On instrumental errors and related correction strategies of ozonesondes: possible effect on calculated ozone trends for the nearby sites Uccle and De Bilt

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Abstract. The ozonesonde stations at Uccle (Belgium) and De Bilt (Netherlands) are separated by only 175 km, but use different ozonesonde types, different operating procedures, and different correction strategies. As such, these stations form a unique test bed for the Ozonesonde Data Quality Assessment (O3S-DQA) activity, which aims at providing a revised, homogeneous, consistent dataset with an altitude-dependent estimated uncertainty for each revised profile. For the Electrochemical Concen-

5   tration Cell (ECC) ozonesondes at Uccle mean relative uncertainties in the $4-6\%$ range are obtained. To study the impact of the corrections on the ozone profiles and trends, we compared the Uccle and De Bilt average ozone profiles and vertical ozone trends, calculated from the operational corrections at both stations and the O3S-DQA corrected profiles.

In the common ECC 1997-2014 period, the O3S-DQA corrections effectively reduce the differences between the Uccle and De Bilt ozone partial pressure values with respect to the operational corrections only for the stratospheric layers below the

10   ozone maximum. The upper stratospheric ozone measurements at both sites are substantially different, regardless the used correction methodology, the origin of which is not clear. The discrepancies in the tropospheric ozone concentrations between both sites can be ascribed to the problematic background measurement and correction at De Bilt, especially in the period before November 1998. The Uccle operational correction method, applicable to both ozonesonde types used, diminishes the relative stratospheric ozone differences of the Brewer-Mast sondes in the 1993-1996 period with De Bilt from about $20-30\%$

15   compared to the standard pump corrections to less than 5%.

The O3S-DQA corrections bring the Uccle and De Bilt ozone trend estimates for 1997-2014 closer to each other in the lower stratosphere and lower troposphere. Throughout whole the vertical profile, these trend estimates are however not significantly different from each other, and only in the troposphere significantly positive. For the entire Uccle observation period $(1969-2014)$, the operational corrections lead to height-independent and consistent ozone trends for both the troposphere and

20   the stratosphere, with rates respectively $+2$ to $+3\% \, \mathrm{dec}^{-1}$, and -1 to $-2\% \, \mathrm{dec}^{-1}$.

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1   Introduction

Although being a minor constituent, ozone is present throughout whole the lower atmosphere. Depending on the location in the atmosphere, the molecule is involved in different chemical reactions and therefore has a different impact on life on Earth. For instance, ozone absorbs both infra-red and ultraviolet (UV) radiation, but the former reaction is more dominant in the

5   tropopause region, where ozone acts as a greenhouse gas with an estimated globally-averaged radiative forcing of 0.40 ± 0.20 Wm^{-2} (IPCC, 2013). On the contrary, the higher ozone amounts in the stratosphere effectively block the harming solar UV radiation and act as a UV-filter for the living beings on earth. At the surface, ozone is an air pollutant that adversely impacts human health, natural vegetation and crop yield and quality (e.g., Cooper et al., 2014).

Since ozone at different (vertical) atmospheric layers is formed and destroyed by different photochemical reactions - and with

10   precursor emissions from both natural and anthropogenic sources - the time variability of the ozone abundance (on seasonal, inter-annual and decadal time scales) highly depends on the location (height) of ozone molecules in the atmosphere. This is illustrated in Fig. 1, in which stratospheric, tropospheric and boundary layer ozone trends are shown for the period $1969-2014$ for the Uccle (Brussels, Belgium) station and for the period $1993-2014$ for De Bilt (Netherlands). The significant decrease of stratospheric ozone in Uccle over the $1969-2014$ period can be ascribed to the growth of the man-made emissions of ozone

15   depleting substances ODSs (with the chlorofluorocarbons as the typical example) until the late 1980s. Thanks to the regulation of these ODSs in the Montreal Protocol (1987), the stratospheric ozone concentrations stopped decreasing during the late 1990s and should recover in the next decades (Newman et al., 2009; WMO, 2014). Tropospheric and especially boundary layer ozone concentrations increased significantly since 1969, see Fig. 1. This increase is caused by growing emissions of e.g. nitrogen oxides (NO_x), methane, carbon monoxide and hydrocarbons in particularly the first (two) decades (e.g., Logan et al., 2012).

