General comments
By analyzing a suite of laboratory and field measurements comparing two ECC ozonesonde types with two different sensing solution concentrations and UV reference standards the authors derived simple transfer functions between the possible combinations. The study builds on former studies with limited data sets. The new study merges those data sets and adds unpublished supplementary data. That enables them to conclude simple transfer functions to be usable in most practical cases in order to homogenize the international ozonesonde data record. Since there is an urgent requirement for a homogenized ozonesonde data record the authors can be congratulated for their efforts. I strongly recommend the publication of the manuscript after some minor revisions.

Specific comments
1. From the description of the data and the way they are used it seems that there is some implemented redundancy not resolved correctly in the BESOS and Laramie data. This should affect the statistical errors. In both cases a set of three ozonesondes with an equal type/sensor combination are compared to another set of three ozonesondes with another type/sensor combination. In the BESOS case each ozonesonde of the first group is compared to each ozonesonde of the second group leading to 9 individual comparisons (line 348, Table 1). In the Laramie case “only” 6 individual comparisons are listed for unknown reasons (Table 1). In any case the 9 or 6 comparisons cannot be seen as independent. There can only be 3 independent comparisons. Each ozonesonde can only be used once or one has to deal with covariance matrices in the error analysis.

   Good catch on the 6 Laramie cases. One of the 0.5% KI ozonesondes was eliminated from the comparison by NOAA due to a poor performance in flight. This is now explained in the text. To test the reviewers comment the covariance and correlation matrices were calculated. The correlation between identical ozonesondes and concentrations in any comparison are > 0.999. The correlation of sondes of different type or the same sonde type with different concentrations is not quite as high, but > 0.998. The correlation coefficients between any combination of the possible comparisons for any six sondes in a comparison are all very similar, which suggests to us that assuming 9 independent comparisons, from any three pairs of sondes, is reasonable. If the reviewer’s comment were followed and only three independent comparisons were made, this independent comparison could be done three different ways. Any one sonde in the first group could be compared with sonde 1, 2, or 3 in the second group, so 3 permutations of the “3 independent” comparisons are possible. The result is back to 9 comparisons. In any case the impact is minimal on our final result, and so the analysis is left as it is.

2. At least Wallops Island used some SP 5A sondes. One should mention that those sondes had no explicit hole for the pump temperature sensor. Can you see any differences in the behaviour? A sentence has been added noting the lack of a specific place for the temperature sensor. There is no obvious difference in the behavior of these sondes.
3. The paragraph (lines 114-125) can be misinterpreted that no other sensing solution concentrations than 0.5% and 1.0% had been used. I recommend adding that few other concentrations, i.e. 2.0%, had been used, too. A sentence has been added to clarify this.

4. The effect of different pH buffers have been mentioned twice (sections 2.4 and 5.) Which buffers had been used in the measurements? Had the same buffer been used everywhere? A paragraph is added to section 3.1 to state that all comparisons were completed using the standard solution concentrations with the recommended buffers and the recipe for the solutions is described here.

Technical corrections
1. Please find a way to mention at least once that the pressure $p$ in $\log_{10}(p)$ has to be used in the unit hPa. Thank you. This was included in the abstract and in the body of the paper the first time this is described.

2. Line 348-349: Table 1 instead? corrected

3. Line 1063: Delete one parenthesis after $d$. corrected

Anonymous Referee #2
Review of Deshler et al., AMT, 2016. Prepared 10 Feb 17

OVERALL COMMENT & RECOMMENDATION. Transfer functions (“TF”) are important for the re-processing of thousands of ozonesonde records and this paper is the first to provide them based on extensive analysis. The paper overall is clearly written and with a few exceptions mentioned below, well-organized, giving TF that the sonde community will accept and use. There is one major short-coming in the paper that needs to be remedied before the paper can be published. There is no uncertainty analysis in the TF, which will propagate to the adjusted ozone reading (refer, for example, to Table 3).

Thank you for this comment. In answering this comment a small error was found in the way the uncertainty for the linear slopes in Table 3 were calculated. This led to some reduction in the calculated uncertainties and thus some modifications to Table 3. A first estimate of the uncertainties in the transfer functions can be obtained by the weighted uncertainties provided for each box in Table 3. These uncertainties now vary from 0.037 to 0.085, depending on the particular comparison considered. Rather than include these individual uncertainties to create the error bars in Figures 4 and 5, as done in the draft submitted, a more general uncertainty was desired which would apply to all the transfer functions and to the comparison of the manufacturers recommendations, SP1.0 and EN0.5. The number settled on was 0.05 as an uncertainty on the slopes. This is an approximate average of the individual uncertainties. This uncertainty, 0.05, at times over-estimates the variations shown in the data and at times underestimates it, but overall does a reasonable job, Figures 4, 5, and 6.

OTHER COMMENTS.
Abstract. Needs an Intro/motivating sentence. Although this is a technical paper in a “measurements” journal, every Abstract needs a “why it matters” opening sentence or two to
give context to the study for some of this paper’s readers. *Several motivating sentences were added to the beginning of the abstract.*

Line 25 – goal of “this study” *corrected*

Lines 39-40: Although ENSCI and SPC are spelled out in line 21, the abbreviations SP and EN are not yet clearly defined at this point. Please fix. *corrected*

End of Introduction. An excellent job in describing the motivation and goals but there is not a user-friendly road map of the upcoming Sections at the end. Please add! The prose gets very specialized very fast. *A road map is added.*

Line 199: All these factor(s) – plural. “superpose” ? *corrected factors and added superpose.*

Consider that the last two paragraphs of section 2.1 and section 2.4 seem to talk about the same thing i.e. stoichiometry. Can they be merged or revised? It gets confusing coming back to this topic. *Thank you. The paragraph from section 2.4 was deleted and the paragraph in section 2.1 was embellished.*

Section 2.5 should be in the Introduction section. It is actually major motivation and important for putting the study into context. Further, Sec 2.5 is not relevant to this section entitled ‘ECC ozonesonde description’. *Thank you for this comment. This section was moved and the introduction subdivided into two sections after the general introduction, this being the second of the two subsections.*


Section 3 title: Transfer function’s’ – plural – there is more than one equation derived. *corrected*

Section 3.1: Be clear up front that only JOSIE data are in controlled environments. The rest of the comparison data came from experiment flown under real atmospheric conditions. *text has been revised to reflect this.*

SOPs during these varied dual/gondola flights in space and time must have been different. Is this not an important source of inhomogeneity? Please address clearly. How do you justify using
these data to come up with unified transfer functions when SOPs have varied? You mention in Section 2.2, line 20 that “ECC ozonesondes prepared according to the SOPs provide very reproducible (<2-3%) measurements.” A section on the impact of varying SOPs should be added, if only to justify the quality of the datasets used. In fact the investigators were all careful to follow the WMO SOPs for the preparation of the dual ozonesonde comparisons. This point is now made in a second paragraph added to section 3.1.

Section 3.1.1 Were all JOSIE measurements used (including the tropical simulations)? All other dual/gondola flight were flown in mid to polar latitude locations so the assumption here is that one would use similar latitude profiles. All the JOSIE data that compared SP/EN at one KI concentration and SP or EN at 0.5/1.0 concentration were used. We did not remove any tropical profiles, nor should we have. The transfer functions are meant to apply throughout the ozonesonde ozone concentration range. Similarly data from McMurdo under ozone hole conditions were used.

Figure 1: Why are there negative backgrounds currents at Wallops? Is that an error? Please explain. The latter background currents show large variations from negative to -0.05uA to close to 1.0. Isn’t the legitimacy of the transfer functions affected by variations on background current? Why do you not use one background current for all the data to remove this artifact and recalculate the transfer functions? What is the dependence of background current on solution, manufacturer, and ground-station equipment to produce zero-ozone? It is understood that backgrounds are poorly understand and that there is a dearth of literature that focuses on this very topic. The negative background currents indicated are included as these were what was reported at the time. Text is added to comment on these negative values. The station manager at the time commented on these values as follows: “During the period July 1995 to late 1999 Wallops experienced about 25 percent negative background currents. At the time, and being new to ozone, I accepted the view of the technician preparing our ECC’s that negative BG currents were acceptable.” In any case since similar background currents would affect both ozonesondes in a comparison the effect on the analysis is negligible. The other questions raised by the reviewer here are interesting, but beyond the scope of the analysis presented in this paper. As pointed out in the paper several times, backgrounds between comparison pairs of ozonesondes were usually quite similar, thus changing these numbers in some uniform fashion, such as one background for all comparisons, would have no impact on the analysis. The differences between ozonesondes of different type or concentration are much bigger than the 0.1 – 0.2 mPa of ozone represented by the background currents, which is less than the minimum ozone measurement considered in the analysis.

Figure 5: ‘DP’-- is that data points? Yes, a note has been added to the caption to clarify this.

Table 2 – Add the sample size whether by profile number or data points - It is worthwhile to add that information here, more so than to the figure: one can save space and increase the font size for clarity! This sounds much easier than it is. To do this would require adding four additional columns to the table, one for the number of data points for each pressure interval. This would make the table unwieldly. The number of data points is added to the figure to give the reader the
idea of the number of data points which went into each comparison. It is not meant to be comprehensive nor to provide the number of data points for each comparison.

Since you refer in detail to this table in term of ‘boxes’ - suggest adding another column numbering them. Such a column was added and the text changed accordingly in reference to Table 2.

Section 4.1
Were the outliers observed and noted in Figure 6 included in developing the transfer functions? If so, why? Outliers denote error or responses outside the norm. By how much would the transfer functions change if outliers were removed? If it is negligible, it should be calculated and noted, even if not included. The only data that were removed were measurements when either measurement of a pair were < 0.5 mPa. Thus some, but far from all, the data in the gray hashed areas in Figure 6 were removed. Thus a significant fraction of the outliers noted were included in the analysis, but their effect was negligible due to the large number of comparisons.

Paragraph on line 511. This sentence is long, convoluted, and unclear. The meaning comes across but the sentence (s) can be written better. The long sentence has been split into 2 sentences.

Line 628. The Logan references (appropriate on Line 302) do not belong here. We’ve removed the Logan references from this line.

Figure 8 – Use the same x-axis scale as Figures 6 and 7. Done

Section 4.4 Are you are applying the ENSCI/1.0% to ENSCI/0.5% transfer function and this is equation 6? This is mentioned nowhere in this section. The error bars should not change as you are merely shifting the profile. The key is that the distribution now centers closer to zero. Yes we are applying the EN1.0 to EN0.5 transfer function, which is Eq. 5, and this is stated in this section, “This data set is used here to illustrate the application of the transfer function OZ_conc defined in Eq. 5.” In fact, the error bars do not change they are just shifted to a new center point.

Methods to homogenize ECC ozonesonde measurements across changes in sensing solution concentration or ozonesonde manufacturer

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Abstract.

