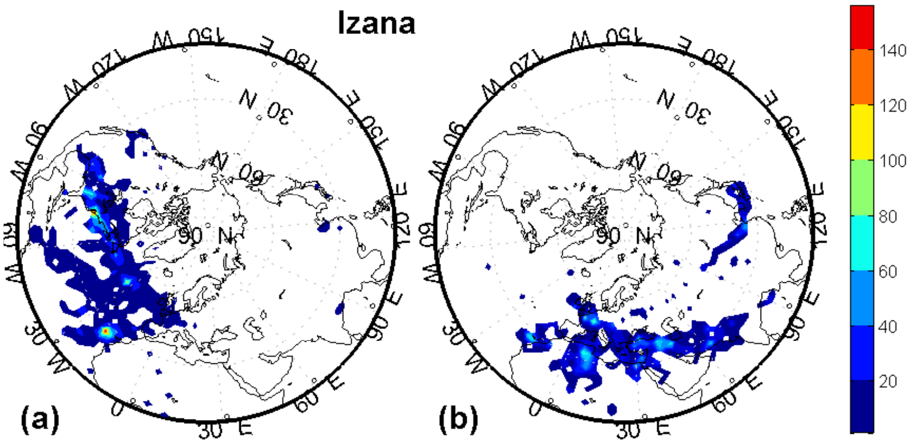
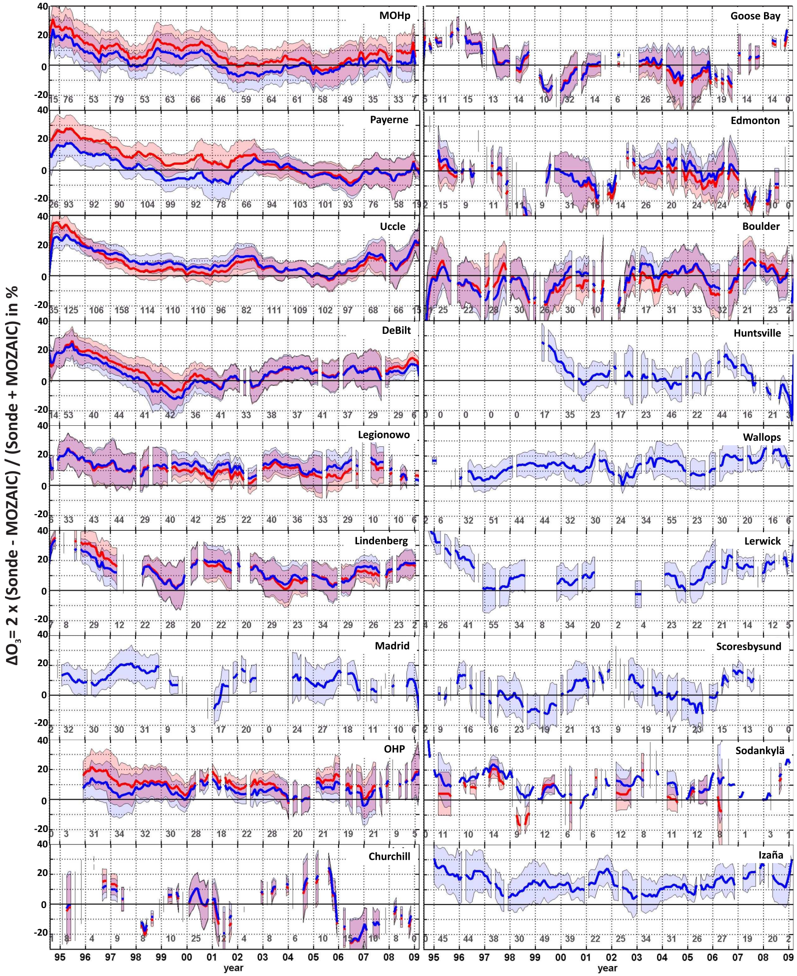


Fig. 8. Spatial distribution of matches between MOZAIC aircraft observations and **(a)** backward trajectories or **(b)** forward trajectories initialized at Izaña at altitudes between 5–15 km. The color bar shows the total number of matches, averaged over a 3° × 3° grid.

