



## In-flight comparison of Brewer-Mast and electrochemical concentration cell ozonesondes

René Stübi,<sup>1</sup> Gilbert Levrat,<sup>1</sup> Bruno Hoegger,<sup>1</sup> Pierre Viatte,<sup>1</sup>  
Johannes Staehelin,<sup>2</sup> and F. J. Schmidlin<sup>3</sup>

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[1] The analysis of 140 dual flights between two types of ozonesondes, namely, the Brewer-Mast (BM) and the electrochemical concentration cell (ECC), is presented in this study. These dual flights were performed before the transition from BM to ECC as the operational ozonesonde for the Payerne Aerological Station, Switzerland. The different factors of the ozonesonde data processing are considered and their influences on the profile of the difference are evaluated. The analysis of the ozone difference between the BM and the ECC ozonesonde data shows good agreement between the two sonde types. The profile of the ozone difference is limited to  $\pm 5\%$  ( $\pm 0.3$  mPa) from the ground to 32 km. The analysis confirms the appropriateness of the standard BM data processing method and the usefulness of the normalization of the ozonesonde data. The conclusions of the extended dual flight campaigns are corroborated by the analysis of the time series of the Payerne soundings for the periods of 5 years before and after the change from BM to ECC which occurred in September 2002. No significant discontinuity can be identified in 2002 attributable to the change of sonde.

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### 1. Introduction

[2] Reliable ozone profile information and its change over time are crucial for describing the effect of man-made release of ozone depleting substances, such as chlorofluorocarbons which lead to stratospheric ozone depletion. In the troposphere, the profile information is also important because ozone is a strong greenhouse gas and its increase since preindustrial time significantly contributed to the increase of radiative forcing. The present concern about the future of the Earth's climate is generating extended climate modeling activities as well as new developments of instruments on board spacecraft. Both disciplines require reference systems for comparison, to verify the ability of the models to reproduce the state of the atmosphere and its evolution over time [e.g., *Stevenson et al.*, 2006] and to validate and maintain the calibration of the present and future remote sensing instruments in space [e.g., *Liu et al.*, 2006]. For this purpose, the ground based instruments need periodical review concerning their capabilities to fulfill their role as reference systems.

[3] Sondes have proved to be reliable instruments for in situ atmospheric ozone measurements. Attached to an aerological balloon, they record the ozone partial pressure

from the ground to more than 30 km. The sonde profiles are characterized by their high vertical resolution ( $\sim 150$  m) which permits measurements of the high ozone gradients present above the tropopause as well as the layered structures of the ozone profile which often occur in the winter-spring period [*Krizan and Lastovicka*, 2005]. However, the low ozone concentration below the tropopause is a challenging issue for in situ measurements. Careful preparation of the sondes is necessary to avoid contamination that may produce a bias in the profile.

[4] Ozonesonde data are extensively used for process studies [*Streibel et al.*, 2006], satellite validations [*Meijer et al.*, 2004] and ozone trend studies [*Logan*, 1985; *World Meteorological Organization (WMO)*, 1998; *Logan et al.*, 1999; *Staehelin et al.*, 2001; *Jeannot et al.*, 2007]. In these publications, questions about the methods used to prepare the sondes and to process the data are repeatedly raised.

[5] Two main designs for the ozonesondes have been used in the last 40 years: the BM type [*Brewer and Milford*, 1960; *Claude et al.*, 1987] and the ECC type [*Komhyr*, 1969]; ECC sondes dominate the global network. The coexistence of these instruments in different monitoring networks and during measurement campaigns requires a detailed comparison of their characteristics to avoid reducing the data quality by instrumental effects.

[6] In the early years, data recording on paper charts and simplified procedures in the preparation of the sondes were the major limitations to measurement precision and accuracy. The introduction of digital recording and standardized and computer controlled operations in the preflight preparation of the ozonesondes have reduced the uncertain-

<sup>1</sup>Payerne Aerological Station, MeteoSwiss, Payerne, Switzerland.

<sup>2</sup>Institute for Atmospheric and Climate Science, ETH Zürich, Zürich, Switzerland.

<sup>3</sup>Wallops Flight Facility, NASA Goddard Space Flight Center, Wallops Island, Virginia, USA.

ties of the measurements. The accuracy limitations appear to come now from manufacturing aspects of the ozonesondes (material used, specifications, sonde provider, etc.) as well as from details of the preparation procedures. The BM sondes are more sensitive to these factors than the ECC sondes and they show a larger variability from sonde to sonde in the simulation chamber [Smit and Kley, 1998] and in atmospheric conditions (see section 4.1).

[7] Over the last 40 years, studies comparing different ozonesonde types were conducted and can be grouped into four classes: (1) large balloon experiment with multiple instrument gondola to characterize the accuracy and precision of the sondes [Attmannspacher and Dütsch, 1970, 1981; Hilsenrath et al., 1986; Beekmann et al., 1994, 1995; Kerr et al., 1994; Komhyr et al., 1995a, 1995b; Margitan et al., 1995; Deshler et al., 2008]; (2) dual flight campaign [De Backer et al., 1998b; Boyd et al., 1998]; (3) comparison with other instrument types, generally ground based remote sensing instruments [Beekmann et al., 1994, 1995; Steinbrecht et al., 1998; Calisesi et al., 2003]; and (4) laboratory and environmental simulation chamber experiments [De Backer et al., 1998a; Smit and Kley, 1998; Steinbrecht et al., 1998].

[8] In the large balloon and laboratory experiments, the presence of a reference instrument (e.g., UV photometer [Proffitt and McLaughlin, 1983]) allows conclusions to be drawn about the accuracy and precision of the ozonesondes, whereas for the other experiments the relative differences between ozonesondes are only inferred. Over the 40 year period that has elapsed since these experiments occurred, ozonesondes have evolved which implies that some of the earlier conclusions could be out of date.

[9] A review of the experiments conducted up to 1998 is presented in the SPARC/IO3/GAW report published in 1998 [Harris et al., 1998, Table 2.9]. This report describes the most relevant problems and uncertainties regarding the ozonesonde measurements. Since this review, further progress has been made with the successive Jülich simulation chamber campaigns JOSIE in 1996, 1998 and 2000 [Smit and Sträter, 2004a, 2004b; Smit et al., 2007] and the recent large balloon experiment BESOS [Deshler et al., 2008].

[10] Among the long-term ozone sounding series available for trend analysis, several stations originally used the BM ozonesondes and later changed to ECC. However the transition from one system to the other generated significant breaks in the data series. Different studies have been published concerning the revision of old BM series to homogenize data sets or to explain the break induced by the transition from BM to ECC sondes. These include the series from Australia [Lehmann and Easson, 2003; Lehmann, 2005], Canada [Tarasick et al., 1995, 2002; Fioletov et al., 2006], Uccle (Belgium) [Lemoine and De Backer, 2001; De Backer et al., 1998a, 1998b] and the Observatoire de Haute-Provence (France) [Guirlet et al., 2000].