Ozone plays a significant role in the chemical and radiative state of the atmosphere. For this reason there are many instruments used to measure ozone from the ground, from space, and from balloons. Balloon-borne electrochemical cell ozonesondes provide some of the best measurements of the ozone profile up to the mid stratosphere, providing high vertical resolution, high precision, and a wide geographic distribution. From the mid 1990s to the late 2000s the consistency of electrochemical cell ozonesonde long-term records from balloon-borne ozonesondes has been compromised by differences in manufacturers, Science Pump (SP) and ENSCI (EN), and differences in recommended sensor solution concentrations, 1.0% potassium iodide (KI) and the one half dilution 0.5%. To investigate these differences a number of organizations independently undertook comparisons of the various ozonesonde types and solution concentrations, resulting in 197 ozonesonde comparison profiles. The goal of this study is to derive transfer functions to allow measurements outside of standard recommendations, for sensor composition and ozonesonde type, to be converted to a standard measurement and thus homogenize the data to the expected accuracy of 5% (10%) in the stratosphere (troposphere). Subsets of these data have been analyzed previously and intermediate transfer functions derived. Here all the comparison data are analyzed to compare: 1) differences in sensor solution composition for a single ozonesonde type, 2) differences in ozonesonde type for a single sensor solution composition and 3) the World Meteorological Organization’s (WMO) and manufacturer’s recommendations of 1.0% KI solution for Science Pump and 0.5% KI for ENSCI. From the recommendations it is clear that ENSCI ozonesondes and 1.0% KI solution result in higher amounts of ozone sensed. The results indicate that differences in solution composition and in ozonesonde type display little pressure dependence at pressures ≥ 30 hPa and thus the transfer function can be characterized as a simple ratio of the less sensitive to the more sensitive method. This ratio is 0.96 for both solution concentration and ozonesonde type. The ratios differ at pressures < 30 hPa such that OZ⁰.⁵% / OZ¹.⁰% = 0.90 ±0.041 • log₁₀(p) and OZ_{SciencePump} / OZ_{ENSCI} = 0.764 ± 0.133 • log₁₀(p), for p in units of hPa. For the manufacturer recommended solution concentrations the dispersion of the ratio (SP-1.0/EN-0.5), while significant, is generally within 3% and centered near 1.0, such that no changes are recommended. For stations which have used multiple ozonesonde types with solution concentrations different from the WMO’s and manufacturer’s recommendations, this work suggests that a reasonably homogeneous data set can be created if the quantitative relationships specified above are applied to the non-standard measurements. This result is illustrated here in an application to the Nairobi data set.
1. Introduction

Ozone is one of the critical atmospheric trace gases. Ozone contributes to the oxidizing capacity of the troposphere, to the absorption of terrestrial IR radiation, and to the absorption of solar UV in the stratosphere. An overabundance of ozone in the troposphere causes air quality problems, while a deficit in the stratosphere leads to enhanced exposure to UV. Ozone measurements are thus required to maintain our understanding of these processes and are required over times scales of hours to years, and from single point measurements to vertical profiles to the mid stratosphere. Measurements are required over hours at single locations to characterize air quality, while regular profiles over decades are required to characterize stratospheric ozone loss and to contribute to climate modeling.

Historically, the first ozone profile information was extracted from the Dobson measurements with the discovery of the Umkehr effect in the 1930s [Götz et al., 1934]. In optimal (blue sky) conditions at sunrise and at sunset two coarse resolution (δz ≈ 7 km) vertical ozone profiles from about 15 to 50 km could be retrieved by this technique and the first stratospheric ozone climatology created [Dobson et al., 1927]. Since this manual measurement method was demanding in personal resources it is only since the mid 1950s that continuous Umkehr measurements are available and the technique continues to be improved [Fioletov et al., 2006; Petropavlovskikh et al., 2005]. In the 1960s, wet-chemical ozonesondes were developed providing in situ high vertical resolution (δz ≈ 0.3 km or less) ozone profiles from the ground to the mid stratosphere [Brewer and Milford, 1960; Komhyr, 1965]. Datasets more than 30 years long are available based on this technique [Harriss et al., 1998; Stähelin et al., 2001; Jeannet et al., 2007]. In the 1970s, the satellite epoch began providing global coverage of the total ozone column [e.g. Labow et al., 2013]. In the 1990s, the active lidar and the passive microwave were developed with an improved time resolution and an extended altitude range up to the mesosphere [Beekman et al., 1994; Calisesi et al., 2003; Moreira et al., 2015]. Today, the full suite of ground based, balloon-borne, and satellite instruments provide significant spatio-temporal coverage of global ozone. Maintaining this coverage requires all three platforms. Satellite instruments have limited life times and require comparison measurements with other instruments for algorithm development and reference measurements. Balloon-borne instruments provide the highest vertical resolution and the highest sensitivity but are limited in spatial and temporal coverage. Ground based instruments are required for long time series with single reference instruments and for daily measurement capability [e.g. Guirlet et al., 2000]. Modeling activities ranging from weather forecasts to climate studies benefit from ozone distribution measurements from all three platforms [Stein et al., 2000, Cionni et al., 2011].

Ozone is recognized as an Essential Climate Variable (ECV) and target observation requirements for satellite based products for climate are defined by the Global Climate Observing System (GCOS), which is a joint undertaking of the World Meteorological Organization (WMO), the United Nations Environmental Program, and others [GCOS, 2010]. The measurement requirements for an ECV represent a challenge even for ground based instruments:

1. Accuracy: 10% (troposphere), 5% (stratosphere).
2. Spatial resolution: Horizontal: 5-50 km (troposphere), 50-100 km (stratosphere).
3. Vertical resolution: 0.5 km (troposphere), 0.5-3 km (stratosphere).
4. Three hourly observing cycle everywhere.
5. Stability: 1 % (troposphere), 0.6% (stratosphere).

1.1 Contributions of electrochemical concentration cell ozonesonde measurements

Since the late 1960s the vast majority of vertical ozone profile information is from individual ozonesonde flights. The instruments used are all based on measurements of an electrical current from an electrochemical galvanic cell which is a measure of the amount of ozone sampled. The current is generated when ozone in the air, which is bubbled through an electrolytic solution, reacts with iodide ions in the electrolyte in the cell. Variations of this principle, described in detail in section 2.1, led to the Brewer-Mast (BM) ozonesonde [Brewer and Milford, 1960], the Japanese KC ozonesonde [Komhyr and Harris, 1965; Kobayashi and Toyama, 1966], and the electrochemical concentration cell (ECC) ozonesonde [Komhyr, 1969]. The BM ozonesonde consists of a single electrochemical cell with a potential applied across the silver anode and platinum cathode immersed in an alkaline potassium iodide (KI) solution. The KC ozonesonde has a platinum cathode and carbon anode
imperfectly understood. The use of a KI concentration was the subject of recommendations for ECC ozone sondes in the mid 1990s, when ENSCI (EN) was formed. Initially, ENSCI recommended a KI concentration of 1.0% for the cathode; however, this was changed to 0.5% after an analysis of comparisons of EN and SP ozonesonde measurements using 1.0% KI indicated that EN ozonesondes recorded more ozone than the SP ozonesondes at the same solution concentration.

These changes created some confusion as recommendations in the preparation of ECC ozonesondes changed. The first results comparing ozonesondes flown with 1.0% and 0.5% KI cathode solution were based on only a few comparisons [Boyd et al., 1998]. More extensive results were obtained from comprehensive intercomparisons in the laboratory [Smit et al., 2007], and in the field [Kivi et al. 2007; Deshler et al., 2008]. These comparisons lead to the current WMO recommendations for ECC ozonesonde preparations [Smit and ASOPOS Panel, 2014]; however, between the mid 1990s and late 2000s the ozonesonde community was using several variations between 1.0% and 0.5% KI cathode cell concentrations in SP and EN ozonesondes. In some cases, other concentrations were also used, such as 2.0%, but we focus here on the manufacturer recommended concentrations of 1.0% and 0.5%.

1.2. Need for homogenization of ECC ozonesonde measurements

The Montreal protocol signed in 1991 has established the publication every four years of an Ozone Assessment [e.g. WMO, 2010; 2014]. One of the most comprehensive reports regarding measurement techniques was the SPARC-IOC-GAW study [WMO, 1998]. An update of this study, the SI2N (SPARC/IGACO/IOC/NDACC) initiative to report the present state of knowledge of the different techniques and to reprocess long time series accordingly is being covered in a special issue of Atmos. Chem. Phys., Atmos. Meas. Tech., and ESSD. A parallel European Space Agency-CCI (Climate Change Initiative) project was established in 2011 to improve the satellites’ products for the prominent "Essential Climate Variables", one being ozone. Comparisons with current satellite measurements of ozone, and future instrumental improvements for new satellite generations, require more accurate ground based data series for validation [Liu et al., 2006; Hubert et al., 2016]. Such comparisons have a rich heritage in previous field campaigns comparing various methods to measure ozone [Hilsenrath et al., 1986; Kerr et al., 1994; Margitan et al., 1995; Komhyr et al., 1995b; Meijer et al., 2004].

The MOZAIC datasets [e.g. Thouret et al., 1998; 2006] obtained from in-service aircraft provide a comparison to tropospheric ozonesonde measurements especially at the tropopause where ozone profiles are at their minimum values. Staufer et al. [2013; 2014] found a systematic difference with ozonesondes when aircraft measurements were compared to ozonesonde measurements determined by matching balloon and aircraft measurements via air parcel trajectory calculations, concluding an overestimation by the ozonesondes on the order of 5-10 % in the upper troposphere - lower stratosphere region. Logan et al. [2012] extensively analyzed tropospheric ozonesonde data by comparison to MOZAIC aircraft’s ascent/descent profiles and to high altitude ground based measurements, pointing out biased and suspicious datasets.

Clear improvement of ozonesonde measurement precision for properly prepared and analyzed instruments is shown in recent comparison experiments for both the EN and SP instruments [Smit et al., 2007; Deshler et al., 2008; Thompson et al., 2012]. A good sign of the stability of these results in the last ten years is confirmed in recent studies, e.g. Logan et al. [2012]. These results are not the case for the accuracy of measurements with "provider-solution" combinations which differ from the recommendations. Such combinations typically deviate from trusted ozone measurements by 5-10%.
These latter deviations are now reasonably well characterized by a large set of comparison measurements, Table 1. Thus, it is time to apply corrections to ozonesonde data measured with provider-solution combinations differing from the standard WMO recommendations. Such applications will homogenize these data sets and thereby improve data quality, usefulness for trend analysis, global homogeneity, and references for satellites and models. The appropriate corrections to apply, using the large comparison data set available, are developed in sections 3 and 4.

To homogenize these records to a single standard requires transfer functions to convert measurements made with any of the various combinations to one of the two WMO recommended standard preparations: 1.0% for SP and 0.5% for EN for the KI concentrations of the cathode electrolyte. Obtaining these transfer functions is the goal of this paper.

The transfer functions will be derived from published and unpublished measurements which compare directly the response of SP and EN ECC ozonesondes using 1.0 and 0.5% concentrations of KI in the cathode cells under identical environments of ozone, pressure, and temperature. These comparisons were done in an environmental simulation chamber in Jülich, Germany [Smit et al., 2007], henceforth JOSIE09, on a multiple ECC ozonesonde gondola [Deshler et al., 2008], henceforth the BESOS experiment, on other multiple ozonesonde balloon flights [Kivi et al., 2007], and on unpublished dual ozonesonde flights from Payner, Switzerland; McMurdo Station, Antarctica; Sodankylä, Finland; Wallops Island, Virginia, USA; and Laramie, Wyoming, USA. Together 197 comparisons of the different possible combinations have been made at these sites with the goal to develop transfer functions to convert measurements made with either manufacturer and with either 1.0 or 0.5% KI concentration to one of the WMO recommendations: SP with 1.0% KI, EN with 0.5% KI.

The basic ECC principles, operation, and uncertainties are described in the next section. Section 3 describes the data base and the methods to establish the transfer functions. Section 4 describes the methods and the results for the derivation of the transfer functions. The results are discussed in section 5 and then conclusions appear in section 6.