AMTD
6, 7099–7148, 2013



AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.

Fig. 9. Time series' of the relative differences between ozonesondes and MOZAIC measurements (ΔO_3 in %) in the upper troposphere. These time series comprise 20 of the 28 launch sites considered in this work (the 8 others have too few matches with MOZAIC to be included here). Note the large differences at nearly all sites from 1994–1998, and the systematic tendency for smaller differences at 10 of these stations thereafter, three using BM sondes (MOHp, Payerne, Uccle) and seven using SP or ES ECC sondes (DeBilt (SP), Legionowo (SP), Lindenberg (SP), Goose Bay (SP, ES), Edmonton (SP, ES), Lerwick (SP, ES), Izana (SP)). Bold lines: ΔO_3 time series with CF (red) and without CF (blue). Numbers at the bottom indicate the number of sondes used for calculating the monthly mean differences. The shaded areas denote the standard error (68 % confidence). Overlapping areas are displayed in light purple.

AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.

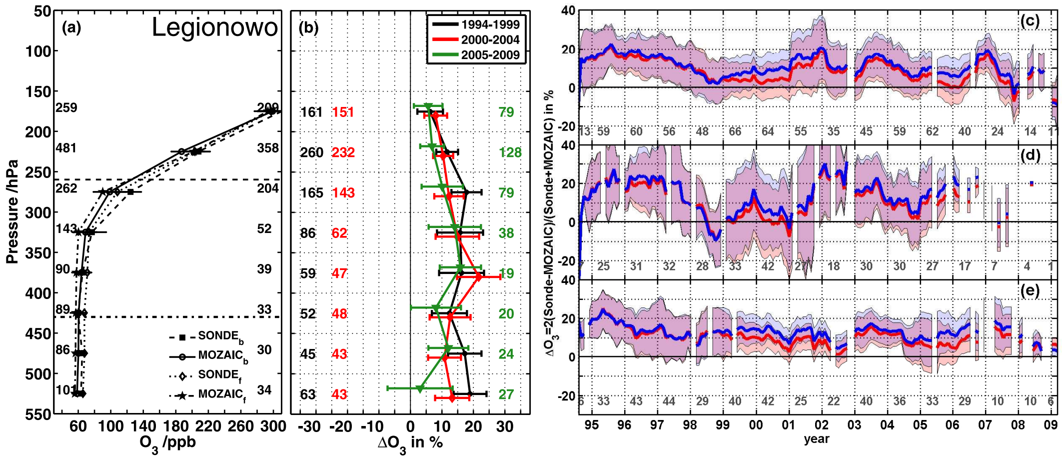


Fig. A1. Comparison of ozonesondes with MOZAIC measurements. As for Fig. 2, but for Legionowo.

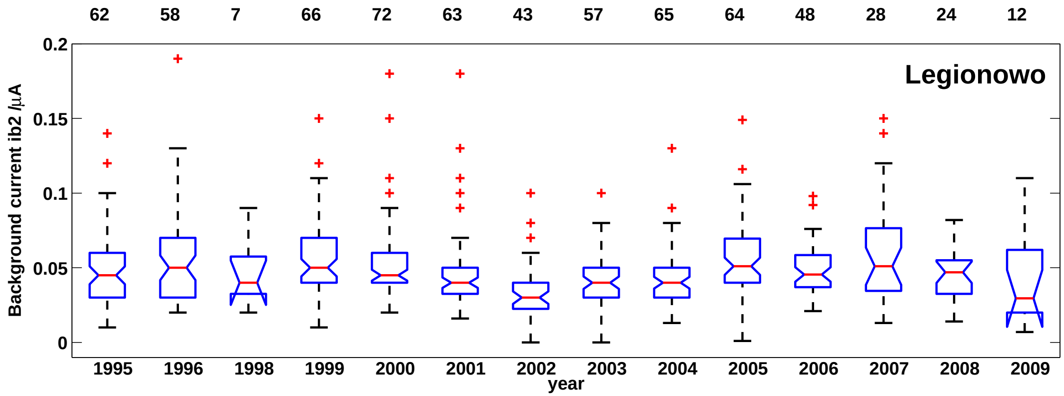


Fig. A2. Annual statistical distribution of the background current ib_2 used to process Legionowo ozonesonde data. Note that ib_2 is not reported to the archives for every launch and data prior to 26 January 1995 were not reported to the WOUDC archive.