[11] The Payerne series were measured with BM sondes until August 2002 and ECC sondes since September 2002. Many aspects of the data quality of the BM series were discussed by Stübi et al. [1998], Calisesi et al. [2003] and Jeannot et al. [2007]. Prior to the transition to ECC sonde, series of dual flights were launched to compare BM and ECC sondes in real atmospheric conditions and to evaluate the impact of the change on the Payerne series. The analysis

of this dual flight data set is presented here. The sensitivity analysis of the data processing methods is presented in sections 2.2 and 2.3. Section 3 provides details on the dual flights performed in Payerne. Section 4 presents the analysis of the dual flights and an evaluation of the consequences of the transition on the Payerne series. The analysis of the continuity of the Payerne series following 5 years of ECC operation is also discussed.

## 2. Method Used to Process BM and ECC Ozonesondes Measurements

### 2.1. Ozonesondes Principles

[12] Ozonesondes were developed in the 1960s [Komhyr, 1969; Brewer and Milford, 1960]. They are based on the chemical reaction of potassium iodide in aqueous solution with atmospheric ozone molecules sampled outside the ozonesonde box by a small pump. Each single reaction, due to oxidation of the KI by ozone contained in the air sample, gives two electric charges producing a current between the two immersed electrodes. Assuming a 100% yield of reaction, the general form of the equation relating the measured current  $i(p)$  ( $\mu\text{A}$ ) to the ozone partial pressure  $O_3(p)$  (mPa) in the atmosphere is:

$$O_3(p) = cst \cdot (i(p) - i_o) \cdot P(p) \cdot T_p(p) \cdot F \quad (1)$$

where the constant  $cst$  (units  $mPa/K C$ ) is:

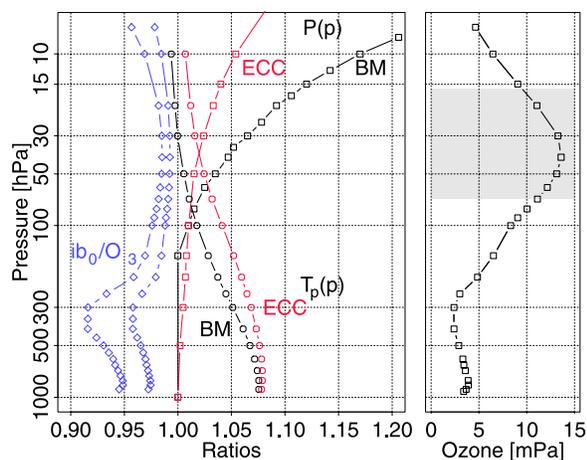
$$cst = \frac{R}{(2 \cdot e \cdot N_A)} = 0.043085 \quad (2)$$

$R$  is the universal gas constant,  $e$  the elementary charge,  $N_A$  Avogadro's number,  $F$  is the flow rate expressed as the time (s) needed to pump  $100 \text{ cm}^3$  of air,  $T_p(p)$  is the temperature (K) of the air when passing through the pump,  $P(p)$  is the pump efficiency correction and  $i_o$  is the background current. Besides the constant and the current, this formula contains terms characterizing the air flow entering the cathode cell. The pump temperature  $T_p(p)$  converts from volume to mass flow rate and the pressure dependant efficiency correction  $P(p)$  adjusts the pumping rate  $F$  measured in the laboratory at ambient pressure.

[13] Equation (1) applies to both BM and ECC ozonesondes but the methods of processing the data are slightly different between the two systems and this has an impact on the observed differences. The parameters of equation (1) are based on physics and chemistry concepts but their use in terms of measured quantities is subject to interpretation (see sections 2.2 and 2.3).

[14] To illustrate the different terms of equation (1), Figure 1 shows their respective contribution for BM and ECC sondes:

[15] 1. The pump temperature profiles  $T_p(p)$  drop gently from  $\sim 300 \text{ K}$  to  $\sim 280 \text{ K}$  (7% change) for the Payerne flight configuration. These are similar for the two types of sondes. The main factors influencing the temperature decrease are the sonde box thermal isolation, the heat source type and positioning (e.g., batteries, pump motor), as well as the thermometer position which is not strictly prescribed. The temperature is expected to be the coldest when the inner box temperature is used (free hanging thermistor) and the



**Figure 1.** (left) Contribution of the different factors of equation (1). Circles indicate pump temperature profiles  $T_p$  relative to 280 K for BM and ECC, diamonds indicate ratio of  $-0.1$  mPa and  $-0.2$  mPa equivalent background currents to the mean ozone profile given in Figure 1 (right), and squares indicate pump efficiency correction for BM [Komhyr and Harris, 1965] and ECC [Komhyr, 1986]. (right) Mean ozone profile measured at Payerne, Switzerland. The lower and upper limits of the grey strip correspond to the 25% and 75%, respectively, of the total ozone column.

warmest when the sensor is inserted in a hole drilled in the pump body. Some stations measure the inlet tube wall temperature assumed to be the same as the entering air temperature.

[16] 2. The background current  $i_0$  measured during the laboratory preparation procedure is  $\sim 0.03 \mu\text{A}$  (equivalent to  $\sim -0.1$  mPa in equation (1)) for the ECC sondes. Values of  $-0.1$  mPa and  $-0.2$  mPa were reported in Figure 1 (diamonds) as percentage deviation of the mean ozone profile shown in Figure 1 (right). Contrary to the smooth pump temperature curves, the background current produces the largest deviation just below the tropopause where ozone concentration is low.

[17] 3. The pump efficiency correction profiles  $P(p)$  smoothly increase with decreasing pressure and predominantly affect the upper part of the ozone profile. The correction profiles are 2 to 3 times larger for BM sondes than for ECC sondes.

[18] After the calculation of the ozone profile with equation (1), the sonde integrated profile is compared to independent total ozone column measurements (e.g., Dobson, Brewer) and a normalization factor is calculated. Applying this factor (Dobson/sonde) to the ozonesonde data guarantees that the column calculated from the profile is consistent with the other ozone column measurements. However, the partial ozone column above the balloon burst altitude has to be evaluated and therefore a hypothesis on the ozone distribution above burst altitude is needed.

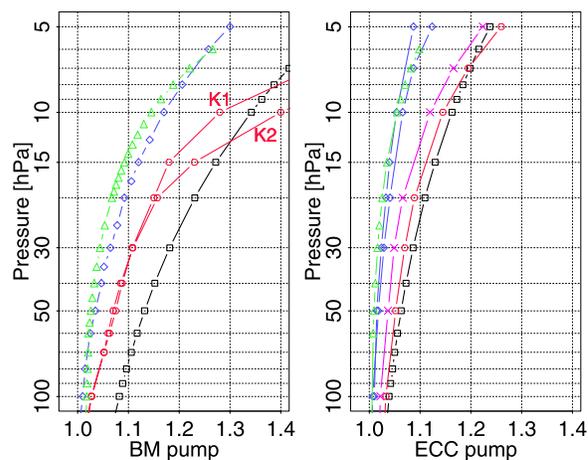
## 2.2. Method Used to Process the BM Data

[19] The standardized operating procedures for the BM ozonesonde, in particular the data processing, are defined in the WMO Standard Operating Procedures (SOP) [Claude et

al., 1987]. Taking into account that neither the pump temperature nor the background current were accurately measured in the past, equation (1) was simplified by (1) setting the background current to zero,  $i_0 = 0$ , and (2) assuming the pump temperature profile constant,  $T_p(p) = 280$  K. Assumption 1 implies that the calculated ozone may be overestimated and a larger relative error is expected in the troposphere where the ozone level is low. Assumption 2 creates an imbalance of the tropospheric part of the profile compared to the stratospheric part due to  $T_p(p)$  change of  $\sim 7\%$ . The two effects 1 and 2 tend to compensate each other from the ground up to about 50 hPa as seen in Figure 1 where the negative contribution of  $i_0$  (diamonds) cancels the positive contribution of  $T_p$  (circles). However, the background current depends strongly on the preparation procedure and assumption 1 may have had a larger variability than assumption 2 during the 40 years of ozone sounding activity.