2. ECC ozonesonde description

2.1. ECC ozonesonde principles

Ozonesondes are based on an electrochemical cell where the chemical potential difference is maintained by differences in the iodide (I) concentration in each half cell. Ozone introduced into the dilute iodide half reacts with iodide and converts it to iodine (I2) in the following reaction [Komhyr, 1969]:

\[
\text{KI} + \text{O}_3 + \text{H}_2\text{O} \rightarrow 2\text{KOH} + \text{I}_2 + \text{O}_2. \quad (1)
\]

followed at the cathode by the reduction of iodine back to potassium iodide:

\[
\text{I}_2 + 2\text{e} \rightarrow 2\text{I}. \quad (2)
\]

The two electrons arise from the electrolyte saturated in iodide at the anode by the oxidation of iodide:

\[
3\text{I} \rightarrow \text{I}_3^- + 2\text{e}. \quad (3)
\]

The outer electrical circuit consists of two platinum electrodes immersed in the half cell electrolytes which are aqueous solutions of KI. The electrolyte is saturated with KI on the anode side and with a dilute KI concentration on the cathode side. Besides KI, the electrolytes also contain potassium bromine and sodium phosphate buffers to maintain a neutral pH solution. The decomposition of each ozone molecule in the dilute electrolyte produces a transfer of electrons (Eq. 2) in the outer circuit following Faraday’s law of electrolysis. Converting the mass of ozone decomposed to partial pressure with the ideal gas law and substituting \(\text{e}\cdot\text{N}_A\) for Faraday’s constant results in the following relationship between ozone partial pressure and electrical current:

\[
O_3(p) = \frac{R}{(z\cdot e\cdot N_A)} \cdot \frac{T_F(p)}{P_E(p)} \cdot \frac{T_F(p)}{FR} \cdot \left(\phi(p) - i_0\right). \quad (4)
\]
$R$ is the universal gas constant, $z$ the number of electrons required to convert the iodine in the dilute electrolyte back to iodide, $e$ the elementary charge, $N_A$ Avogadro’s number, $i(p)$ the measured cell current as a function of atmospheric pressure, $p$, $i_0$ the background current, $\varphi(p)$ the effective stoichiometry factor of the chemical conversion of ozone into iodine, $T_{\text{p}}(p)$ the pump temperature, a surrogate for the temperature of the air sampled, $FR$ the flow rate, $P_{\text{eff}}(p)$ the pump efficiency correction to account for decreasing flow rate at low pressures. Operationally, for $i(p)$ in units of $\mu$A, $z = 2$, and $FR$ in units of $\text{ml s}^{-1}$, the leading term in Eq. 4, $R(z \times N_A)$ is replaced by $4.3085 \times 10^6$ which gives ozone partial pressure in mPa.

Each of the terms in Eq. 4 has an uncertainty deduced from the measurement method but they also have an additional uncertainty which is more difficult to quantify, especially at high altitude (low pressure) levels. The background current, $i_0$, the flowrate, $FR$, the pump temperature, $T_{\text{p}}(p)$, and the stoichiometry of ozone to iodine, $\varphi(p)$, require special mention.

The background current $i_0$ is a measure of the residual signal with "zero-air" (no ozone) at the ozonesonde inlet. Originally, $i_0$ was attributed to side chemical reactions with oxygen and therefore, expected to decrease with altitude; however, Thornton and Niazy [1982] and Thornton [1983] and other empirical evidence from laboratory tests, suggested that $i_0$ is constant independent of altitude. Figure 1 gives the range of $i_0$ measured at the different stations prior to the measurement flights considered in this analysis. A mid value of 0.03 $\mu$A from Figure 1 corresponds to 0.1 mPa and produces an offset of the ozone profile and a diminution of the ozone column by 5.4 DU, 0.1 mPa integrated from 1000 hPa to 1 hPa, or 1.8% for a 300 DU ozone column. These $i_0$ values will be discussed again in section 3.2, but the value of $i_0$ does not affect comparisons between cells with similar backgrounds, and is thus less important for the work here.

The flow rate $FR$ is well characterized in the laboratory preparation at surface pressure as shown in the box and whisker plot of the flow rates, Figure 2, measured at the different stations. The median values for each ozonesonde type are within 0.3 ml s$^{-1}$, $< 10\%$, showing the good concordance between stations, although there is a 0.2 ml s$^{-1}$ systematic difference between the majority of SP and EN ozonesondes. The real flow rate is much less certain at low pressure (high altitude) conditions where the pump efficiency decreases and pressure against the flow from the height of the solution in the ECC cell, on the order of 1-2 hPa, becomes non-negligible. Thus a pump efficiency correction is applied through the factor $P_{\text{eff}}(p)$; however, there is still disagreement on the correct $PE(p)$ to use [Johnson et al., 2002]. The pumps are designed for a constant rotation speed during a flight, a characteristic which has been checked by several investigators here but not published.

The effective stoichiometric factor, $\varphi(p)$ as formulated in Eq. 4, is composed of two factors: the absorption of ozone in the electrolytic solution, and the stoichiometric efficiency of the reaction of O$_3$ and KI to produce I$_2$. This latter factor is more difficult to characterize. In theory it is close to 1. The absorption efficiency has been measured in the laboratory with two ECC ozonesondes measuring in series. These tests have shown that the absorption of ozone is somewhat dependent on the amount of solution used (e.g. 2.5 ml vs. 3.0 ml) [Tarasick et al., 2016; Davies et al., 2003], with the absorption increasing from 0.96 as the amount of solution increases from 2.5 ml. The increase of the gas diffusion rate during balloon ascent due to the pressure decrease should limit this ozone conversion efficiency loss to the lower part of the profile [Davies et al., 2003].

All these factors are eventually time and pressure dependent and they superpose each other which makes their individual contribution particularly difficult to determine. The factors which are of particular concern for the issue of varying ozonesonde manufacturer and cathode solution concentration are contained in the terms $z$ and $\varphi(p)$.

The details of the differences in ozonesonde manufacturer and how they affect these terms can only be speculated on at this time. The SP and EN ozonesondes are in principle the same, yet the EN ozonesondes consistently indicate a higher ozone amount when compared to an SP ozonesonde with the same sensor KI electrolyte concentration. The differences in cell manufacturer may affect the efficiency of electron release at the anode and electron gain by iodine at the cathode, which could be related to differences in the platinum electrode sensitivity and the ion bridge conductance. The inner surface properties of the cell walls are different and may affect reactions of ozone and the cell walls. Either of these would affect $z$. 
2.2. Review of the standard WMO recommendations

The preparation before the flight of an ECC ozonesonde is crucial for its performance. The Standard Operating Procedures (SOPs) for ECC ozonesondes have been established by a group of experts under the guidance of the World Calibration Center for Ozone sondes (WCCOS) of the WMO [Smit and the ASOPOS panel, 2014]. These results are from a ten year coordinated process to improve the different aspects of ECC ozonesonde preparation and data processing. Operating the ECC ozonesondes under these prescribed rules has been tested extensively in different JOSIE experiments at the Jülich Research Center [Smit et al., 2007]. The large balloon experiment BESOS was designed as an extension in the real atmosphere of these laboratory developments [Deshler et al., 2008]. In addition to 18 ozonesondes, the BESOS gondola included also the Jülich reference UV spectrometer [Proffitt et al., 1983, Smit et al., 2007] to replicate as far as possible the test procedure of the JOSIE experiments. The two experiments agree on the following conclusions:

1. ECC ozonesondes prepared according to the SOPs provide very reproducible (<2-3%) measurements.
2. The results depend on the ozonesonde manufacturer (e.g. EN vs. SP) and on the sensing solution concentration (e.g. 0.5% vs. 1.0%). The order of magnitude of the differences is 5%.
3. The combinations "EN-0.5%" and "SP-1.0%" as "provider-solution" agree with each other to within 2%, but overestimate the reference UV photometer in the stratosphere by 5-10%.
4. The total ozone column estimated from these combinations on the BESOS gondola agreed with a collocated Dobson spectrophotometer.
5. It is possible to reconcile the measurements made with other "provider-solution" combinations and the photometer with the help of a simple linear in ln (p) transfer function.

JOSIE and BESOS were completed under conditions not reflecting directly the diversity of the operational services around the world. Each sounding station has specific instrumentation and operators even though they follow the same procedures. It is therefore important to verify that the conclusions from JOSIE and BESOS are also reflected in results of several operational stations. At the stations, it is not possible to fly a reference UV instrument so only relative differences can be derived from dual or multiple ozonesonde flights. In the present analysis the JOSIE and BESOS results will be analyzed in a similar way as the other dual multi instrument flights.

2.3. Review of previous solution concentration and provider comparisons

In the mid 1990s the problems of differences in "provider-solution" combinations were still of marginal importance [SPARC, 1998]. The EN-SCI company had entered the market only a few years earlier and the disparate preparation procedures prevented clearly identifying problems. The conclusions at that time were that the effect of changes in ECC KI solution concentrations were complex and required further study before clear recommendations could be provided.

McPeters et al. [1999] reports a 2% consistency from five triple ECC flights during a validation campaign at Mauna Loa in 1995 using EN 1.0% ozonesondes. The authors report that the ozonesondes overestimate the Dobson measurements by an average of 5%. In profile, above 25 km, the ozonesonde measurements are greater than the lidar and microwave measurements by a similar amount. Boyd et al. [1998] presented ozone profile differences from EN ozonesondes with 1.0 and 0.5% KI solutions at Lauder, New Zealand. A 5-6% systematic overestimation of ozone by the 1% solution compared to the 0.5% with the EN ozonesondes is evidenced by comparison of the ozone profile and total column collocated lidar and Dobson measurements.

For an analysis of the transition from Brewer Mast to ECC (EN - 1.0%) ozonesondes, the ECC data were normalized to the Dobson column to be consistent with the Brewer Mast SOPs [Stübi et al.,
2008]. Stübi et al. found that the ECC ozonesondes systematically overestimated the total ozone column with a mean normalization factor of 0.95 for more than 100 dual flights between ECC and BM indicating an overestimation of 5% of the ozone column by EN-1.0 ozonesondes.

Kivi et al. [2007] analyzed a series of dual and multiple ozonesonde flights with SP and EN instruments using 0.5% and 1.0% sensing solutions. For the homogenization of ozone profiles from the Northern high latitude stations the authors derived a third order polynomial correction based on altitude to correct the overestimation of ozone from EN-1.0% compared to SP-1.0%.

The laboratory work (Smit et al. [2007] along with the several field measurements (Boyd et al. [1998]; McPeters et al. [1999]; Stübi et al. [2008]; Kivi et al. [2007]; Deshler et al. [2008] all indicate a relatively consistent systematic bias, on the order of 5%, between the different ozonesonde manufacturers with the same electrolytic concentration, and between different electrolyte concentrations in ozonesondes from a single manufacturer.

### 2.4. Chemistry of the ECC ozonesonde

The early stoichiometric work on the yield of iodine from ozone showed varying results with much of the uncertainty arising from the variety of KI sensing solutions, pH buffers, and sensors used [Saltzman and Gilbert, 1959; Boyd et al., 1970; Dietz et al., 1973; Pitts et al., 1976; Lanting, 1979]. Common to many of the references was the suggestion of a secondary reaction producing additional iodine perhaps from reactions of iodide with the phosphate buffers. Johnson et al. [2002] showed that the same type of ECC ozonesonde operated with differing amounts of KI, and corresponding changes in the phosphate buffers, provide slightly different stoichiometric ratios of iodine to ozone. In fact these differences were very apparent in the initial development of the ECC ozonesondes [Komhyr, 1969; 1986].