AMTD
6, 7099–7148, 2013

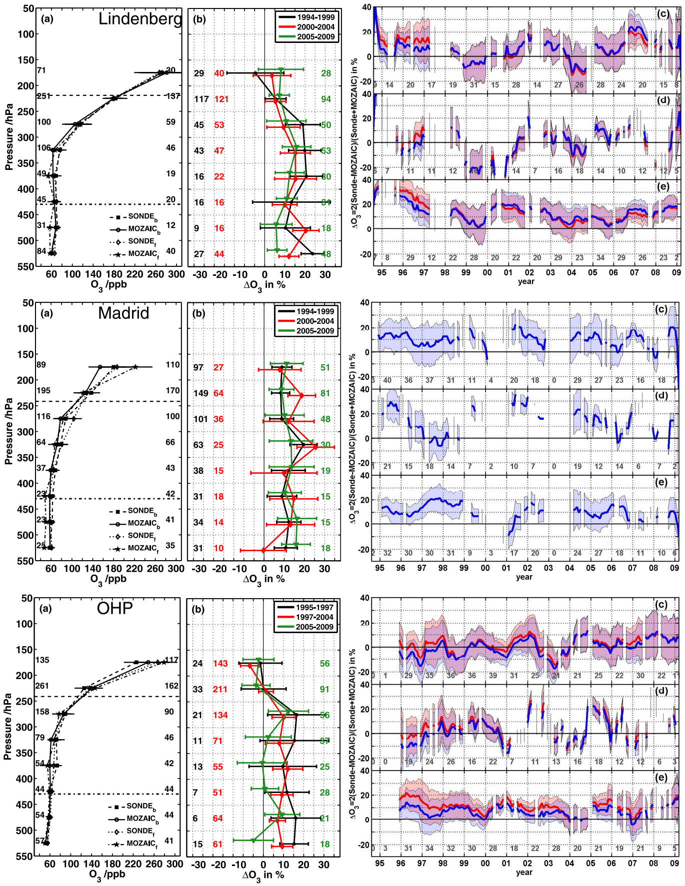


Fig. A3. Comparison of ozonesondes with MOZAIC measurements. As for Fig. 2, but for Lindenberg, Madrid and the Observatory Haute Provence (OHP).

AMTD

6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◀| |▶
◀ ▶
Back | Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