[20] A third assumption (assumption 3) concerns the pump correction factor  $P$ . The metal/plastic assembly that constitutes the BM pumps leads to reduced efficiency at low pressure. The BM pump requires that the piston be lubricated and this affects the pump leakage and possibly leads to some ozone destruction. In the SOP, it is recommended to use the correction measured by Komhyr and Harris [1965] reproduced in Figure 1 which shows that this correction is larger than 20% below 8 hPa.

[21] Figure 2 gives different pump efficiency correction profiles proposed in the literature for BM and ECC sondes. The large scatter illustrates the difficulties involved in correctly determining methods to improve the original [Komhyr and Harris, 1965] corrections and to evaluate



**Figure 2.** Pump efficiency correction profiles from different authors for the BM and ECC sondes. (left) Correction for BM from De Backer et al. [1998b] (squares), K1 and K2 from Steinbrecht et al. [1998] (circles), Claude et al. [1987] (diamonds), and unpublished data from NASA/GFSC (B. Hoegger, private communication, 2002) (triangles). (right) Correction for ECC from De Backer et al. [1998a] (squares), Komhyr [1986] and Komhyr et al. [1995b] (diamonds), CMDL (circles) and UWY (crosses) data from Johnson et al. [2002], and NASA/GFSC data measured during the SONDEX campaign (mean of ECC sondes) (triangles).

**Table 1.** Parameters for the ECC and BM Data Processing Method Referring to Equation (1)<sup>a</sup>

| Parameter      | BM Method   | ECC Method   |
|----------------|---|--|
| $P(p)$         | <b>(1) standard [Komhyr, 1969]</b> , (2) Hohenpeissenberg [Steinbrecht et al., 1998], and (3) Uccle [De Backer et al., 1998a] | <b>(1) [Komhyr, 1986]</b> , (2) [Komhyr et al., 1995b], and (3) measured for individual sonde [e.g., Johnson et al., 2002] |
| $T_p(p)$       | (1) constant: 300 K [Claude et al., 1987] and <b>(2) constant: 280 K (Payerne)</b>  | <b>(1) measured in each flight</b>   |
| $i_o$          | <b>(1) NO correction</b> ( $i_o = 0$ )  | <b>(1) correction by a constant <math>i_o</math></b> and (2) decreasing $i_o$ with altitude                                |
| Dobson scaling | <b>(1) applied</b> and (2) not applied  | <b>(1) applied</b> and (2) not applied   |

<sup>a</sup>The methods written in bold are used in the present study.

possible changes in the performance of the sondes over the last four decades. Steinbrecht et al. [1998] proposed two alternatives (see Figure 2, left): the first (K1) is based on laboratory measurements and the second (K2) is an empirical correction deduced from the comparisons of measured ozone profiles by BM sondes and a colocated LIDAR. De Backer et al. [1998a] derived a parametric correction profile based on laboratory experiments. The curve (squares) presented in Figure 2 corresponds to the mean BM correction profile given by De Backer et al. [1998b] (their equation 1 with  $b = 1.56$ ). Measurements (triangles) from the NASA/GFSC laboratory [Torres, 1981; B. Hoegger, unpublished data, 2002] show BM pump efficiency corrections very close to the original Komhyr and Harris [1965] (diamonds) results. An additional temperature sensitivity of the pump efficiency correction  $P(T_p)$  was reported by De Backer et al. [1998b]. However, the pump temperature decrease compensates for the increase of pump efficiency associated with this temperature change. The net effect could be low sensitivity of the BM sondes to the pump temperature.

[22] The SOPs for BM sondes call for the normalization of the profiles which reduces the sonde to sonde variability and correct the  $\sim 10\%$  low bias of the ozone column evaluated from the BM profile. The largest contribution to the column comes from the ozone layer. This is emphasized in Figure 1 (right) by the grey strip that encompasses 50% of the total column. The residual column above the burst altitude represents 10–20% of the whole column and it is evaluated by assuming a constant mixing ratio (CMR). The CMR method was tested for the BM sondes by comparison with an ozone microwave radiometer [Calisesi et al., 2003]. The error associated with the CMR method to evaluate the residual column was estimated to be  $\sim 3\%$ .

[23] The contribution of the background current to the total column can be evaluated directly and it amounts to about 5.4 DU/0.1 mPa (integration from 1000 hPa to 1 hPa). For a column of 300 DU, this corresponds to a contribution of 1.8%/0.1 mPa of background signal.

[24] Table 1 summarizes the method and the parameters of equation (1) used in the standard BM data processing and the reference to other methods published in the literature has been listed.

### 2.3. Method Used to Process ECC Ozone Sonde Data

[25] Referring to equation (1), in ECC sondes the background current  $i_o$  is measured at ground level prior to the launch and the pump temperature profile  $T_p(p)$  is recorded during the flight. Therefore, the open questions regarding the ECC sondes data processing are the hypothesis of an altitude-independent background current, the pump efficiency

correction profile and the normalization of the ozone profile.

[26] Reid et al. [1996] compared ECC sondes with a chemiluminescent analyzer in the troposphere and found that the measurements agree within 4% if a constant background current correction is applied. This was confirmed in a dual flight at Payerne with ozone eliminating charcoal filters connected to the inlet tube of the two sondes during the flight. Both BM and ECC signals gave a constant residual signal within  $\pm 0.1$  mPa. Therefore in this analysis, the background current is assumed to be constant during the ascent.

[27] ECC pumps are made of Teflon and their efficiency is better than BM pumps at low pressure as seen in Figure 2 (right). However there is a difference by a factor of 2 between the different measurements reported in the literature. The recent JOSIE [Smit et al., 2007] and BESOS [Deshler et al., 2008] experiments show that the smaller pump corrections give better results compared to the UV photometer reference available in these two experiments. In the present study, the pump efficiency correction was selected according to the manufacturer recommendation [Komhyr, 1986].

[28] The normalization was systematically applied to the ECC data with the CMR method for the residual column to be consistent with the BM data processing.