20   Thereafter, a levelling off of the ozone amounts took place due to declining anthropogenic ozone precursor emissions (e.g., Cooper et al., 2014).

The observations used in Fig. 1 to construct the integrated ozone amount time series are gathered with ozonesondes, light-weight instruments attached to weather balloons and electronically coupled with a standard meteorological radiosonde for data transmission to a ground receiver. Ozonesondes provide the vertical distribution of ozone at very high vertical resolution

25   (typically a few 100 metres), up to altitudes in the range 30-35 km. They have been launched worldwide already more than half a century, and therefore constitute the most important data source to derive long-term ozone trends with sufficient vertical resolution up to about 20 km (SPARC-IOC-GAW, 1998). A major concern for any research with ozonesonde measurements is the data homogeneity and consistency, because every profile is obtained with a unique instrument, and different types of ozonesondes exist. Consequently, every ozonesonde needs to be calibrated thoroughly prior to launch. To have consistency

30   between different ozonesonde stations, it is essential to have agreement on procedures for preparation as well as agreement on procedures for data processing and analysis (Smit et al., 2011). Therefore, within the SI[2]N Initiative[1] on "Past Changes in the Vertical Distribution of Ozone" (Harris et al., 2011), an Ozonesonde Data Quality Assessment (O3S-DQA) has been initiated

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[1]this is a joint initiative under the auspices of SPARC (Stratosphere-troposphere Processes And their Role in Climate), the International Ozone Commission (IO3C), the ozone focus area of the Integrated Global Atmospheric Chemistry Observations (IGACO-O3) programme, and the Network for Detection of Atmospheric Composition Change (NDACC). To aid digestion, an acronym of acronyms, SI[2]N, was adopted.

with the aim to provide a revised, homogeneous dataset with corrections being applied for biases related to instrumental changes (such as sonde type or electrolyte solution) in those cases where comparisons or laboratory experiments provide strong evidence for such corrections (Harris et al., 2012). This exercise should result in a significantly improved ozonesonde record with a reduced uncertainty (from $10-20\%$ down to $5-10\%$), giving more solid information about the atmospheric

5 changes that have occurred, as well as a better dataset for comparison with satellite measurements. The Canadian ozonesonde record has recently been re-evaluated regarding the O3S-DQA principles and the results are presented in Tarasick et al. (2015).

In this paper, for the two nearby sites Uccle (Belgium) and De Bilt (The Netherlands), ozonesonde data processed by the principles of O3S-DQA are presented for the first time. However, both stations also developed their own data correction algorithms. Both sites are separated only 175 km from each other. The typical horizontal ozone correlation length is about

10 $500\,\mathrm{km}$ in the troposphere (Liu et al., 2013) and $1500\,\mathrm{km}$ in the stratosphere (Liu et al., 2009). Time scales of autocorrelation vary between about 1.5 and 3.5 days in the troposphere and at 2-6 days in the stratosphere (Liu et al., 2009). Therefore, Uccle and De Bilt have a similar vertical distribution of ozone (see Van Malderen et al., 2014, and Fig. 1). As a consequence, these data enable us to investigate the impact of different correction strategies on the vertical ozone profiles and the derived trends. This research is a follow-up study of Van Malderen et al. (2014), in which the comparison of the ozonesonde data

15 of both stations was used to identify the origin of anomalous high tropospheric ozone in the Uccle data during a 2.5 year period (beginning of 2007 to mid-2009). Moreover, De Backer et al. (1996) report on differences between profiles obtained at Uccle and De Bilt with different types of ozonesondes. They show, although real atmospheric differences cannot be ruled out completely, a systematic altitude-dependent difference between both data sets, ranging from more than 20% near the ground to about -15% at the burst level.