### 2.5. Need for homogenization for data comparison

The Montreal protocol signed in 1991 has established the publication every four years of an Ozone Assessment [e.g., WMO, 2010; 2011]. One of the most comprehensive reports regarding measurement techniques was the SPARC-IOC-GAW study [WMO, 1998]. An update of this study, the SI2N (SPARC/IGACO/IOC/NDACC) initiative to report the present state of knowledge of the different techniques and to reprocess long-time series accordingly is being covered in a special issue of Atmos. Chem. Phys., Atmos. Meas. Tech., and ESSD. A parallel European Space Agency-CCI (Climate Change Initiative) project was established in 2011 to improve the satellites’ products for the prominent “Essential Climate Variables”, one being ozone. Comparisons with current satellite measurements of ozone, and future instrumental improvements for new satellite generations, require more accurate ground based data series for validation [Liu et al., 2006; Hubert et al., 2016]. Such comparisons have a rich heritage in previous field campaigns comparing various methods to measure ozone [Hilsenrath et al., 1986; Kerr et al., 1994; Margitan et al., 1995; Komhyr et al., 1995b; Meijer et al., 2004].

The MOZAIC datasets [e.g. Thouret et al., 1998; 2006] obtained from in-service aircraft provide a comparison to tropospheric ozonesonde measurements especially at the tropopause where ozone profiles are at their minimum values. Stauffer et al. [2013; 2014] found a systematic difference with ozonesondes when aircraft measurements were compared to ozonesonde measurements determined by matching balloon and aircraft measurements via air parcel trajectory calculations, concluding an overestimation by the ozonesondes on the order of 5-10% in the upper troposphere – lower stratosphere region. Logan et al. [2012] extensively analyzed tropospheric ozonesonde data by comparison to MOZAIC aircraft’s ascent/descent profiles and to high altitude ground based measurements, pointing out biased and suspicious datasets.

Clear improvement of ozonesonde measurement precision for properly prepared and analyzed instruments is shown in recent comparison experiments for both the EN and SP instruments [Smit et al., 2007; Deshler et al., 2008; Thompson et al., 2012]. A good sign of the stability of these results in the last ten years is confirmed in recent studies, e.g. Logan et al. [2012]. These results are not the case for the accuracy of measurements with “provider solution” combinations which differ from the recommendations. Such combinations typically deviate from trusted ozone measurements by 5-10%.

These latter deviations are now reasonably well characterized by a large set of comparison measurements, Table 1. Thus, it is time to apply corrections to ozone data measured with
3. Methods to establish the transfer functions

3.1. Summary of datasets considered in the present analysis.

The present analysis of dual ozonesonde measurements is an extension of the JOSIE and BESOS experiments to link short term instrument comparison campaigns to routine operations at regular soundings. The JOSIE experiments were conducted in a controlled atmospheric chamber. BESOS and the other comparisons were conducted via balloon flights under real atmospheric conditions. JOSIE and BESOS used the same reference UV photometer [Proffitt et al., 1983] for the final comparisons and the results of those comparisons confirmed the high precisions and good accuracy of well-prepared ozonesondes. For the extensive additional data presented here an independent (e.g. photometric) reference is not available, rather the ozonesondes are compared pairwise. The JOSIE and BESOS data are included here also pairwise, Table 2. The first of these dual ozonesonde comparisons began in the late 1990s at different locations. Although there was no coordinated effort, the motivation at each station was similar. The need for homogenization of the long term ozonesonde record at the station. Table 1 summarizes the datasets used for the present analysis. Differences in the details of these comparisons at the different stations are described below.

While the differences are described below the comparisons were the same in following the SOPs established by Smit and the ASOPOS panel [2014] in the preparation of each ozonesonde, and in using only 0.5% of 1.0% KI buffered cathode solutions. The 0.5% KI cathode solutions were created by diluting the standard buffered 1.0% KI solution by 50% with distilled deionized water. The standard 1.0% KI solution is created by dissolving the following in distilled deionized water: 10 g KI, 25 g KBr, 1.25 g NaHPO₄·2H₂O, and 5 g Na₂HPO₄·12H₂O, and then adding enough distilled water to make 1 liter of solution.

3.1.1 JOSIE09 – Ozone profile simulation chamber, Jülich, Germany

The JOSIE experiments have been described by Smit and Kley [1998], Smit and Sträter [2004a, b] and Smit et al. [2007] so only the experimental principles are reviewed here. Four ozonesondes can be placed simultaneously in the atmospheric simulator. Pressure and temperature can be regulated from surface conditions to 10 hPa and -70°C. The ozone flow is controlled in a glass cavity and measured in parallel by the ozonesondes and a reference UV photometer. Different types of "temperature-pressure-ozone" profiles are generated to simulate high-, middle- or tropical-latitude profiles. In the present analysis, only pairs of ozonesondes, representing different provider-solution combinations, simultaneously operated in the chamber are considered. This explains the low number of comparisons available for these data.

3.1.2. BESOS – Balloon-borne multi instrument gondola, Laramie, Wyoming

The BESOS experiment was described fully by Deshler et al. [2008]. A collaborative team of ozonesonde experts prepared a balloon gondola (100 kg) with 16 ozonesondes, the Jülich UV photometer [Proffitt et al., 1983], a Vaisala radiosonde, and a data acquisition system. Dobson and Brewer spectrophotometers were available at the launch site. The flight to 32 km was completed on 13 April 2004 from Laramie, Wyoming. The data from this flight are used here similarly to the JOSIE data by considering the ozonesondes pairwise. The payload had a set of 12 standard ozonesondes, 6 from EN and 6 from SP; out of each set of 6 ozonesondes, 3 had a 0.5% and 3 a 1.0% KI solution concentration. Thus, a set of nine pairs are available for comparison for each "provider-solution" combination, Table 2. The correlation matrix for any one of the 3 pairs of a comparison indicated little variation in the correlation coefficient for any combination of the two types of ozonesondes being compared. This suggests that covariance does not negate the independence of the 9 comparisons for any combination of ozonesonde type or solution concentration.

3.1.3. Payerne, Switzerland – Balloon-borne dual ozonesonde gondolas
The Payerné station is run under the responsibility of MeteoSwiss and the radiosondes used were the SRS model from the Swiss company Meteolabor. SRS radiosondes are not capable of interfacing two sondes, so for the dual flights two independent receiving systems were used. These were synchronized at the time of the launch to better than one second and the sampling frequency was about 7 seconds. For the analysis, the data are interpolated to a common time scale to avoid any problems related to a difference of the pressure readings from the two sondes. The datasets consist in two campaigns embedded in the operational service as a dual flight for the Wednesday sounding. The reference (operational) ozonesonde being the EN-0.5%. The data sets consist of:

1) 48 pairs EN-0.5% and EN-1.0% - June 2002 - July 2003.
2) 26 pairs EN 0.5% and SP 1.0% - May 2005 - December 2006.

3.1.4. McMurdo Station, Antarctica – Balloon-borne dual ozonesonde gondolas

Measurements from McMurdo Station, Antarctica, were conducted by the University of Wyoming during the ozone hole period, August –November, 1986-2010 [Mercer et al., 2007]. From this record 18 flights with two EN ozonesondes interfaced to a single microprocessor and Vaisala RS80 radiosonde were completed. The years (number of flights) are: 1996 (3), 1999 (1), 2000 (1), 2002 (6) and 2006 (7). In each case EN-1.0% and EN-0.5 % KI concentration solutions were compared. The low temperature conditions in Antarctica require a heater near the cells to prevent the solution freezing at high altitude. This preventive action is visible on Figure 3 with a mean pump temperature which stays close to 24°C at high altitude. The background currents are low with a slightly higher value for the ozone sondes with 1.0% solution compared to the 0.5%, Figure 1. This dataset is characterized by a large variety of ozone profiles from typical ozone hole to more conventional polar conditions; ozone column ranges are 126 - 423 DU.

3.1.5.  Sodankylä, Finland – Balloon-borne dual and multi ozonesonde gondolas

The Sodankylä station is run by the Finnish Meteorological Institute. The radiosondes used for the dual sonde and multiple sonde measurements were the Vaisala RS80. In the dataset used here there is a mix of 5 dual flights and 4 larger balloon flights with “6-sonde” payloads. The larger balloon payloads were recovered and flown again the next day with reused ozonesondes. The mean pump temperature profile shown in Figure 3 is characterized by the leveling of pump temperature at about 22°C due to the use of a heater in case of the comparisons made under cold stratospheric conditions.

For both the multiple and dual ozonesonde payloads a single RS80 radiosonde was applied per two ozonesondes, using interface extension boards provided by EN-SCI. Thus one receiving system was involved per two ozonesondes. The data set consists of

1) 6 pairs EN 0.5% and EN 1.0% - September 2003 - July 2004.
2) 5 pairs EN 0.5% and SP 1.0% - May 2003 - March 2005.
3) 8 pairs EN 1.0% and SP 1.0% - September 2003 - July 2004.

3.1.6.  Wallops Island, Virginia - Balloon-borne dual ozonesonde gondolas

Resources for ozonesonde measurements, with Sippican radiosondes, from Wallops Island have been, and continue to be, provided by NASA Headquarters. The Wallops Island practice is to use the background current measured during the day-of-flight preparation prior to exposing the ECC to moderate ozone (5μA) for 5 minutes; these backgrounds are smaller than the others shown in Figure 1. The negative backgrounds are unusual, but are included as those were what was reported in the period 1995-1999 when Wallops Island was beginning its ozone measurements. The values of i are, however, close to each other within the comparison pairs so this difference, even if negative, has a negligible effect on the comparison measurements analysis which were coordinated by matching the elapsed times of flight of the two comparison systems, similar to the procedure for Payerné. The data sets consist of:

1) 7 pairs SP 5A-ECC’s 0.5% vs. 1.0% in 1996.
2) 11 pairs SP 6A-ECC’s 0.5% vs. 1.0% in 2004.

3.1.7 Laramie, Wyoming – Balloon-borne multi ozonesonde gondola

These results were obtained from a collaboration between the Climate Monitoring and Diagnostic Laboratory (CMDL) of the National Oceanic and Atmospheric Administration and the University of Wyoming. CMDL prepared the gondola and the University of Wyoming conducted the flight
operation. The measurements were obtained from a gondola containing 6 EN ozonesondes, 3 with
0.5% KI and 3 with 1.0% KI. The instruments were synchronized to a common data system and an
RS80 Vaisala radiosonde. The flight occurred on 20 June 1996 and reached an altitude of 32 km. Post
data analysis eliminated one of the 0.5% KI EN ozonesondes from the comparison. Results from this
sonde were significantly different from the other two 0.5% KI ozonesondes, which agreed quite
well.

3.2. Data Processing
The ozone data processing from the measured current is based on Eq. 4 with little variability
among the datasets. The major difference is in the sampling frequency of the measurements which
ranged from 0.2 to 1.0 Hz. The typical e-folding response of an ozonesonde is of the order of 0.05 Hz
[Smit and Kley, 1998]. All the sampling rates here are faster than this, but are the same for every
pairwise comparison so the sampling rate will not affect a comparison. However, since the data
analysis is based on the individual pair differences, it is necessary to average the high frequency
measurements to a common time scale to avoid unduly weighting the high frequency data relative to
the lower frequency measurements. Ultimately the high frequency data were averaged to a frequency
of 0.2 Hz so that when the data weighted means of the comparisons were calculated each comparison
profile was weighted about equally.