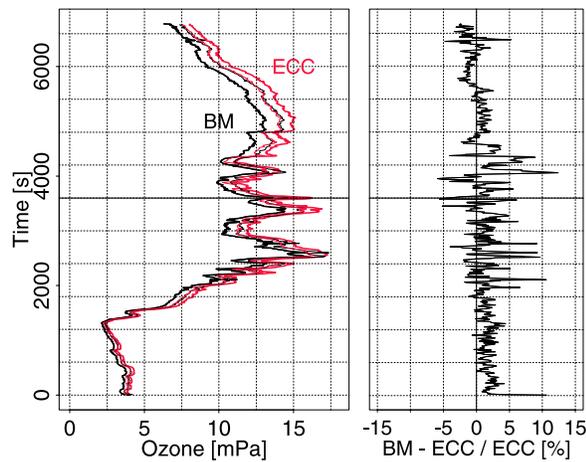
## 3. Measurements Used in This Study

### 3.1. Ozone Sondes Measurements at Payerne

[29] Payerne is a small city located on the Swiss plateau between the Jura Mountains in the northwest and the Alps in the southeast. The regular ozone sonde measurements, started in 1968, were done every Monday-Wednesday-Friday using the BM ozone sondes. The history of the BM series and the methods to minimize inhomogeneities are described by Jeannot et al. [2007]. The SOP for BM sondes was applied for the preparation of all the BM sondes in his study. The constant pump temperature was set to 280 K instead of 300 K specified in the SOP document [Claude et al., 1987] because it better reflected the Payerne flight conditions. This change does not affect the ozone profile but the normalization factor increases by 7%.

[30] The ECC sondes are provided by two manufacturers, ENSCI and SPC companies. Recent experiments have shown that the ECC sondes from the two providers produce slightly different results [Smit et al., 2007; Deshler et al., 2008]. Moreover, today they recommend using different sensing solution concentration with their sondes.

[31] The preparation procedure for ECC sondes follows the recommendations of the manufacturer [Komhyr, 1986]



**Figure 3.** Ozone profile of the dual flight from 19 April 2000. (left) BM and ECC ozone profiles before (thick lines) and after (thin lines) the normalization. The normalization factors are 1.09 for BM and 0.94 for ECC. (right) Relative difference of the normalized ozone profiles.

and the general practices of the ECC user community. One week in advance of the flight, the sonde cell is cleaned with a high level of ozone air flow and then filled with the sensing solution. After this, the sonde is operated for a sequence of three periods of 10 min: the first and third periods with “clean air” and the second period with a mixture of “clean air” and 0.2 ppm of ozone ( $\sim 20$  hPa). Then the cathode cell is completely filled with solution and the sonde is stored for a week. On the day of flight after a change of the sensing solution, the conditioning cycle is repeated about 3 h before the launch. The background current is measured at the end of the last sequence. These operations are computer controlled in Payerne (except for the change of solution) and therefore the preparation is reproducible from instrument to instrument. The pump temperature is measured during the ascent with a thermometer placed in a hole drilled in the pump block.

[32] The total ozone column from the Dobson located at Arosa (210 km East of Payerne) is used for the normalization of both profiles. Under cloudy conditions, satellite data are used instead.

### 3.2. Dual Flight Campaigns

[33] As already mentioned, since September 2002 the ECC ozonesondes have been the operational instruments for Payerne. To prepare the transition from BM to ECC, a large number of dual flights were launched to compare the two sonde types in operational conditions. Dual flights consist of attaching two independent sondes under the aerological balloon assuring the sampling of the same atmospheric environment, the two sondes being separated by only a 2 m boom. The data acquisition systems of each sonde are synchronized at the start which assures a time difference smaller than 1 s. The actual sampling of the data is about 7 s, representing the time needed for a complete cycle of measurements of temperature, pressure, humidity and two ozone parameters (current and pump temperature). The time elapsed since the launch is the more accurate coordinate to compare the sondes’ data.

[34] In Figure 3, an example of dual flight results is given with the profiles measured by the BM and ECC sondes (Figure 3, left). The two ozone profiles before the normalization are  $\sim 15\%$  apart (thick lines) whereas after the normalization (thin lines) they agree within  $\pm 5\%$  as show in Figure 3 (right).

[35] In the analysis, the time series of the differences between the two sondes are calculated and converted to a profile of ozone difference versus altitude for each flight. These difference profiles are then grouped in altitude layers (2 km wide) and the mean difference and the standard deviation within each layer are computed. The result is a mean ozone difference profile between the two sondes computed for different subsets of the whole set of dual flights. The use of altitude as the vertical coordinate is arbitrary; pressure could have been used as well because the temperature profile is available to convert one coordinate to the other.

[36] Strict criteria were applied to select the valid flights for the statistical analysis:

[37] 1. Flights with long transmission interruptions ( $>10$  min,  $\sim 3$  km) are excluded.

[38] 2. For shorter interruptions, the missing part is disregarded and not interpolated.

[39] 3. Short-term perturbations ( $<30$  s) are removed.

[40] 4. Flights where the difference between the two sensors increases with time are excluded. This is a sign of a significant drift of one or both sondes for unexplained reasons.

[41] The BM-ECC dual flight data set was collected during two main campaigns organized in Payerne:

[42] 1. SONDEX was a 2 week campaign (26 April to 4 May 1996) with 29 successful ascents. The sondes preparations were executed by the NASA/GSFC team for the ECC sondes and by the Payerne team for the BM sondes. The ECC sondes from two manufacturers (ENSCI, SPC) were used during this campaign, while the BM was produced by one manufacturer (Mast-Keystone).

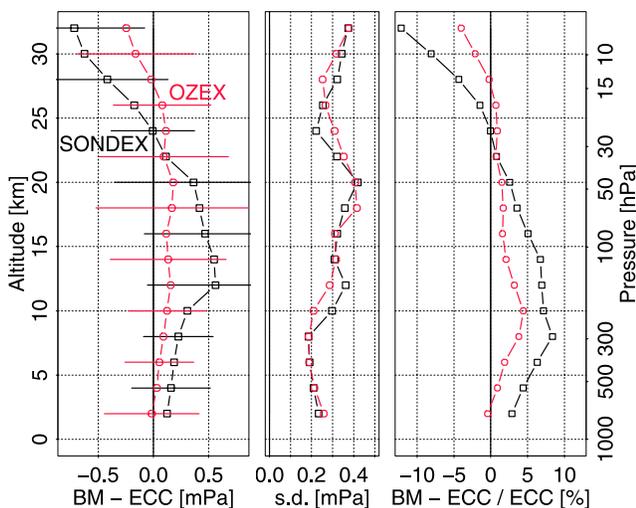
[43] 2. OZEX was a multiple-year campaign of regular dual flights (March 1998 to August 2002). The schedule of this campaign varied from one flight a week (intensive periods 1998–1999 and 2001–2002) to one flight a month (1999–2001). A total of 141 dual flights were recorded, out of which 111 passed the quality control and were used in the final analysis.

[44] In addition, 9 dual flights with 2 ECC sondes and 6 dual flights with 2 BM sondes were launched to evaluate the variability of each sonde type. Except for 22 sondes from SPC in the SONDEX campaign, all other ECC were purchased from ENSCI Corporation and all ECC sondes were operated with a 1% KI solution.