20 This paper is organised as follows: in the next section, we will describe the data, pre-flight preparations and post-flight data processing at the two sites. An uncertainty assessment of the ozone profile data is also provided. In Sect. 3 we assess the impact of these different post-flight data processing steps and methods on the average profiles of both sites. For different time periods, the vertical trends at Uccle and De Bilt are calculated and compared. The impact of the data handling procedures on these trends is analysed in Sect. 4. The last section 5 presents the conclusions of our study.

25 2   Data

Since the 1960s, three main types of electrochemical ozonesondes are in use: the Brewer-Mast (BM, Brewer and Milford, 1960), the Electrochemical Concentration Cell (ECC, Komhyr, 1969), and the Japanese carbon iodine cell (KC, Kobayashi and Toyama, 1966). At present, most sites use ECC sondes. ECC sondes are manufactured either by Science Pump Corporation (SPC; model type 5A and 6A), or, since the early nineties, by the Environmental Science Corporation (ENSCI; model type

30 Z). In 2011 ENSCI was taken over by Droplet Measurement Technologies (DMT). These two types of ECC sondes only have minor differences in construction and differences in recommended sensing solution strength (SST) and of its phosphate buffer (Hassler et al., 2014). For the BM and the ECC sondes, the principle of operation is based on the chemical titration of O_3 in a potassium iodide (KI) sensing solution. For each molecule of O_3 entering the solution in the cell with the help of a very stable

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miniature piston pump, two iodide ions (I^-) are oxidised to form iodine (I_2), which is subsequently reduced back to I^- at the electrodes, generating an electric current of two electrons. This current can directly be related to the number of moles of ozone, sampled per second and cm^3, by the formula (Smit et al., 2011):

$$n_{O_3} = \frac{(I_M - I_B)}{(\eta_c \cdot 2 \cdot F \cdot \Phi_p)} \tag{1}$$

with I_M and I_B respectively the measured electric cell current and background current (both in µA), η_c the conversion efficiency, F the Faraday's constant ($=9.6487 \times 10^4$ C mole^{-1}), and Φ_p the pump flow rate in $cm^3 s^{-1}$. The factor 2 in the denominator points to the number of electrons produced in the sensor cell per ozone molecule. The pump flow rate Φ_p and the background current I_B are measured prior to launch. By applying the ideal gas law the corresponding partial pressure of ozone can be expressed as

$$P_{O_3} = n_{O_3} \cdot R \cdot T_p = 0.043085 \cdot \frac{T_p}{(\eta_c \cdot \Phi_p)} \cdot (I_M - I_B) \tag{2}$$

with T_p the measured pump temperature (K) and R the universal gas constant ($=8.314$ J K^{-1}mole^{-1}).

Uncertainties may change during flight as the pump efficiency degrades with increasing altitude, or due to inaccurate pump temperature measurements or the presence of a background current that is subtracted from the measured current (Staufer et al., 2014, and references therein). Within the O3S-DQA initiative, an uncertainty analysis has been developed and the overall relative uncertainty of P_{O_3} is expressed as a composite of the contributions of the individual uncertainties of the different listed instrumental parameters above (Smit et al., 2011):

$$\frac{\Delta P_{O_3}}{P_{O_3}} = \sqrt{\frac{(\Delta I_M)^2 + (\Delta I_B)^2}{(I_M - I_B)^2} + \left(\frac{\Delta \eta_c}{\eta_c}\right)^2 + \left(\frac{\Delta \Phi_p}{\Phi_p}\right)^2 + \left(\frac{\Delta T_p}{T_p}\right)^2} \tag{3}$$