3.2.1. Background current $i_0$
In Figure 1, the background currents measured for the different data sets are summarized as box
and whisker plots. For all sites except Wallops Island, these are the background currents after
exposure to moderate ozone and just prior to flight. The $i_0$ used at Wallops Island was prior to
exposure to moderate ozone on the day of flight preparations. This may explain the slightly lower
backgrounds obtained at Wallops Island. The medians of $i_0$ from the various sites are all below 0.03
$\mu$A indicating the quality and consistency of the ozonesonde preparations. The $i_0$ values for the 1.0%
solution tend to be slightly higher compared to the 0.5% solution suggesting the impact of the larger
buffer amount [Johnson et al., 2002]. Boyd et al. [1998] argue that the large difference between their
ascent and descent ozone (tropospheric) profiles is attributable to an increase in the background
current after exposure to high ozone in the stratosphere. They have observed this increase with the 1%
solution but not with the 0.5%. Similarly in the laboratory preparation, the ozonesondes are exposed to
high ozone for ten minutes and the slightly higher $i_0$ values for the 1.0% solution could be related to
such a memory effect. The origin of the background current is still poorly understood [Smit et al.,
2007]. Vömel and Diaz [2010] measured the rate at which ozonesondes approach background in the
laboratory, with some implications for measurements in the tropics of very low tropospheric ozone
concentrations.

3.2.2. Pump flow rate
The pump flow rate is the second parameter measured in the laboratory preparation of each
ozonesonde. Figure 2 shows the coherency of the pump flow rates at the 5 field measurements sites. In
about half the measurement sets, the inner quartiles of the variations amongst the instruments
measured are less than 3% of the median, and in all cases except one the inner quartiles are less than
6% of the median. The figure also shows a systematically 0.2-0.3 ml s$^{-1}$ higher flow rate for SP pumps
compared to EN pumps.

3.2.3. Pump efficiency correction
The application of pump efficiency corrections vary amongst the datasets. In general Komhyr
[1986] is used for SP and Komhyr et al. [1995a] is used for EN ozonesondes. Since the comparisons
analyzed are amongst pairs of identical ozonesondes, the pump efficiency does not play an important
factor unless significantly different pump efficiencies were applied separately to the ozonesondes in a
measurement pair. In most cases the same efficiency factors were applied to both ozonesondes of a
pair. The one exception is the Wallops Island data, where individual pump efficiency curves were
applied prior to mid 2000 when the system failed. The pairwise comparisons of these data, however,
were quite similar to the Wallops Island data where identical pump efficiencies were used, and to the pairwise comparisons from the other data sets.

### 3.2.4. Pump temperature

For the data processing, individual pump temperatures are used as illustrated in Figure 3 for the mean pump temperature profiles for each dataset. It should be noted that the SP 5A ozonesondes used at Wallops Island did not have an explicit hole to insert the pump temperature sensor. The standard deviations of the temperature range from 1°C for McMurdo Station to 5°C for Sodankylä. The pump temperature decrease over a profile is around 7–10°C for the ozonesondes with a heater and 20–23°C for the ozonesondes without a heater. This parameter also is reproducible within the ozonesondes pairs and thus doesn’t impact significantly the pairwise comparisons.

### 4. Comparisons of ozone partial pressure

In Figure 4, an example of a dual flight from Payerne is illustrated. The two ozonesondes separated by a 1.5 m long boom were hanging under the same balloon and the data transmitted to two independent receiving systems on the ground. The ozone profiles have identical structures and differences increase near the ozone maximum at pressures less than 50 hPa, indicating some dependence on both ozone partial pressure and atmospheric pressure. The increased sensitivity of the 1.0% solution is clear throughout the profile.

#### 4.1. Differences between 1.0% and 0.5% KI cathode solutions for EN and SP ozonesondes

The simplest way to analyze the data is to compare ozone partial pressures measured by ozonesonde pairs operated simultaneously, either in the atmosphere or in the simulation chamber. Scatter-plots of ozone partial pressure measured with ECC ozonesondes with 1.0% solution, x-axis, against simultaneous measurements with a 0.5% solution, y-axis, are shown in Figure 5 for EN ozonesondes flown from a) Payerne and b) McMurdo Station, and SP ozonesondes from c) BESOS and d) JOSIE09. The color coding distinguishes four pressure ranges to highlight the altitude (pressure) dependence. Only ozone partial pressures > 0.5 mPa have been considered to remove large differences resulting from comparisons of small numbers near the measurement limit of the ozonesondes. Figure 5 demonstrates a near linear relationship between the 0.5 and 1.0% ozonesonde measurements in the four pressure ranges considered, independent of the ozonesonde manufacturer. The mean and standard deviation of the measurement ratios in the various pressure intervals are given in the legends. The mean ratios are used to construct linear fits, which pass through the origin, to the measurements in each pressure range and are displayed in the Figure. The mean ratios and standard deviations for all the measurements from JOSIE09, BESOS, Payerne, and McMurdo Station at the four pressure intervals are given in Table 2. Table 2 also contains bulk fits to all data from Sodankylä, Wallops Island, and Laramie without differentiation according to pressure. Figure 5 and Table 2, upper two boxes 1 and 2, display the remarkable consistency amongst all the data from the varied sources. Note the consistency of the mean ratios and their standard deviations from all data sources. Figure 5 and Table 2 also indicate a consistent 3-4% underestimation of ozone from a 0.5% KI solution compared to a 1.0% solution from the ground up to 30 hPa for both SP and EN ozonesondes. An increase of the difference to 6-8% at pressures below 30 hPa is also shown by the four data sets, Figure 5.

Table 2 also shows additional analysis in three other comparison groups: the third and fourth boxes 3 and 4 correspond to a change of provider keeping the same concentration and the final box, 5, a fit to a mix of SP 1.0% and EN 0.5%. The tendency of a decrease of the linear term at lower pressures is present in most data sets except for this last group where the linear fit is not statistically different than the fits at pressures above 30 hPa. The correlation coefficients for the data are all above 0.998. There are four cases in Table 2, two in the Payerne, box 1 and 5, and two in the BESOS, box 3 and 5, datasets, with standard deviations > 0.1. These are all in the column for pressures > 500 hPa. The origin of the large standard deviations for Payerne, EN-1.0 vs EN-0.5, probably lies in the outliers apparent in Figure 5a) at pressures > 500 hPa. Such discrepancies are less obvious in the other Payerne comparison and in the BESOS data. The cause of these larger standard deviations was not investigated further in light of small standard deviations in all datasets at pressures less than 500 hPa.
Considering the strong linear relationship of the dual measurements for the differences in concentration in the same ozonesonde type, and differences in ozonesonde type with the same sensor concentration, it is natural to simply use a single ratio to characterize the relationship of the two measurements at pressures above a certain threshold pressure, and then to use a linear relationship in $\log_{10}(p)$ to fit the ratio at lower pressures, insuring that the two systems merge at the threshold pressure. The ratio of the measurements from a single manufacturer at two cathode concentrations is illustrated in Figure 6 as an ensemble of red dots for the same data sets as in Figure 5. The dual flight measurements at Payerne and McMurdo Station, Figure 6a), 6b), show a larger spread of the data but the number of measurements are considerably larger and the atmospheric conditions more diverse than in the BESOS and JOSIE experiments, Figure 6c), 6d). Occasionally, individual flights from Payerne and McMurdo Station deviate from the majority of comparisons, seen as a set of points separated from the main cloud of points. These few comparisons are clear outliers compared to the majority of measurements, but have not been removed from the analysis. The only limit placed on the data were to insure that each ozonesonde in a comparison pair measured ozone above 0.5 mPa.

These comparisons suggest that measurements from ozonesondes using a 1.0% KI concentration in the cathode can be used to derive measurements which would have been obtained from measurements with a 0.5% KI solution. To do this, if the-measurements using the 1.0% KI solution are modified using a pressure independent ratio at pressures above some threshold pressure and a pressure dependent ratio below the threshold pressure. Different values for the threshold pressure to switch from a single ratio to a pressure dependent ratio were tested but the results were not very sensitive to this value and it has been fixed at 30 hPa.

With the threshold pressure level established each data set was used to calculate a mean concentration ratio at pressures, $p \geq 30$ hPa and a linear, in $\log_{10}(p)$, fit at $p < 30$ hPa, for $p$ in units of hPa. The results of this analysis are displayed in the upper two boxes of Table 3 for all datasets listed in Table 1. The second column provides the concentration ratio and its standard deviation for $p \geq 30$ hPa, the third column the number of individual ozonesonde to ozonesonde comparisons (N). Recall each dataset was standardized to a sampling frequency of 0.2 Hz to balance the weights of the high frequency and low frequency data equally. Thus the number of data points represents primarily the number of individual ozonesonde to ozonesonde comparisons within each dataset. For $p < 30$ hPa columns 4, 5, and 6 list the fitting parameters providing the slope in $\log_{10}(p)$ and then the offset, corresponding to the value of the ozone concentration ratio at 1 hPa. Two offsets are listed. The first is the one used. The second offset is derived without a requirement to match the $p \geq 30$ hPa transfer function. The small differences between these two offsets reflects the fact that the function chosen is doing a good job of well in representing the data even without a fitting constraint. Column 7 provides the number of data points (N) at $p < 30$ hPa, column 8 the number of dual ozonesonde measurements.

The coefficients for the transfer function representing the ratio of ozone sensed at the differing concentrations were calculated as a weighted mean and standard deviation (according to sample size) of the individual parameters given in Table 3 for all datasets considered in the analysis. These values comprise the final row in each box in Table 3. Not all data from each dataset were used due to unstable ratios at particularly low ozone concentrations, or during clearly deficient ozonesonde performance. The primary examples-regions where some of the data were excluded are displayed as dark gray areas in Figure 6. These Data in these regions were excluded for the following specific reasons:

1. McMurd Station: Some of the dual measurements were completed in ozone hole conditions, and in these cases ozone drops to near zero producing highly divergent ratios,
2. JOSIE: At three points during the simulated profiles, the ozone flow was stopped to measure the residual signal and the response time, producing very low ozone and thus likewise ratio divergences.
3. BESOS: in the first minutes of the flight, the data acquisition unit was unstable and too noisy to consider in the present analysis.

The common transfer function to analyze differences in KI concentration, $OZ\text{conc}$, is given in Eq. 5 and presented in Figure 6 as a blue line for $p \geq 30$ hPa and a black line for $p < 30$ hPa. The function displayed in Eq. 5 accounts for a change of solution concentration independently of the ozonesonde provider.
\[
\frac{OZ_{0.5\%}(p)}{OZ_{1.0\%}(p)} = 0.96, \quad \text{for } p \geq 30 \text{ hPa}, \quad (5)
\]
\[
\frac{OZ_{0.5\%}(p)}{OZ_{1.0\%}(p)} = 0.90 + 0.041 \log_{10}(p), \quad \text{for } p < 30 \text{ hPa}.
\]

4.2. Difference between EN and SP with the same solution concentration

In Figure 7, profiles of the ratio of SP and EN ozonesondes with the same KI solution concentration are shown in the same format as Figure 6. The upper panels show the difference between the SP and EN ozonesondes with a 1.0% solution concentration while the lower panels are for the 0.5% solution concentration. Figure 7a) is from multiple dual flights at Sodankylä during the period 1995-2002 while the other panels present the analysis of the JOSIE and BESOS experiments. Using these data, and following the procedure used to reconcile the two solution concentrations in the same ozonesonde provider, section 4.1, the common transfer function to correct a change from one provider to the other was derived. Similar to the analysis in section 4.1 the results from fits to each data set and their weighted mean are provided in the third and fourth box in Table 3. Combining the results from the boxes comparing EN and SP ozonesondes at 1.0% and EN and SP ozonesondes at 0.5% results in the transfer function, \(OZ_{prov}\), given in Eq. 6.

\[
\frac{OZ_{SP}(p)}{OZ_{EN}(p)} = 0.96, \quad \text{for } p \geq 30 \text{ hPa} \quad (6)
\]
\[
\frac{OZ_{SP}(p)}{OZ_{EN}(p)} = 0.764 + 0.133 \log_{10}(p), \quad \text{for } p < 30 \text{ hPa}
\]

As for \(OZ_{concs}\), the coefficients of the transfer function \(OZ_{prov}\) were calculated individually for the five data sets and an overall mean, weighted by the number of comparisons and flights, calculated. The JOSIE data present a larger spread than in the previous case and individual simulator runs are visible. Aside from the BESOS data for 0.5% KI at \(p > 150 \text{ hPa}\), Figure 7d), where there is a tropospheric bias of 2%, the \(OZ_{prov}\) curve reproduces the results from the different data sets. The \(\log_{10}(p)\) coefficient is slightly larger in \(OZ_{prov}\) than in \(OZ_{concs}\) producing a lower value for the constant term (intercept at 1 hPa) since the constant ratio terms are the same (ratio\(OZ_{concs}\) = ratio\(OZ_{prov}\) = 0.96) for the \(p \geq 30\text{hPa}\) domain, while the decrease in the ozone ratio between the providers increases at lower pressures comparable to the decrease in ozone ratio at differing solution concentrations, Figure 6.