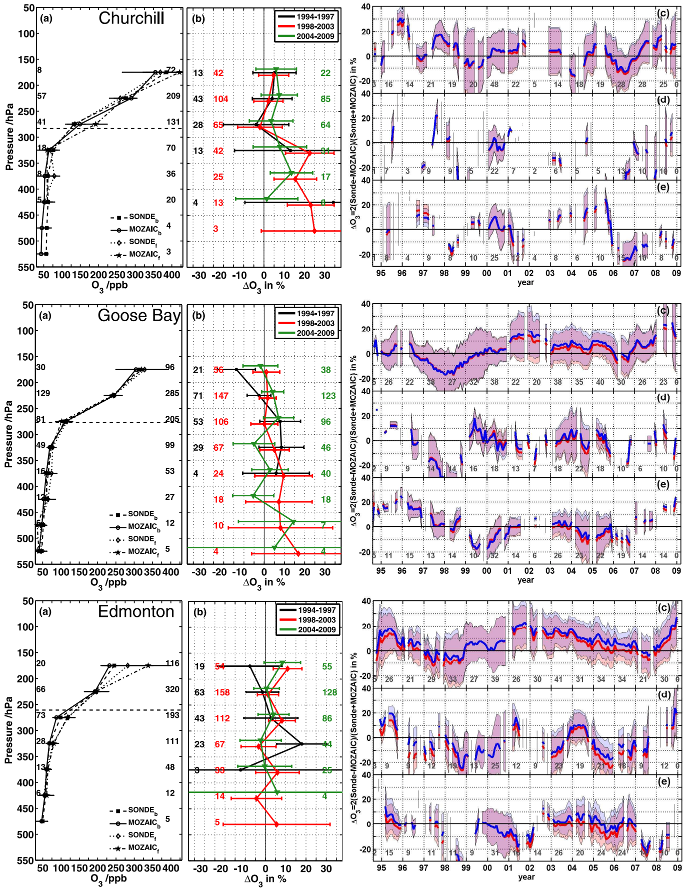


Fig. A4. Comparisons of ozonesondes with MOZAIC measurements. As for Fig. 2, but for Churchill, Edmonton and Goose Bay.

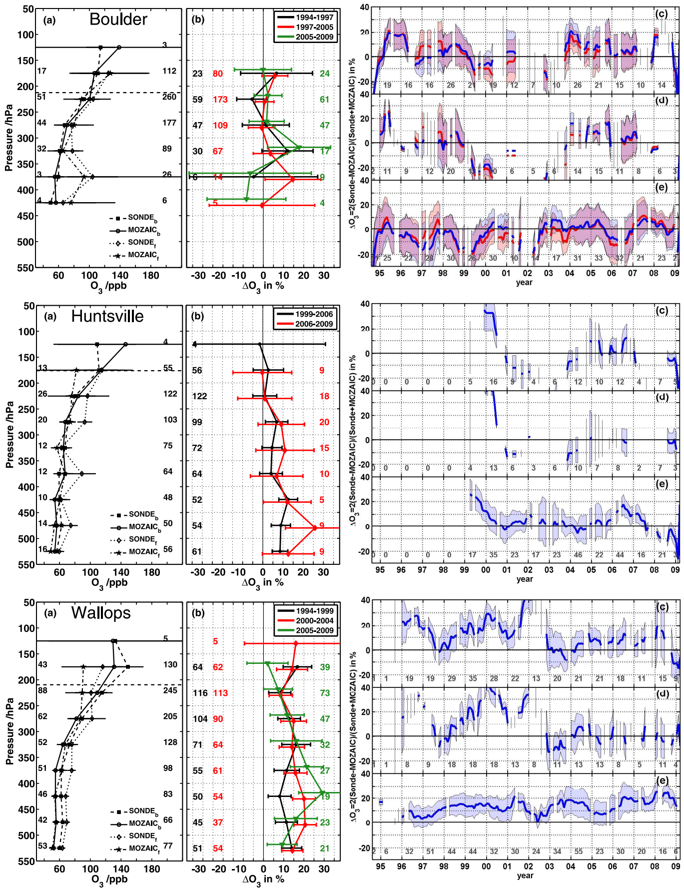


Fig. A5. Comparison of ozonesondes with MOZAIC measurements. As for Fig. 2, but for Boulder, Huntsville and Wallops Island. For each period in **(b)** a different sensing solution was used at Boulder and Huntsville.