As some of the contributions depend on the air pressure, the overall uncertainty of the ozone measurement is a function of pressure or altitude. The O3S-DQA initiative therefore provides this uncertainty estimate for each ozone measurement of the vertical profile. It should however be noted that this uncertainty estimation does not take into account the uncertainty due to the time lag of the response of the I_M, T_p and even I_B measurements. For the ECC ozonesondes at Uccle, the average profiles of the relative uncertainties and the contributions from the individual uncertainties of the different instrumental parameters as defined in Eq. 3 are shown in Fig. 2. At Uccle, the overall uncertainty in the stratosphere is between 4 and 5%, while in the troposphere it varies between 5 and 6%. Overall, the conversion efficiency is the predominant uncertainty ($\approx 3.6\%$) at Uccle, and the background current (BGC) has the largest influence on the overall uncertainty at the lowest O_3 concentrations in the upper troposphere. Unfortunately, the physico–chemical origin of the BGC is not well understood and further research is required to better understand its origin and its appropriate measurement and treatment (Smit et al., 2007; Vömel and Diaz, 2010; Smit et al., 2011). The pump efficiency uncertainty contributes significantly to the overall uncertainty at altitudes starting from the ozone maximum, which, in altitudes relative to the tropopause, is in Uccle located around 10 to 15 km (Fig. 2). The average altitude of the tropopause at Uccle is about 11 km.

The relative uncertainties of BM sondes are even harder to estimate, because the BM sonde response is strongly dependent on manufacturing aspects (material used, specifications, provider, etc.) and the preflight preparation procedures employed

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(Tarasick et al., 2002; Stübi et al., 2008). Therefore, the results from previous comparisons of BM sondes with other types of sondes, either on dual flights or in the laboratory, or with other instrument types are not consistent (Smit and Kley, 1998; Stübi et al., 2008). The performance of the BM sondes in the troposphere is even more problematic than in the stratosphere, and the quality of tropospheric data from earlier European BM sondes has been questioned by Schnadt Poberaj et al. (2009) and Logan

5 et al. (2012). The BM sondes flown operationally at Hohenpeissenberg, Payerne, and Uccle from 1994–1997 overestimate O_3 by up to 25 % in the upper troposphere compared to the MOZAIC aircraft measurements (Staufer et al., 2014).

2.1 Meta data

The ozonesonde stations of Uccle (50°48′ N,4°21′ E, 100 m asl) and De Bilt (52°10′ N,5°18′ E, 4 m asl) are located only about 175 km from each other, in urbanized environments. Uccle is in the southern, residential area of Brussels (about 1

10 million inhabitants). It is classified as a suburban station, according to European standards (2008/50/EC, 2008). De Bilt is on the east of Utrecht (about 300000 inhabitants), and about 50 km to the south of Amsterdam, the capital of the Netherlands. The ozonesonde program in Uccle started in January 1969 and is the third longest time series in Europe (after Payerne and Hohenpeissenberg). In the 1980s there were some minor interruptions. The launch frequency is 3 times a week (on Monday, Wednesday and Friday). The De Bilt ozonesonde time series dates back to November 1992, and measurements are made

15 weekly, preferentially on Thursday (or Tuesday, but not on the same day as Uccle).

In Table 1, an overview of the ozonesonde properties of both stations is given. At Uccle, two types of ozonesondes have been used: in April 1997, the BM sondes were operationally replaced with Model-Z ENSCI ECC sondes. However, during the period October 1996 - December 1997, both types were launched interchangeably, either on double soundings (34 pairs, see De Backer et al., 1998a) or individually. The De Bilt time series is built up with a single ozonesonde type, SPC ECC, hereby two

20 model types have been used during the record: SPC 5A and 6A. The latter is in use since 24 July 1997, but with an interception of more than one year (30 September 1999 − 1 March 2001), when the SPC 5A has been launched again. Both stations used the same radiosonde types during their overlap period (Vaisala's RS80 and RS92), but the switch was made at different dates, see Table 1. Before 1990, VIZ radiosondes have been launched at Uccle. Although both ozonesonde stations used different ECC ozonesonde types, they both consistently stuck to the manufacturer's recommended sensing solution strengths (SST),

25 0.5% and 1% KI for ENSCI Z (Uccle) and SPC (De Bilt) respectively. Therefore, the response of both ozonesondes should be very similar, as assessed in an environmental simulation chamber (Smit et al., 2007) and on a balloon experiment (Deshler et al., 2008). It should however be noted that the amount of the sensing solution at De Bilt changed from 2.5 to 3.0 cc on the 23rd of November, 1994.