Also included in the last row in each box in Table 3 is the weighted standard deviation of the comparison ratios. These provide an indication of the uncertainty in the ratios, and thus in the corresponding transfer functions, ranging from 0.037 – 0.084. To establish a single uncertainty which could be applied to all the transfer functions, the individual comparison standard deviations were averaged and found to be near 0.05. Thus 0.05 was adopted as the single uncertainty which could be applied to all the transfer functions proposed. This value is used to apply the error bars shown in Figures 6 and 7 and shows how well the transfer functions, including the uncertainty represent the data. Clearly 0.05 both under and over estimates the ratios, depending on the particular comparison; however, overall does a reasonable job.

4.3. Similarity between the combinations EN-0.5% and SP-1.0%

With the similarity of the two transfer functions \(OZ_{concs}\) and \(OZ_{prov}\), it is natural to counterbalance them and compare EN ozonesondes with 0.5% solution and SP ozonesondes with 1.0% solution. The results are given in Figure 8 for a) Payerne, b) Sodankylä, c) BESOS, and d) JOSIE09. The horizontal axis of Figure 8 is expanded compared to Figures 6 and 7 and the number of comparisons are low for JOSIE (3 cases) and Sodankylä (5 cases). The agreement between the ozonesonde pairs is not as clear.
as for the previous cases as illustrated by the Payerne data which present a somewhat larger dispersion (10%) in Figure 8 compared to Figure 5 (5%). The BESOS data have a distinct behavior below and above the tropopause (200 hPa) while the five Sodankylä flights show a constant 3% overestimations by the SP ozonesondes. However, if no simple transfer function would allow to reconcile these observations, it is noticeable that the majority of the points are within ±5% around 1.0 illustrated by the gray zone in Figure 8. Table 3 last 3 lines also reveal no marked departure from unity. The gray area of ±5% is the same uncertainty as used for the transfer functions and some of the comparisons shown in Figure 8 are better than, but not all.

The present conclusion is that the interchange between the EN-0.5% and SP-1.0% combinations would not have a negative impact on the continuity of a time series. It may increase the variability, but no noticeable break should appear at the transition between these two systems.

### 4.4. Transfer function application on the Nairobi data set

The Kenyan Meteorological Department (KMD), in collaboration with MeteoSwiss, operates the Nairobi aerological station, within the SHADOZ (Southern Hemisphere ADditional OZone station) network, [Thompson et al., 2012]. Weekly ozone soundings began in 1996. In summer 2010, due to interruption of the Vaisala RS80 radiosonde production, new equipment based on the RS92 was installed at Nairobi. Coincidently, the ozonesonde solution concentration was changed from 1.0% to 0.5%, keeping the same ozonesonde provider, EN. This data set is used here to illustrate the application of the transfer function $OZ_{con}$ defined in Eq. 5. In Figure 9, a time series on three pressure levels is illustrated. Color separates the measurements with the different solution concentrations. In the troposphere, the ozone partial pressure is low (2.5 mPa) and the variability is too high to detect changes of a few percent. Therefore the 500 hPa data illustrated in Figure 9 do not show the change of sensor concentration. At 30 hPa, the quasi biennial oscillation is the dominant signal and this requires at least a complete cycle after the change to correctly see the effect; however, there is a clear decrease of the mean ozone value before and after the change of concentration. Finally at 10 hPa, the lower variability and the absence of geophysical cycles in the data allow the effect of the concentration change to be clearly seen.

To quantify the concentration change, the mean ozone profiles before and after 2010 have been calculated and appear in Figure 10, with black squares for 1996 to 2010 and blue circles after 2010. Red triangles correspond to the 1996 - 2010 data after correction of each profile with the transfer function $OZ_{con}$. The difference profiles are illustrated on the right side of Figure 10 in black for the original data and in red for the corrected data for the period 1996 - 2010. The error bars combine the variabilities of the two original mean profiles. Even though the differences were not significant for the pressure >30 hPa, the correction shows a net improvement for the higher levels.

For a total ozone column comparison three estimations are available for Nairobi station: the ozonesonde integrated profile, a Dobson D018 co-located spectrophotometer and the OMI (Ozone Monitors Instrument) satellite overpass measurements. The change of sensing solution and the corrections shown in Figure 10 have affected the ratios of the total column ozone as shown in Table 4.

### 5. Discussion

There has been a significant effort to reconcile ozonesonde measurements completed with instruments from the two ozonesonde providers, Science Pump and ENSCI, with various combinations of the recommendations for the KI sensor solution concentrations 1.0% and 0.5%. The motivation for this effort rests on characterizations of the precision and accuracy achievable with well-prepared ozonesondes through laboratory tests [Smit and Strüter, 2004a; 2004b; Smit et al., 2007] and field tests [Logan et al., 1999; 2012; Kivi et al., 2007; Deshler et al., 2008]. These results have shown that the precision of an ECC ozonesonde is better than observed systematic differences between ozonesonde type or solution concentration. The results presented here demonstrate that the differences in ozonesonde type, with the same solution concentration, are quite systematic and thus can be characterized, to within experimental uncertainties, with a single relationship for both 0.5 and 1.0% KI concentrations. Similarly, systematic differences between sensor solution concentrations in the same
ozonesonde for both SP and EN ozonesondes can also be characterized by a single relationship. These results attest to the consistency in ozonesonde manufacturing for both companies and that both ozonesonde types have similar differences in performance when the KI solution concentration is varied, pointing again to the strength of the instrumental technique and the instruments.

The rationale employed in this analysis was to find a simple set of relationships which could be applied throughout all the data analyzed. Clearly there are differences in the various datasets as shown in Figure 6. In this case the recommended relationship, Eq. 5, for the relationship between 0.5% and 1.0% KI does not optimally fit the BESOS SP data, Figure 6c, but it does quite well against the Payerne and McMurdo Station data, Figure 6a, 6b. The overestimation of the BESOS SP data is counterbalanced by the under estimation of the JOSIE09 SP data, Figure 6d). This relationship does well against the BESOS EN 0.5 – 1.0% and the Wallops Island SP 0.5-1.0% comparisons (not shown).

The relationship is not steep enough for the Sodankylä measurements at pressures < 30 hPa. Differences such as these led to the alternate transfer functions using first to third order polynomials in log pressure derived by Kivi et al. [2007] and Deshler et al. [2008]. Neither of these relationships, however, would do well across the full data set analyzed here. In particular the third order polynomial provided by Kivi et al. was required due to the significant ratio decrease at pressures below 50 hPa.

Similar comments may arise from the analysis of the ozonesonde type comparisons, Figure 7, although in general the proposed relationship requiring a more significant decrease in the ozonesonde type ratio at pressures below 30 hPa is better at reproducing the ozonesonde type comparisons. The only dataset not shown in this comparison is from JOSIE09 comparing the ozonesonde types at 1.0%. That ratio profile compared to the relationships recommended is quite similar to the comparison at 0.5%, Figure 7c).

The reasons behind the increase in ozone sensed with increase in KI concentration has not been fully explored and is beyond the scope of this paper. The discussions of this effect have centered on the importance of the sodium phosphate hydrate buffers used to maintain the pH of the solution. These buffers, which vary in proportion to the KI concentration, may lead to secondary reactions between iodide ions and the buffer leading to excess iodine, thus indicating additional ozone [Saltzman and Gilbert, 1959; Johnson et al., 2002]. Similarly there have been discussions on the reasons behind the increased sensitivity of the EN ozonesondes compared to SP ozonesondes. Speculation has centered on the efficiencies of the platinum electrode in scavenging the iodine, the conductance of the ion bridge, or the surface properties of the SP Teflon cells versus the EN molded plastic cells, but there has been no systematic investigation of this effect. This also remains beyond the scope of the work presented here.

For a transfer function to have wide acceptance within the community it must have reasonable application to the widest possible set of comparisons. Specialized transfer functions have been derived for particular subsets of the data [Kivi et al., 2007; Deshler et al., 2008] but it has not been demonstrated that these functions are useful beyond the specific data from which they were derived. The analysis here sought to develop as simple a relationship as possible based on the full comparison data set available. This was achieved through weighting of the ratio fits by the number of profile comparisons to arrive at the final four relationships described in Eqs. 5 and 6. Once derived the individual datasets were compared against the derived transfer functions and a subset of these shown in Figures 6 and 7. While Kivi et al. [2007] did not show such a comparison, Deshler et al. [2008] did. Figure 5 from Deshler et al. could be compared here against Figures 6c), 7b), and 7d). Compared to Deshler et al. the fits proposed here improve the comparisons of ozonesonde type while not significantly diminishing the comparisons of sensor concentration. Coupling this with the ability of the fits to reproduce nearly all data sets within the uncertainty of the fits provide strong support for the validity of the proposed transfer functions. This is not to argue that the relationships proposed here should be used instead of results of an individual investigation of a particular comparison dataset; however, such an individual transfer function must be supported by the appropriate measurement set, and made available publicly through the refereed literature. For investigators without access to the resources to conduct such a study, the transfer functions proposed here will do an adequate job of data homogenization.
The final comparison investigated here is between the two manufacturer’s recommendations. This was done through 43 comparison profiles summarized in Figure 8. There was no attempt to derive a fitting function for these data and as the figure illustrates such an exercise would be difficult. The ratio of SP-1.0 to EN-0.5 has a wide dispersion, which is amplified in the figure by the reduced axes for the ratios, from 0.8 – 1.2 compared to Figures 6 and 7. Figure 8 suggests some bias in the smaller datasets investigated, with ratios > 1 for Sodankylä and BESOS, but < 1 for JOSIE09, while Payerne, by far the largest dataset, shows no systematic bias. The objective analysis shown in Tables 2 and 3 quantify these differences but also show that the differences are on average generally not different than 1.0 in contrast for the results of the solution concentration and ozonesonde type comparisons. Thus the data here suggest that the two, manufacturer and WMO, recommended ozonesonde type and solution concentration packages can be used directly and should be widely comparable.