## 4. Results and Discussions

### 4.1. Results of Dual Flights

[45] The data set allows us to determine the average ozone difference profile between the two sonde types and also to evaluate the influence of the data processing on the comparability of these two systems. In Figure 4, the results are reported for the SONDEX and OZEX campaigns as ozone differences (mPa) in Figure 4 (left), the standard deviation (mPa) in Figure 4 (middle) and the relative ozone



**Figure 4.** Ozone difference profiles BM-ECC for the two campaigns: SONDEX 29 dual flights (squares) and OZEX 111 dual flights (circles). (left) Direct difference in mPa with the 90% confidence intervals. (middle) Standard deviation (mPa). (right) Relative difference BM-ECC/ECC (%). On the left side of the figure, the altitude scale is given together with the approximate pressure scale on the right side.

differences (%) in Figure 4 (right). The ozone differences are within the limits of  $\pm 0.7$  mPa for the SONDEX campaign (29 flights) and of  $\pm 0.3$  mPa for the OZEX campaign (111 flights). The 90% confidence interval in Figure 4 (left) is associated with the hypothesis that the difference BM-ECC is null. This hypothesis is verified for the OZEX campaign at all altitudes, whereas for the SONDEX campaign this is not the case above 30 km. Some doubts regarding the hypothesis exist also between 10 and 17 km. The standard deviations range from 0.2 to 0.4 mPa and present similar patterns for both campaigns. The relative ozone differences are  $\pm 10\%$  for SONDEX and  $\pm 5\%$  for OZEX. Generally, the SONDEX ozone differences are 2–3 times larger than the OZEX ones except at  $\sim 23$  km (ozone layer altitude) where the differences are close to zero. Subsets of the data have been considered in the analysis to explain these differences linked to the specificity of each campaign. The SONDEX data set was split according to the launch time within the day, the total column ozone, and the ECC sondes manufacturer. Only the last case is presented here, the other two cases show no influence of the tested parameter. The SONDEX flights with SPC (20 flights) and ENSCI (9 flights) ECC sondes were analyzed separately and the results are illustrated in Figure 5. The “BM-SPC” subset presents smaller differences compared to the “BM-ENSCI” subset. Therefore the presence of the SPC sondes in SONDEX does not explain the difference between the two campaigns seen in Figure 4 because the OZEX data set contains only “BM-ENSCI” pairs. Other recent studies have also reported the difference between the two manufacturers of ECC sondes and presently an effort is being made to understand and quantify this effect [Smit et al., 2007; Deshler et al., 2008].

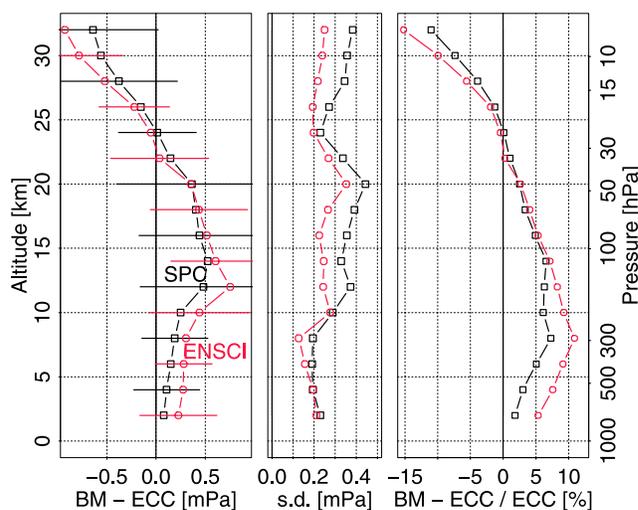
[46] Another reason for the difference between the two campaigns could be the short preflight preparation time

between the up to five successive dual flights a day during the SONDEX campaign. The BM sondes are particularly sensitive to the preparation of the sonde and in an intensive campaign it is difficult to assure a preflight preparation comparable to the operational service. It is difficult to quantify this effect but it could also be the reason for contradictory results from the past campaigns (e.g., Hilsenrath et al. [1986] versus Kerr et al. [1994]). Despite the absence of clear explanations for the difference between the two campaigns, the discrepancies between the two profiles shown in Figure 4 stay within  $\pm 5\%$ , which is within the uncertainty observed in the past campaigns.

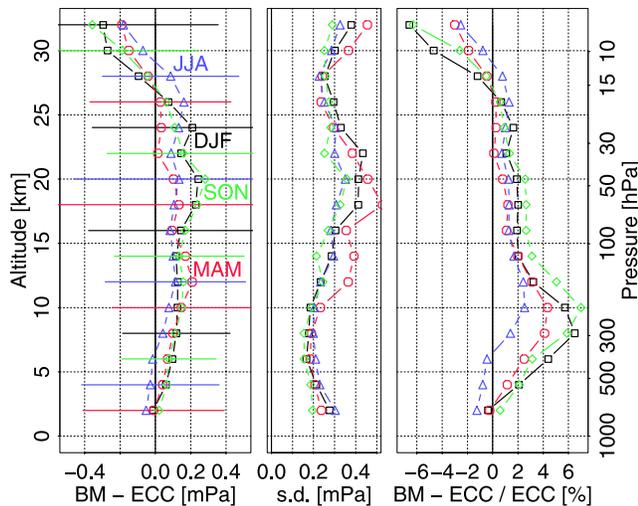
[47] Contrary to SONDEX, in the OZEX campaign all the preparations were done in accordance with the operational procedures. Therefore, the result of the OZEX campaign is certainly more representative of the difference between BM and ECC sondes in the conditions of the operational service. To confirm the robustness of the OZEX results, the data set was split according to different conditions.

[48] To check whether the two intensive periods 1998–1999 and 2001–2002 give the same results, the dual flights launched before and after 2000 were analyzed separately. The two BM-ECC difference profiles were compared but no significant difference appears and therefore they are not reproduced here. This result concludes that over the 1998–2002 period the data are consistent as regards the annual mean difference and no effect associated with different batches of sondes is detected.

[49] Seasonal effects were looked at by the calculation of the four seasonal difference profiles reported in Figure 6. The 90% confidence intervals indicate that the seasonal influence is not significant. But Figure 6 shows an underlying systematic bias within  $\pm 0.3$  mPa on the direct ozone differences or  $\pm 6\%$  on the relative differences depending on the season. In summary, the analysis of the two sub samples of the OZEX data sets support the conclusion that the BM-ECC difference is not sensitive to the season and the results



**Figure 5.** Ozone difference profiles BM-ECC similar to Figure 4 for the SONDEX campaign. The data set was separated according to the manufacturer of the ECC sondes: 9 dual flights with “ECC-ENSCI” sondes (circles) and 20 dual flights with “ECC-SPC” sondes (squares).



**Figure 6.** Ozone difference profiles similar to Figure 4 for the seasonal analysis of the OZEX data set: DJF (squares), MAM (circles), JJA (triangles), and SON (diamonds).

remained stable over the period 1998–2002 of the OZEX campaign.