The largest difference in the operating procedures between the Uccle and De Bilt stations (see also Table 1) is the measure-

30 ment of the BGC. At Uccle, this value is measured in the laboratory before exposure to ozone. In De Bilt, I_B is measured after exposure to ozone and the value is kept small by changing (refreshing) the chemical solutions in the cell several times. The measurement, through the radiosonde system, takes places in the laboratory and/or at the launch field during the inflation of the balloon, typically a couple of hours after the ozonesonde preparation.

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2.2 Data correction methods

The sonde data are processed according to Eq. 2, but design changes (e.g. the presence and location of the pump temperature sensor), differences in pre-flight operating procedures, evolving guidelines following inter-comparison campaigns led to wide variety of post-processing algorithms applied in the ozonesonde network. For instance, the background current is measured

5   at different times during pre-flight preparation, e.g. before or after the sonde is exposed to a sampling flow with about 100 ppbv. This BGC can be assumed constant during the flight, equal to 0 for BM sondes, or alternatively, a pressure dependent BGC correction can be used[2], assuming a small oxygen dependence with a gradual decline that is proportional with decreasing pressure and is negligible in the upper troposphere and stratosphere (Komhyr, 1986). In this latter case, the BGC is assumed to be caused by a small interference of oxygen reacting with KI in the cathode and therefore generating a small additional current

10   (Smit et al., 2007).

Furthermore, it has been observed that at reduced air pressure, the pump flow rate Φ_p in Eq. 2 declines due to pump leakage, dead volume in the piston of the pump, and the back pressure exerted on the pump by the cathode cell solution (Komhyr, 1967; Steinbrecht et al., 1998). This decrease in pump efficiency is corrected by multiplying the pump flow rate in Eq. 2 with a pump correction factor C_{PF} as function of air pressure, based on laboratory measurements of the pump efficiency at reduced

15   pressures (Smit et al., 2007). The different pump efficiency correction profiles C_{PF} used worldwide for the BM and ECC sondes are e.g. shown in Fig. 2 of Stübi et al. (2008). They all smoothly increase with decreasing pressure and predominantly affect the upper part of the ozone profile.

Another common practice is the normalisation (linear scaling) of the ozonesonde profiles to an independently determined total ozone amount (measured by e.g. a Brewer or Dobson spectrophotometer). This is in particular important for BM sondes,

20   because they have a typical response equivalent to about $80-90\%$ of the actual ozone amount (SPARC-IOC-GAW, 1998). Therefore, the partial ozone column above the balloon burst altitude has to be estimated, either by the assumption of a constant mixing ratio or by applying satellite climatologies (e.g. McPeters and Labow, 2012).

2.2.1 O3S-DQA corrections

From the discussion in the previous paragraphs, it is obvious that there is a need for a standardisation of the operating procedures

25   and a homogenisation of the ozonesonde time series (not only between different stations, but also for a given station), which is the aim of the already mentioned O3S-DQA activity. This activity is however restricted to ECC sondes only, not for BM sondes. Consequently, for Uccle, the time series of ozonesonde measurements homogenised according to the O3S-DQA principles, starts with the introduction of ECC sondes in 1997.

The rationale, recommendations and guidelines of the O3S-DQA activity are described in Smit et al. (2012) and can be

30   consulted there. We here shortly give an overview of the proposed corrections for Uccle and De Bilt, also summarized in

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[2]A pressure dependent background current has typically the form $I_B = I_{B0} \times \frac{P}{P_0}$, where I_{B0} is the background current measured during pre-flight preparations at surface pressure P_0 (Smit et al., 2011). The Vaisala manual however proposes a second order correction for the SPC ECC sensor: $I_B = I_{B0} \times \frac{A_0 + A_1 \times P + A_2 \times P^2}{A_0 + A_1 \times P_0 + A_2 \times P_0^2}$, with $A_0 = 0.00122504$, $A_1 = 0.0001241115$, and $A_2 = -2.687066 \cdot 10^{-8}$