6. Summary and Conclusions

Measurements with various combinations of ozonesonde type, Science Pump or ENSCI, and with differing combinations of the KI solution concentration, 1.0% or 0.5%, have led to variations in ozonesonde preparation at a number of ozonesonde stations throughout the world. These changes began in the mid 1990s and played a role in the analysis of ozonesonde data between then and the late 2000s [Mercer et al., 2007; Tarasick et al., 2016]. Recognizing that these differences exceeded the accuracy and precision that is possible from ozonesondes [Smit et al., 2007; Deshler et al., 2008] led many investigators to independently explore the differences that occur when the same ozonesonde is operated with differing solution concentrations, and when differing ozonesonde types are operated with the same solution concentrations [Johnson et al., 2002; Kivi et al., 2007; Smit et al., 2007; Deshler et al., 2008]. Measurements from these investigators and other unpublished comparisons have been analyzed in this paper. The analysis has focused on three basic comparisons: 1) Sensor solution composition differences in ozonesondes of the same type, 2) Ozonesonde type differences using the same sensor solution concentration, and 3) Differences of the manufacturer and WMO recommendations, Science Pump 1.0% and ENSCI 0.5% KI solution concentrations. Using the published and unpublished data has resulted in the analysis here of 116 profile comparisons for solution concentration differences, 38 profile comparisons for ozonesonde type differences, and 43 profile comparisons of the manufacturer’s solution concentration recommendations. The datasets used in the comparisons have been obtained from the laboratory (JOSIE09), multi-sonde balloon-borne gondolas (BESOS, Sodankylä), and dual ozonesonde balloon-borne gondolas (Payerne, McMurdo Station, Sodankylä, Wallops I., Laramie), involving at least 6 different scientific groups.

Overall the measurements display a satisfying coherence when solution concentrations or ozonesonde type are compared. At pressures above 30 hPa, the surface to 30 hPa, the two measurements can be characterized with a simple ratio displaying almost no pressure dependence. In addition this ratio is, within experimental uncertainty, the same, 0.96, whether the difference is in solution concentration with the same ozonesonde type, or ozonesonde type with the same solution concentration. Ozone concentrations are higher for 1.0% versus 0.5% KI and for ENSCI compared to Science Pump ozonesondes. At pressures below 30 hPa there is a pressure dependence which is linear in \( \log_{10} \) of pressure. This pressure dependence is more pronounced for differences in ozonesonde type.

The results arrived at here are simpler than previous recommendations, but are based on a much more comprehensive dataset and include all of the data used in deriving the previous transfer functions [Kivi et al., 2007; Deshler et al., 2008] both of whom arrived at a relationship requiring a polynomial in altitude or log of the pressure for all pressures. As evidenced here, when the full datasets are investigated, the complexity of these relationships is not justified by the data.

The conclusions arrived at from the analysis described here are the following:

For differences in solution concentration independent of ozonesonde type:

\[
OZ_{0.5\%} = 0.96 \cdot OZ_{1.0\%}, \text{ for } p \geq 30 \text{ hPa},
\]

\[
OZ_{0.5\%} = [0.90 + 0.041 \cdot \log_{10}(p)] \cdot OZ_{1.0\%}, \text{ for } p < 30 \text{ hPa}.
\]

For differences in ozonesonde type independent of solution concentration:
\[ OZ_{\text{Science Pump}} = 0.96 \cdot OZ_{\text{ENSCI}} \text{ for } p \geq 30 \text{ hPa}, \]
\[ OZ_{\text{Science Pump}} = [0.764 + 0.133 \cdot \log(p)] \cdot OZ_{\text{ENSCI}} \text{ for } p < 30 \text{ hPa}. \]

We recommend that all ozonesonde measurements completed with 1.0% KI in ENSCI ozonesondes or 0.5% KI in Science Pump ozonesondes should adjust their data according to the relationships shown above such that the final data product would be representative of 0.5% KI ENSCI or 1.0% Science Pump. This should be done for any data prepared for analysis and for public availability. An uncertainty of 0.05 can be ascribed to the application of these transfer functions.

The investigation of 43 profiles comparing 1.0% KI in Science Pump ozonesondes and 0.5% KI in ENSCI ozonesondes found that the dispersion in the comparisons was centered on a ratio of 1.0. Thus there is no recommendation to alter data obtained from instruments using the recommended concentrations.

If these recommendations are followed it can be expected that datasets experiencing variations in the use of ozonesonde type and solution concentration will see their long term data converge to within the expected ±5% for ozonesondes, and that offsets at the times of transition between the ozonesonde type, or solution concentration change, or both, will be minimized. This will improve significantly the reliability of long term ozone measurements derived from ozone soundings, and indirectly stabilize, in space and time, the long term series of ozone measurements obtained from satellites.

These recommendations have been implemented in the WMO/Global Atmospheric Watch (GAW)’s guidelines for the homogenization of ozonesonde data [Smit and O3S-DQA-Panel, 2012], and recommended to the ozone sounding stations of the Network for the Detection of Atmospheric Composition Change (NDACC) and to SHADOZ stations [Thompson et al., 2012; Witte et al., 2016]. The effort for the ozonesonde investigators to accomplish these corrections will be significant, but in the end the health of the network is dependent on such quality control measures being implemented, and it will greatly add to the value of the measurements. All future measurements should use the WMO/GAW recommendations for solution composition. Any deviation from these recommendations should be justified and carefully researched prior to a change.

Author contributions. T. Deshler with R. Stübi performed the data analysis. T. Deshler was responsible for the majority of the writing with input from all co-authors. R. Stübi provided the Payner data, F. J. Schmidlin the Wallops Island data, J. L. Mercer, B. Nardi the McMurdo Station data, H. G. J. Smit the JOSIE09 data, B. J. Johnson, the Laramie data, R. Kivi the Sodankylä data, T. Deshler the BESOS data.

Data Availability. The ozonesonde measurements used in this analysis are available from the individual investigators and the majority of these data are on the NDACC data base. These data and the code used to created Figures 4-8 are also compiled at ftp://cat.uwyo.edu/pub/permanent/balloon/Manuscripts/Oz_Transfer_Functions/. The data for Figures 9 and 10 are available at the NDACC and SHADOZ data bases.

Acknowledgments. This work was encouraged by the ozonesonde working groups of the WMO/GAW, NDACC, and SHADOZ. The following agencies have supported the generation of these data sets: MeteoSwiss, Finish Meteorological Institute, World Ozoneonde Calibration Facility, US National Science Foundation, US National Aeronautics and Space Administration, US National Oceanic and Atmospheric Administration.

References


Table 1. Datasets used in the analysis giving experiment or location, years of the comparisons, location latitude and longitude, ozonesonde and solution strengths compared, number of comparisons, sampling frequency of the data (Hz), and the platform. The multi-sonde platforms contained from 4 to 12 ozonesones.

<table>
<thead>
<tr>
<th>Location</th>
<th>Dates</th>
<th>Lat.</th>
<th>Long.</th>
<th>Provider-Concen. Compared</th>
<th>No.</th>
<th>Freq (Hz)</th>
<th>Platform/ Gondola</th>
</tr>
</thead>
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<tr>
<td>JOSIE09</td>
<td>1996-2000</td>
<td>50.9</td>
<td>-6.4</td>
<td>EN-0.5 EN-1.0</td>
<td>3</td>
<td>1</td>
<td>Simulator</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>SP-0.5 SP-1.0</td>
<td>3</td>
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<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>SP-1.0 EN-1.0</td>
<td>7</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
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<td></td>
<td>SP-0.5 EN-0.5</td>
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<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>SP-1.0 EN-0.5</td>
<td>3</td>
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<td></td>
</tr>
<tr>
<td>BESOS</td>
<td>April 2004</td>
<td>41.3</td>
<td>105.7</td>
<td>EN-0.5 EN-1.0</td>
<td>9</td>
<td>0.5</td>
<td>Multi-sonde</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>SP-0.5 SP-1.0</td>
<td>9</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
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<td></td>
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<tr>
<td></td>
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<td>SP-0.5 EN-0.5</td>
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<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>SP-1.0 EN-0.5</td>
<td>9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Payerne</td>
<td>2002-2003</td>
<td>46.8</td>
<td>-6.9</td>
<td>En-0.5 En-1.0</td>
<td>48</td>
<td>0.15</td>
<td>Dual-sonde</td>
</tr>
<tr>
<td></td>
<td>2005-2006</td>
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<td></td>
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<tr>
<td>McMurdo</td>
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<td>166.7</td>
<td>En-0.5 En-1.0</td>
<td>18</td>
<td>0.15</td>
<td>Dual-sonde</td>
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<tr>
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<td>-26.6</td>
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<td>SP-1.0 En-0.5</td>
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<td>Wallops I.</td>
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<td>37.8</td>
<td>75.5</td>
<td>SP-0.5 SP-1.0</td>
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<td>Dual-sonde</td>
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<td>Laramie</td>
<td>1996</td>
<td>41.1</td>
<td>105.6</td>
<td>En-0.5 En-1.0</td>
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<td>Multi-sonde</td>
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<td>Total</td>
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<td>EN-0.5 EN-1.0</td>
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<td>SP-1.0 EN-0.5</td>
<td>43</td>
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<td>Recommended</td>
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Table 2. Mean and standard deviations of the various comparison measurements. The mean ratio is equivalent to the slope of a linear fit to the data which passes through the origin. For the datasets with the most data and the most varied comparisons the means and standard deviations are given for the pressure intervals indicated at the top. For the other datasets the means and standard deviations are given for all the data without regard to pressure.

<table>
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<th>Source</th>
<th>Sonde/KI</th>
<th>From</th>
<th>To</th>
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<th>500&gt;p&gt;100</th>
<th>100&gt;p&gt;30</th>
<th>30&gt;p</th>
<th>box</th>
</tr>
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<td>JOSIE09</td>
<td>EN</td>
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<td>0.5%</td>
<td>0.96±0.02</td>
<td>0.95±0.04</td>
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<td>0.94±0.06</td>
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<td>0.96±0.03</td>
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<td>BESOS</td>
<td>0.5%</td>
<td></td>
<td></td>
<td>1.00±0.07</td>
<td>0.98±0.03</td>
<td>0.96±0.03</td>
<td>0.96±0.02</td>
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</tr>
<tr>
<td>JOSIE09</td>
<td>SP-EN</td>
<td></td>
<td></td>
<td>1.01±0.04</td>
<td>0.99±0.03</td>
<td>1.00±0.04</td>
<td>0.98±0.03</td>
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<tr>
<td>BESOS</td>
<td></td>
<td></td>
<td></td>
<td>1.02±0.13</td>
<td>1.03±0.03</td>
<td>1.01±0.03</td>
<td>1.02±0.02</td>
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</tr>
<tr>
<td>Payerne</td>
<td></td>
<td></td>
<td></td>
<td>1.03±0.46</td>
<td>1.00±0.06</td>
<td>1.00±0.04</td>
<td>1.01±0.03</td>
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</tr>
<tr>
<td>Sodankylä</td>
<td></td>
<td></td>
<td></td>
<td>1.02±0.03</td>
<td></td>
<td></td>
<td></td>
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</tr>
</tbody>
</table>
Table 3. Transfer function parameters summary. The second and third columns are for p ≥ 30 hPa providing the ratio and number of data points. Columns 4 – 7 are for p < 30 hPa, providing the slope of a linear fit in log₁₀(p), the offset adjusted to match the ratio at 30 hPa, the offset without constraint to the data below and number of data points. The offset is the value of the ratio at p=1 hPa. The final column is the number of dual sonde measurements for each data set for each application.