[50] The confidence intervals are determined by the standard deviations that combine the variability of both ECC and BM sonde types. To get an estimate of the individual contribution, a few dual flights with sondes of the same type were launched. In Figure 7 (left), the results of the analysis of 9 ECC-ECC compared with 6 BM-BM dual flights are presented as the RMS difference profiles since there is no distinction between the two sondes in these flights. Even though the number of flights is quite small, the BM sondes show a larger dispersion than the ECC sondes, particularly near the surface and at the top of the profile. Therefore, the standard deviation of the Figures 4–6 are dominated by the poorer BM reproducibility which is in accordance with the conclusion of the JOSIE 1996 experiment [Smit and Kley, 1998].

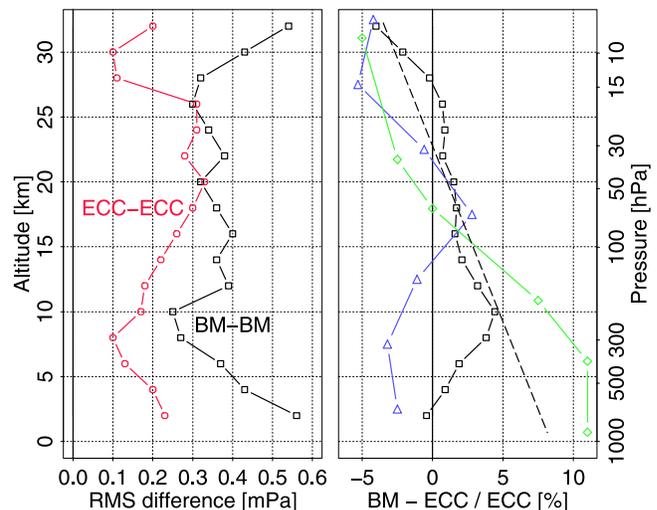
[51] Only a few similar studies have been published in the last 10 years. De Backer et al. [1998b] have published the analysis of 26 BM-ECC dual flights and their relative ozone difference profile is reproduced in Figure 7 (right) (triangles). The largest differences with the present OZEX analysis are seen in the troposphere which is probably linked to the much larger normalization factor at Uccle station (mean factor 1.2) compared to Payerne or Hohenpeissenberg stations (mean factor 1.05 to 1.10).

[52] The JOSIE 1996 experiment produced ozone difference profiles for BM and ECC sondes compared to the reference photometer [Smit and Kley, 1998]. However in 1996 the responses of the different ECC sondes were still presenting large differences due to various preparation procedures. But from the JOSIE 1998 campaign, a reliable difference profile between ECC sondes and the same reference photometer was determined [Smit and Sträter, 2004b]. Therefore, the combined JOSIE 1996 and JOSIE 1998 results allow calculation of the BM-ECC ozone difference profile from the simulation chamber experiments. In Figure 7 (right), this calculated profile is reproduced (diamonds). The agreement above 15 km is good, even

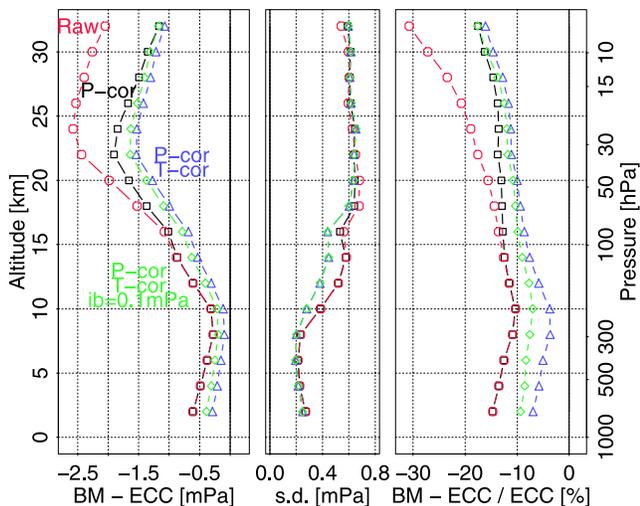
though the number of simulations is limited to 6 in JOSIE 1996. Below 15 km, the difference is larger, mainly because of the “ECC-photometer” differences because the BM sondes in the JOSIE 1996 were very close to the photometer.

[53] Other authors have deduced BM-ECC ozone difference profiles using satellite data as a transfer mechanism to link measurement periods done with BM and with ECC sondes [Lemoine and De Backer, 2001; Lehmann, 2005; Fioletov et al., 2006]. In accordance with the present results, these analyses showed that a good agreement was found between the BM and ECC sondes at the ozone layer altitude and a 3–6% negative bias was present above it. No reliable differences were obtained in the troposphere because of the limited satellite coverage below the tropopause.

[54] The conclusions based on the OZEX campaign are valid for the ECC sondes from ENSCI and with a 1.0% KI solution. The influence of the solution strength (e.g., 1% KI versus 0.5% KI) on the ozone profile was quantified [Smit et al., 2007; Deshler et al., 2008]. A crude empirical correction to simulate a change of solution concentration from 1% to 0.5% KI is proposed by Deshler et al. [2008]. This linear correction in  $\ln(p)$  corresponds, in terms of relative difference, to a tilt of the vertical axis by 11% between 1000 mPa and 10 mPa as illustrated by the dashed line in Figure 7 (right). This line is placed so that it crosses the vertical axis at ~23 km (ozone layer) to reproduce the effect of the normalization which minimizes the difference at that altitude. The dashed line shows that the ozone relative difference between BM and ECC sondes with a 0.5% KI solution would have been smaller than with the 1.0% KI solution used here (square line closer to the dashed line than to the vertical axis). In the lower troposphere, the differences are



**Figure 7.** (left) Mean RMS difference profiles for dual flights with sondes of the same type: 9 ECC-ECC (circles) and 6 BM-BM (squares). (right) Relative ozone difference BM-ECC/ECC (%) for different campaigns: OZEX (squares) and JOSIE (diamonds) experiments [Smit and Kley, 1998; Smit and Sträter, 2004b] and De Backer et al. [1998b] (triangles). The dashed line illustrates the empirical correction for a change of ECC solution concentration [Deshler et al., 2008] (see text).



**Figure 8.** Ozone difference profiles at the different steps of the BM data processing applied to the OZEX data set. The ECC data before the normalization are used here. The different steps are raw BM data (circles), with pump efficiency correction (squares), including the pump temperature (triangles), and a -0.1 mPa equivalent background current (diamonds).

larger but the crude correction from *Deshler et al.* [2008] is not better than  $\pm 5\%$  in the troposphere.

[55] The overestimation of the ozone of the 1.0% KI solution compared to 0.5% KI solution was also confirmed at different ozone sounding stations where dual flight “ECC-0.5%”–“ECC-1.0%” campaigns were organized. The results of these campaigns have been analyzed and a separate publication is under preparation.

[56] The results of the analysis presented in this paragraph are based on the data processing discussed in sections 2.2 and 2.3. It is necessary to quantify the influence of the processing by doing a sensitivity analysis that will be presented in the next section.