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Table 2. The main focus of O3S-DQA is on the development and application of transfer functions to convert either 1.0% KI concentration measurements to 0.5% KI SST, or ENSCI measurements to SPC measurements, or vice versa, so that all ozonesonde data can be traced back to one of the two standards, SPC 1.0% or ENSCI 0.5%. As the ECC data of De Bilt and Uccle respectively are measured with those standards, there is no need of applying a transfer function, as the ratio is 1.0 to within 1.0%. However, during the first two years of operation, the ozone sensors at De Bilt have been charged with only 2.5 cm^3 of cathode sensing solution. In this case, only $\sim 96\%$ of the ozone is captured by the sensing solution at 1000 hPa ground pressure, but this deficit vanishes rapidly with decreasing pressures (Davies et al., 2003). Therefore, for these data, the **conversion efficiency** η_c is not longer equal to one and its composite, the absorption efficiency, was processed by a pressure-dependent expression for pressures above 100 hPa.

For the O3S-DQA correction, both Uccle and De Bilt stations subtracted the **background current (BGC)** measured prior to launch from the measured electrical currents, i.e. the BGC is kept constant. As at Uccle the recommended BGC measurement after ozone exposure is only since recently available, the value recorded before ozone exposure is used. The former is higher than the latter, but never exceeds 0.1 μA at Uccle, because this is the established upper limit for accepting the ozonesonde for launch. In De Bilt, to reduce the I_B, the following strategy has been adopted: after exposure to ozone, the chemicals in the cell were changed (refreshed) as many times as necessary to get the I_B to a small value (< 0.2 μA from 1998 onwards, < 0.1 μA from 2003, see Fig. 3). The value for the BGC that is actually used for the correction, is measured through the radiosonde system, at the end of the calibration procedure. This is typically one or two hours after the rest of the procedure to condition and calibrate the ozone sensor. Normally the I_B has gone down significantly in this period. Before 1998, this value was measured in the laboratory, immediately after the calibration of the radiosonde. From November 2005 onwards the I_B was measured at the launch field during the inflation of the balloon. Between 1998 and 2005 the I_B was measured both in the lab and on the field, see Fig. 3. The value that is used for correcting the ozone profile changed in 2003 from "lab" to "field". The "field" values are typically lower than the "lab" values. Although the constant BGC subtraction with the measured value has been applied for the O3S-DQA correction, this remains questionable for the De Bilt record, as the measured BGCs are too high in the early years. Instead, the O3S-DQA guidelines recommend to use a climatological value of 0.045 μA$\pm$0.03μA for the BGC after exposure of ozone. In this paper, we nevertheless use the measured BGC at De Bilt.

The ECC sondes now used in Uccle and De Bilt are equipped with a thermistor, mounted in a hole drilled in the pump body, to measure the **pump temperature** T_p. However, the pump temperature needed in Eq. 2 is the actual temperature inside the cylindrical housing of the moving piston of the pump, which is about 1-3 K smaller than the measured T_p, depending on the pressure (Smit et al., 2012). Within O3S-DQA, a correction (with an uncertainty of about ± 0.5 K) is proposed, based on simulation chamber measurements. For the periods during which the thermistor was located only in the box (and not in the pump) at Uccle and De Bilt (see Table 1), an additional pressure-dependent correction is applied (Eq. 9 in Smit et al., 2011), because the frictional heating of the moving piston of the pump gives an internal temperature within the pump base that is higher than the external pump temperature. Measurements in the simulation chamber pointed out that the differences between both temperatures were between 0.5 and 2 K at ground pressure, but increased to a maximum in the range $7-10$ K at 50 hPa and then slightly decreasing towards lower pressures (Smit et al., 2007).