<table>
<thead>
<tr>
<th>Dataset</th>
<th>Ratio, p&gt;30</th>
<th>N(p&gt;30)</th>
<th>Slope Log₁₀(p)</th>
<th>Offset adjusted</th>
<th>Offset no adjustment</th>
<th>N(p&lt;30)</th>
<th>N. dual sondes</th>
</tr>
</thead>
<tbody>
<tr>
<td>EN 0.5% to EN 1.0%</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>JOSIE09</td>
<td>0.943±0.056</td>
<td>1918</td>
<td>-0.176</td>
<td>1.203</td>
<td>1.052</td>
<td>949</td>
<td>3</td>
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<tr>
<td>BESOS</td>
<td>0.951±0.028</td>
<td>5003</td>
<td>0.041</td>
<td>0.981</td>
<td>0.882</td>
<td>1854</td>
<td>9</td>
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<tr>
<td>Payerne</td>
<td>0.965±0.546</td>
<td>34855</td>
<td>0.031</td>
<td>0.920</td>
<td>0.910</td>
<td>14744</td>
<td>48</td>
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<tr>
<td>Sodankylä</td>
<td>0.941±0.027</td>
<td>2291</td>
<td>0.109</td>
<td>0.780</td>
<td>0.762</td>
<td>524</td>
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<td>McMurdo</td>
<td>0.977±0.124</td>
<td>15030</td>
<td>0.118</td>
<td>0.802</td>
<td>0.785</td>
<td>4197</td>
<td>18</td>
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<tr>
<td>Laramie</td>
<td>0.974±0.044</td>
<td>1290</td>
<td>0.063</td>
<td>0.882</td>
<td>0.848</td>
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</tr>
<tr>
<td>Weighted</td>
<td>0.966±0.42075</td>
<td>60387</td>
<td>0.041</td>
<td>0.904</td>
<td>0.886</td>
<td>22676</td>
<td>88</td>
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<tr>
<td>SP 0.5% to SP 1.0%</td>
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<td></td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>JOSIE09</td>
<td>0.955±0.129</td>
<td>1919</td>
<td>-0.164</td>
<td>1.198</td>
<td>1.051</td>
<td>949</td>
<td>3</td>
</tr>
<tr>
<td>BESOS</td>
<td>0.975±0.105</td>
<td>5005</td>
<td>0.014</td>
<td>0.954</td>
<td>0.914</td>
<td>1854</td>
<td>9</td>
</tr>
<tr>
<td>Wallops I.</td>
<td>0.937±0.065</td>
<td>10123</td>
<td>0.052</td>
<td>0.860</td>
<td>0.865</td>
<td>2083</td>
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<tr>
<td>Weighted</td>
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<td>0.962</td>
<td>0.919</td>
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<td>SP 1.0% to EN 1.0%</td>
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<tr>
<td>JOSIE09</td>
<td>0.945±0.074</td>
<td>4475</td>
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<td>0.749</td>
<td>0.744</td>
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<tr>
<td>BESOS</td>
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<td>0.756</td>
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<tr>
<td>Sodankylä</td>
<td>0.963±0.026</td>
<td>4041</td>
<td>0.112</td>
<td>0.798</td>
<td>0.784</td>
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<td>8</td>
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<tr>
<td>Weighted</td>
<td>0.956±0.494053</td>
<td>13521</td>
<td>0.133</td>
<td>0.757</td>
<td>0.756</td>
<td>5032</td>
<td>24</td>
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<tr>
<td>SP 0.5% to EN 0.5%</td>
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<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>JOSIE09</td>
<td>0.954±0.054</td>
<td>3193</td>
<td>0.179</td>
<td>0.690</td>
<td>0.680</td>
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<tr>
<td>BESOS</td>
<td>0.977±0.027</td>
<td>5003</td>
<td>0.119</td>
<td>0.801</td>
<td>0.786</td>
<td>1854</td>
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<tr>
<td>Weighted</td>
<td>0.968±0.400037</td>
<td>8196</td>
<td>0.147</td>
<td>0.750</td>
<td>0.737</td>
<td>3441</td>
<td>14</td>
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<tr>
<td>SP 1.0% to EN 0.5%</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>JOSIE09</td>
<td>0.998±0.055</td>
<td>1919</td>
<td>0.880</td>
<td>-0.301</td>
<td>0.142</td>
<td>949</td>
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</tr>
<tr>
<td>BESOS</td>
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<td>5004</td>
<td>0.112</td>
<td>0.842</td>
<td>0.864</td>
<td>1854</td>
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<tr>
<td>Payerne</td>
<td>1.008±0.251</td>
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<td>0.912</td>
<td>0.927</td>
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<tr>
<td>Sodankylä</td>
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<td>2462</td>
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<td>1.019</td>
<td>1.015</td>
<td>642</td>
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</tr>
<tr>
<td>Weighted</td>
<td>1.009±0.094054</td>
<td>26508</td>
<td>0.143</td>
<td>0.797</td>
<td>0.850</td>
<td>10514</td>
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</table>
Table 4. Ratios of total column ozone at Nairobi, Kenya, measured either with the OMI satellite instrument or a Dobson Spectrophotometer, compared to EN ozonesondes using 1.0% and 0.5% KI concentrations. The ratios are also shown after correction of the EN-1.0% to EN-0.5% (OZ_{conc}). Two approaches for the residual ozone column above the balloon burst are given: constant mixing ratio (MR) and using climatology [McPeters and Labow, 2012], the WMO recommendation.

<table>
<thead>
<tr>
<th>Ratios</th>
<th>OMI / EN-1.0%</th>
<th>OMI / OZ_{conc}</th>
<th>OMI / EN-0.5%</th>
<th>Dobson / EN-1.0%</th>
<th>Dobson / OZ_{conc}</th>
<th>Dobson / EN-0.5%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Medians</td>
<td>0.93 ± 0.04</td>
<td>0.98 ± 0.04</td>
<td>0.96 ± 0.04</td>
<td>0.92 ± 0.05</td>
<td>0.96 ± 0.05</td>
<td>0.94 ± 0.05</td>
</tr>
<tr>
<td>Constant MR</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Medians</td>
<td>0.95 ± 0.03</td>
<td>0.98 ± 0.03</td>
<td>0.94 ± 0.03</td>
<td>0.92 ± 0.04</td>
<td>0.96 ± 0.04</td>
<td>0.92 ± 0.05</td>
</tr>
<tr>
<td>Climatology</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
</tbody>
</table>

Figure 1. Boxplot of the background current (µA) measured at the five stations which flew a number of dual or multi ozonesonde gondolas: Sodankylä (Sod), Wallops Island (Wi), McMurdo Station (McM), Payrme (Pay), and Laramie (Lar). Following the station location, the ozonesonde manufacturer is identified (EN or SP) and then the KI solution concentration (0.5% or 1.0%). Medians are the thick black segments, the inter-quartile range the box height, 1.5 times the inter-quartile range the whiskers, outliers denoted as circles, the square root of the number of measurements is reflected in the box width. A value of 0.03 µA corresponds to 0.1 mPa of ozone.
Figure 2. Ozonesonde pump flow rates (cm$^3$ s$^{-1}$) measured at the different stations, following the nomenclature of Figure 1 for the station and ozonesonde.
Figure 3. Mean profile of the measured pump temperature for the different dual flight data sets. The higher pump temperature at the upper part of the profiles for the McMurdo and Sodankylä stations is due to a heat source to avoid freezing of the solution. Error bars are one standard deviation of the measurements.
**Figure 4.** Example of the results of a dual flight profile from Payerne. The only difference between the ozonesondes is the KI solution concentration.
Figure 5. Scatter plots of ozone partial pressures measured with KI concentrations of 1.0% (x-axis) and 0.5% (ordinate-axis) using EN ozonesondes flown from a) Payerne, b) McMurdo Station, and with SP ozonesondes from c) BESOS, and d) JOSIE09. The data are subdivided into four pressure intervals and averages and standard deviations of the ratios of y:x are listed on the panels along with the number of measurements (noted as DP for data points). Only ozone measurements above 0.5 mPa are included. At p ≥ 30 hPa there is little variation in the ratios either by pressure interval, location, or ozonesonde type. At p <30 hPa ratios are decreasing. The 1:1 line is shown in gray.
a) Payene Ensci 0.5% vs Ensci 1.0%
\[ r = 0.900 + 0.041 \log_{10}(p), \text{ at } P < 30 \text{ hPa} \]
\[ r = 0.900, \text{ at } P > 30 \text{ hPa} \]
No. of dual sound comparisons = 48

b) McMurdo Ensci 0.5% vs Ensci 1.0%
\[ r = 0.900 + 0.041 \log_{10}(p), \text{ at } P < 30 \text{ hPa} \]
\[ r = 0.900, \text{ at } P > 30 \text{ hPa} \]
No. of dual sound comparisons = 18

c) Besos SPC 0.5% vs SPC 1.0%
\[ r = 0.900 + 0.041 \log_{10}(p), \text{ at } P < 30 \text{ hPa} \]
\[ r = 0.900, \text{ at } P > 30 \text{ hPa} \]
No. of dual sound comparisons = 9

d) Josie09 SPC 0.5% vs SPC 1.0%
\[ r = 0.900 + 0.041 \log_{10}(p), \text{ at } P < 30 \text{ hPa} \]
\[ r = 0.900, \text{ at } P > 30 \text{ hPa} \]
No. of dual sound comparisons = 3
Figure 6. Vertical profiles of ratios of ozone partial pressure using ozonesondes with different KI concentrations (0.5%, 1.0%) in ozonesondes of the same manufacturer, EN or SP. a) Payerne - EN, b) McMurdo – EN, c) BESOS – SP, and d) JOSIE09 – SP. The hashed areas are regions of low ozone in: ozone hole measurements, McMurdo; tests in the laboratory, JOSIE09; or during initial problems with the BESOS data system at the lowest altitudes. Fits to the data using the transfer functions from Eq. 5, and included at the top of each panel, are shown in blue and black. The numbers in parantheses behind each slope are the uncertainties of the fit.
Figure 7. Vertical profiles of ratios of ozone partial pressure using ozonesondes of different manufacturer (SP, EN) with the same KI concentrations in both ozonesondes, either 1.0% (top two panels a,b) or 0.5% (bottom two panels c,d). a) Sodankylä – 1.0%, b) BESOS – 1.0%, c) JOSIE09 – 0.5%, d) BESOS – 0.5%. The hashed areas denote regions of low ozone in tests in the laboratory during JOSIE09, or during initial problems with the BESOS data system at the lowest altitudes. Fits to the data using the transfer functions from Eq. 6, and included at the top of each panel, are shown in blue and black. The numbers in parantheses behind each slope are the uncertainties of the fit.
8. Ratios of SP-1.0% with EN-0.5% ozonesondes. The measurements are from a) Payerne, b) Sodankylä, c) BESOS, and d) JOSIE 2009. The filled regions represent 35% around 1.0. The hashed areas are as before to indicate regions of low ozone concentration during tests in JOSIE 2009 or difficulty with the data system in BESOS.
**Figure 9.** Ozone time series from the Nairobi station at three pressure levels: 500, 30 and 10 hPa. The black symbols correspond to the data from 1996 - 2010 (EN-1.0% solution) and the red symbols from 2010 onwards (EN-0.5%). The horizontal segments are the mean values over the two periods. No corrections to the EN-1.0% data have been made for this figure.
**Figure 10.** On the left (upper scale), mean ozone profile for the EN-1.0% period in black, EN-0.5% period in blue and corrected EN-1.0% profile using $OZ_{conc}$ in red for the Nairobi data set. On the right (lower scale), the difference between the mean (1996-2010) ozone profiles and the mean (2010-2015) profiles before (after), black (red) correction for the change of solution concentration that occurred in 2010. A small offset of the pressure scale is used to avoid overlapping error bars.