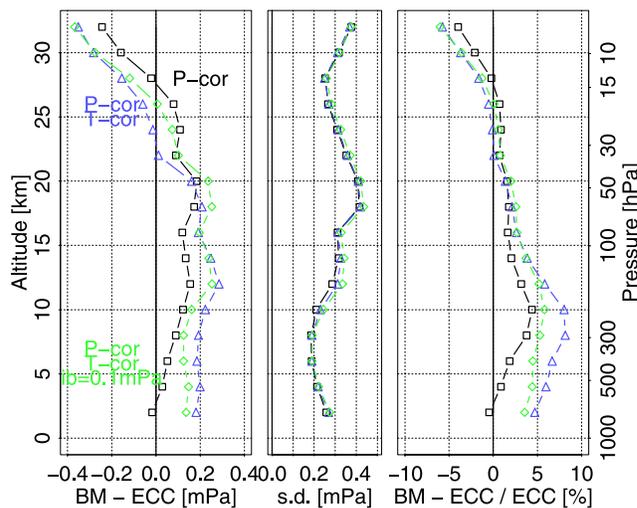
#### 4.2. Sensitivity Analysis of the Method Used to Process BM Data

[57] In this sensitivity analysis, the processing method for the BM data is changed while the ECC data processing is analogous to the previous section. As discussed in 2.2, the data treatment consists of different steps which are illustrated in Figure 8 with the OZEX data set. The leftmost line (circles) shows the ozone difference between the ECC data (before normalization) and the raw BM data obtained by the conversion of the measured current according to equation (1) with a constant temperature ( $T_p(p) = 280$  K) and the measured pump flow  $F$ . The negative differences are as much as  $-2.5$  mPa at the ozone layer altitude. The standard deviations increase from 0.2 mPa in the troposphere to 0.6 mPa in the stratosphere. The pump efficiency correction is then applied (squares) which affects the upper part of the profile. The differences are now reduced to  $-2$  mPa and it appears in Figure 8 (right) that the relative difference profile is approximately constant. This means that the BM and ECC sondes responses differ by a constant factor. The last two curves in Figure 8 were calculated to show the effect of

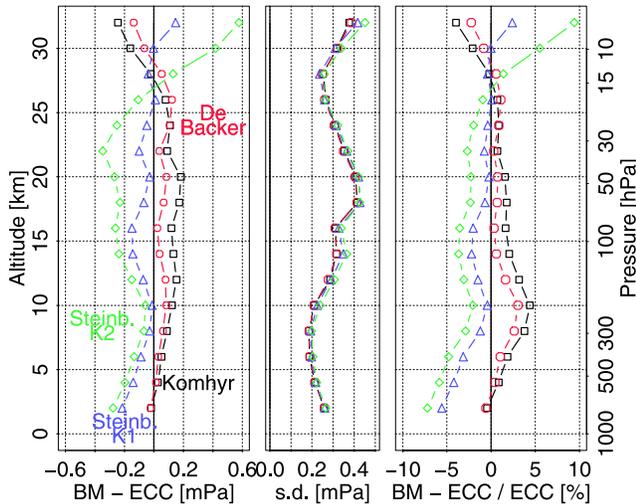
releasing hypotheses 1 and 2 described in section 2.2 for the analysis of the OZEX data. The line with triangles corresponds to the BM data calculated with the mean pump temperature profile measured during the OZEX campaign. The line with diamonds further simulates a  $-0.1$  mPa equivalent background current. These curves illustrate the magnitude of the different terms of equation (1) which tends to compensate or attenuate their individual effect.

[58] In Figure 9, the results of the same procedure as for Figure 8 are given but the normalization of the data is done after each step. The relative difference of  $\sim 15\%$  presented in Figure 8 (right) is reduced as expected. The BM data are increased by  $\sim 10\%$  (BM underestimate ozone) while the ECC data are reduced by  $\sim 5\%$  (ECC overestimate ozone) by the normalization. This is in accordance with the observation that ENSCI with 1.0% KI solution ECC sondes overestimate column ozone while the opposite is true for BM sondes [*Smit et al.*, 2007; *Deshler et al.*, 2008]. The use of a real pump temperature profile  $T_p(p)$  for the BM sondes increases the difference at the top and the bottom of the profile. It is possibly linked to the low pump temperature sensitivity mentioned in section 2.2. The background current reduces the effect of the pump temperature. The ozone differences for the three curves in Figure 9 (left) is of the order of  $\pm 0.3$  mPa but the standard deviations decrease notably in the stratosphere (0.3–0.4 mPa) compared to Figure 8 (0.6 mPa).

[59] The use of normalization by independent total ozone is presently being debated. While BM sondes are corrected according to the SOP, ECC sondes are usually not normalized. The analysis of the present data set shows that the normalization introduces a bias that is comparable to ones associated with other factors of the data processing method (e.g., real pump temperature profile and background current). Another debate concerns the use of the normalization for the tropospheric ozone measurements from the BM sondes [*WMO*, 1998]. On the basis of comparisons with aircraft measurements of the MOZAIC project, *Thouret et*



**Figure 9.** Same results as in Figure 8 (without raw BM data) but the normalization of the data is applied after each step of the data processing.



**Figure 10.** Ozone difference profile for the OZEX data set calculated with different pump efficiency corrections for BM sondes: K1 (triangles) and K2 (diamonds) [Steinbrecht et al., 1998], Komhyr and Harris [1965] (squares), and De Backer et al. [1998a] (circles).

al. [1998] questioned the use of normalization for tropospheric ozone. Another recent comparison between aircraft data and BM data from Payerne and Hohenpeissenberg provides evidence that the response time of the BM sensors needs to be taken into account and that agreement is better when using normalized ozonesonde measurements [Schnadt Poberaj et al., 2007].

[60] From the discussion of sections 2.2 and 2.3, it follows that the pump efficiency correction is the most uncertain parameter for ozonesonde data processing. Different laboratory measurements of  $P(p)$  give contrasting results both for the BM and the ECC sondes. In Figure 10, the result of the analysis of the OZEX data set with different BM pump efficiency corrections is presented. Changing this correction has an impact not only at the top of the profile but also at the bottom due to the normalization because the residual column increases for larger pump correction. The correction proposed by De Backer et al. [1998b] (circles) gives an ozone difference profile comparable to the SOP case (squares [Komhyr and Harris, 1965]) while the two alternatives (K1 and K2) proposed by Steinbrecht et al. [1998] produce pronounced changes of the difference profiles.

[61] As shown in this section, the pump temperature, the background current, the pump efficiency correction and the normalization all have a significant impact on the BM ozone profile and consequently on the BM-ECC difference profile. It is therefore necessary that the methods used to process ozonesondes data are documented by the data providers. The BM data processing method defined in the SOP produces ozone profiles which agree well with the ECC ozone profile provided that both data sets are normalized. Under these conditions, the ozone differences are not significant at the 90% confidence level. As a result, it was concluded that the change of the BM sonde to the ECC sonde should not imperil the consistency of the Payerne long-term ozone sounding series. After the first 5 years of

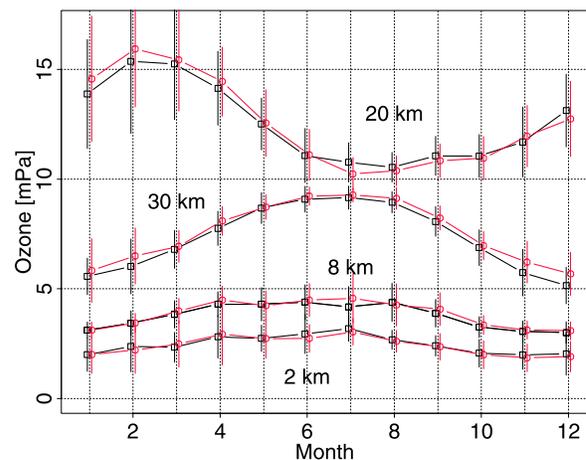
ECC soundings at Payerne, it is possible to test this conclusion as presented in the next section.