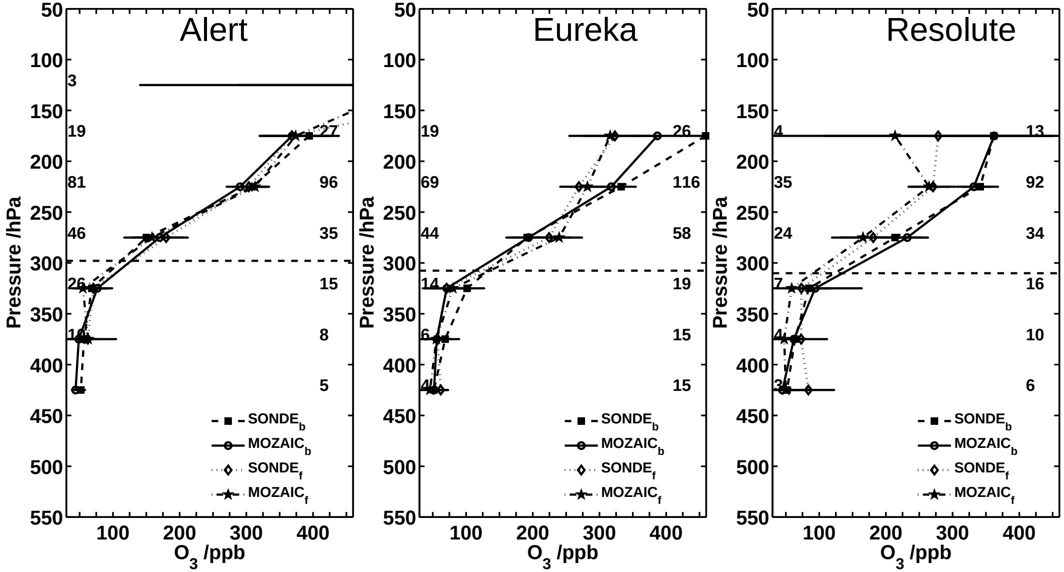


Fig. A6. 16 yr average (1994–2009) O_3 profiles from sondes flown at Alert, Eureka and Resolute, and from MOZAIC. Data are grouped into 50 hPa layers. Numbers on the left and right indicate the number of soundings using 6 day backward-only (subscript b) and forward-only trajectories (subscript f), respectively.

AMTD

6, 7099–7148, 2013

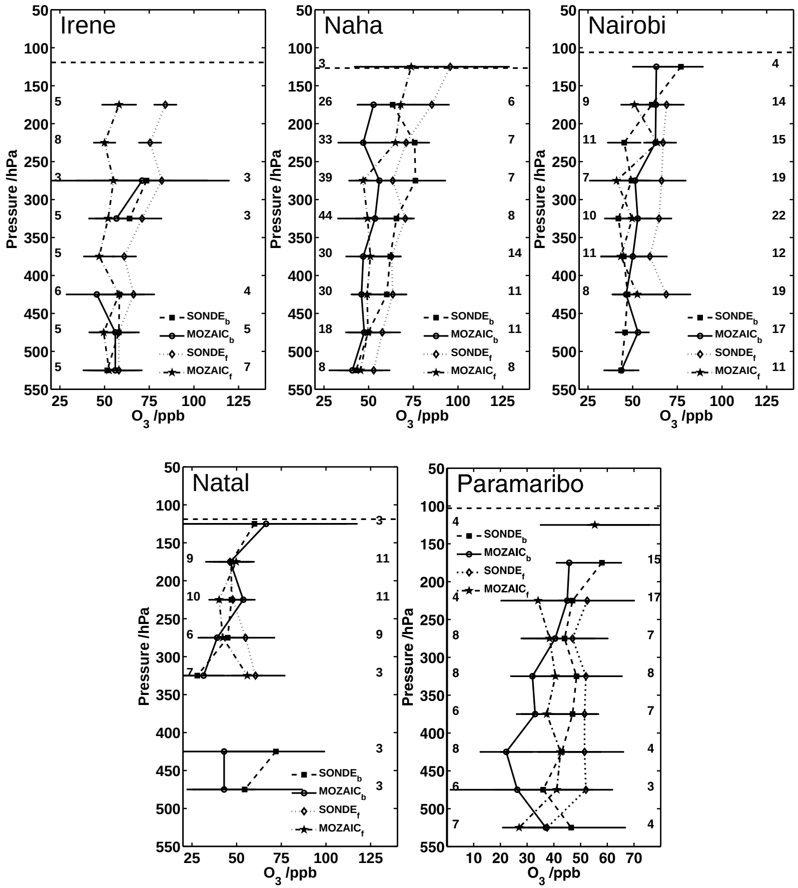


Fig. A7. Comparison of MOZAIC ozone measurements at tropical and southern latitude ozonesonde stations. As for Fig. A6, but for Irene, Naha, Nairobi, Natal and Paramaribo.

AMTD
6, 7099–7148, 2013

Trajectory matching of ozonesondes and MOZAIC – Part 2: Application

J. Staufer et al.