#### 4.3. Time Series Analysis of the BM to ECC Transition

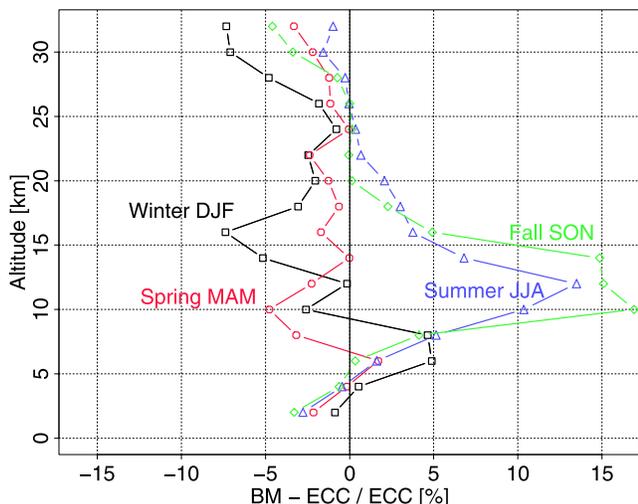
[62] As has already been mentioned, the operational ozone sonde for the Payerne station was changed in September 2002. It was decided to use the ECC sondes from ENSCI Corporation with the 0.5% KI solution for the operational service. In this section, the continuity of the Payerne soundings series covering the last 5 years of BM data and the first 5 years of ECC data is analyzed. The midlatitude trend over this period is negligible as described in the recent ozone assessment [WMO, 2007].

[63] The data was averaged for monthly values and for 16 altitude layers (2 km wide). In Figure 11, the mean yearly cycles for 4 altitude layers are presented separately for the BM 1997–2002 period and the ECC 2002–2007 period. The standard deviation bars overlap at all altitudes and no systematic difference between BM and ECC is seen in Figure 11. Each of these 16 time series was analyzed with standard homogeneity testing tools [Easterling and Peterson, 1995; Alexandersson and Moberg, 1997]. The large interannual variability of the ozone limits the detection of an underlying rupture of the monthly mean time series. Even for the altitudes presenting the lower variability, the homogeneity tests do not reveal a significant break (90% level) at the time of the change from BM to ECC sondes.

[64] The mean seasonal profiles for the last 5 years of BM ozone soundings and for the first 5 years of ECC ozone soundings at Payerne were calculated separately. In Figure 12, the relative differences of the corresponding ozone profiles are given for each season. The relative differences are between  $\pm 7\%$  except for the summer and fall profiles in the altitude range 10–15 km where the differences are  $\pm 15\%$ . These larger differences reflect the important interannual variability of the ozone between the tropopause and the ozone layer altitudes observed at midlatitude stations. The comparison of Figures 6 and 12



**Figure 11.** Mean annual cycle at four altitudes measured with BM sondes (squares) between September 1997 and August 2002 and with ECC sondes (circles) between September 2002 and August 2007. The standard deviations are also reported. The symbols are shifted to prevent an overlap.



**Figure 12.** Relative difference of the mean seasonal ozone profiles calculated on the 5 year period before (BM sondes) and on the 5 year period after (ECC sondes) the change from BM to ECC ozonesondes in Payerne.

is inappropriate because first, the operational ECC sondes are operated with a 0.5% concentration solution instead of 1.0% used for the OZEX campaign and second, Figure 12 is not the difference of coincident BM and ECC ozone measurements.

[65] The two independent analyses presented in sections 4.1 and 4.3 of the ozone difference between the BM and the ECC ozonesondes show consistent results. From the dual flight campaigns, it is concluded that the observed ozone differences between BM and ECC are not significant. From the 10 year time series analysis, it is concluded that no significant ruptures are detected at the time of the change of ozonesonde type. However, the reliability of the combined series for long-term trend analysis in the upper troposphere and the lower stratosphere will be further evaluated with MOZAIC regular aircraft measurements and with surrounding stations when a longer time period becomes available.

## 5. Summary and Conclusions

[66] In this study, two analyses of the difference between the BM (Brewer-Mast) and the ECC (electrochemical concentration cell) ozonesondes are presented. The first analysis is based on a data set of 140 dual flights resulting from a campaign in 1996 and a program of regular dual flights launched between 1998 and 2002. The second analysis is based on the Payerne time series of the last 5 years of BM soundings (1997–2002) compared to the first 5 years of ECC soundings (2002–2007).

[67] The first analysis shows that the BM and ECC ozone profiles agree within  $\pm 5\%$  or  $\pm 0.3$  mPa with a standard deviation between 0.2 and 0.5 mPa depending on the altitude. The BM-ECC ozone differences are not significant at the 90% confidence level. This result is obtained provided that the data of both sondes are normalized to an independent ozone column measurement.

[68] The second analysis shows that no discontinuity in the Payerne sounding series (1997–2007) is presently detected at the time of the change of sonde from BM to ECC which occurred in September 2002. The observed differences are not significant compared to the ozone variability over the 10 year period considered.

[69] For BM sondes, the data processing method is defined in the standard operating procedures. For the ECC sondes, the operating procedures have been optimized in recent years and they are now rather uniform within the global ozone sounding networks. But no standard has yet been approved.

[70] On the basis of the dual flight data set, a sensitivity analysis is presented for the different factors affecting the ozone profile calculation, e.g., the pump temperature, the background current, the pump efficiency correction and the normalization. Each factor has a significant influence on the ozone profile which was quantified for the BM ozonesondes. However, these influences are not independent in the final ozone profile as a drawback of the normalization.

[71] The present study allows us to conclude that it is possible to change from BM to ECC ozonesondes without affecting the quality the long-term ozone sounding series.

[72] The current comparison of BM and ECC ozonesondes in atmospheric conditions is complementary to the JOSIE experiments in the Jülich simulation chamber [Smit and Kley, 1998; Smit and Sträter, 2004b]. Both results are in agreement.

[73] **Acknowledgments.** The authors are grateful to the sounding team from Payerne Aerological Station for having performed these dual soundings over many years in parallel to their regular operational duties. The NASA team in charge of the ECC sondes during the SONDEX experiment is warmly acknowledged.

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B. Hoegger, G. Levrat, R. Stübi, and P. Viatte, Payerne Aerological Station, MeteoSwiss, CH-1530 Payerne, Switzerland. (rene.stubi@meteoswiss.ch)

F. J. Schmidlin, Wallops Flight Facility, NASA Goddard Space Flight Center, Wallops Island, VA 23337, USA.

J. Staehelin, Institute for Atmospheric and Climate Science, ETH Zürich, CH-8092 Zürich, Switzerland.