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In Uccle, **the pump flow rate** is measured in the laboratory with a Brooks volume calibrator with a mercury ring. In De Bilt, a bubble flow meter is used for this measurement. However, this latter technique is susceptible to an offset due to the evaporation of water from the detector cell, which is positioned between the pump and the bubble flow meter: this is called the "humidification effect". The proposed correction method for this effect (Smit et al., 2011) is based on the temperature

5 and relative humidity at laboratory conditions. These have been recorded in De Bilt for the majority of the flights. In the few cases when these conditions have not been recorded, they have been estimated from the meteorological conditions during the preparations of the sensor. More in general, the equilibrium pump temperature turns out to be about 2 K higher than the room temperature in which the volume calibrator is located (Komhyr et al., 1995; Smit et al., 2012). As a consequence, the actual pump flow rate at ground will be larger than the measured one by a factor of 1.007, and is corrected for accordingly for both

10 stations. This value is then multiplied in Eq. 2 with the already mentioned pressure-dependent pump correction factor C_{PF}, obtained from the laboratory measurements described in Komhyr (1986) for SPC (De Bilt), and described in Komhyr et al. (1995) for ENSCI (Uccle). These two curves differ by about 1 % at 10 hPa and 3 % at 5 hPa.

Finally, the O3S-DQA initiative recommends not to use the total ozone normalisation for ECC ozonesondes, but still to calculate and report the scaling factor when distributing the data through international databases. It can be used as an additional

15 quality indicator of the ozone sounding data. Furthermore, although both Uccle and De Bilt switched from RS80 to RS92 radiosondes and the corresponding change in the pressure sensor affects the vertical ozone profile (Steinbrecht et al., 2008; Stauffer et al., 2014; Inai et al., 2015), we follow the O3S-DQA recommendation not to apply any altitude correction to the profile. Additionally, this radiosonde change also caused a change in the Vaisala interface card, and hence the pump temperature sensor, so that an effect on the recorded pump temperatures cannot be excluded (see Fig. 2 in Van Malderen et al., 2014, which

20 shows a 2°C decrease at 700 hPa). Since this effect is not quantified, no correction can be applied.

2.2.2 The Uccle corrections (PRESTO)

In Uccle, after using BM sondes for about 25 years, the transition was made to ENSCI ECC sondes in 1997. Therefore, the operational post-flight algorithms at Uccle are developed primarily to construct a homogeneous time series, without any break caused by this transition. The details of these corrections can be found in De Backer (1999) and are presented in Table 2. The

25 main aim of the correction strategy is to combine the **pump efficiency correction** with the **total ozone normalisation**, as the latter is required for BM sondes. Therefore, we will use the acronym PRESTO (PRESsure and Temperature dependent total Ozone normalisation) for this correction method in the remaining of the paper. This method is operationally applied only at Uccle, but could also be adapted to other ozonesonde site datasets. In practice, the following steps are taken (see also Table 2). Performing steady-state measurements with BM and ECC sondes in a vacuum chamber at different pressures and

30 temperatures, De Backer et al. (1998a) found that the efficiency of the miniature pumps is not only a function of pressure, but is also dependent on the temperature of the pump, especially for BM sondes, and the following temperature correction was derived

$$k(T) = a_{0,0} + a_{0,1} \cdot T + a_{1,0} \cdot \log_{10}(p) + a_{1,1} \cdot T \cdot \log_{10}(p) \quad (4)$$

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where $k(T)$ represents the factor by which the time to pump 100 ml ($\propto 1/\Phi_P$ in Eq. 2) of air at 20°C must be multiplied to obtain the time at temperature T (in °C), and $a_{i,j}$ regression coefficients. These factors are visualized for different pressures and temperatures in Fig. 2 and Fig. 3 in De Backer et al. (1998a) for BM and ENSCI ECC sondes respectively. Then, based on vacuum chamber steady-state measurements with varying pressure (but now with fixed temperature) of 200 BM sondes and

5   150 ENSCI sondes, De Backer et al. (1998b) obtained pump correction factors C_{PF} that are higher than the corresponding standard correction factors (Komhyr and Harris, 1965; Komhyr et al., 1995, for BM and ENSCI ECC sondes respectively). Both sets of the obtained measurements could be fitted by a similar equation for the time needed to pump 100 ml of air at pressure p:

$$t(p) = t(p_0) \frac{1 + \sqrt{\frac{b}{p}}}{1 + \sqrt{\frac{b}{p_0}}} \tag{5}$$

10   with p_0 the ground pressure and b a parameter depending on the sonde type. Inspired by this equation, De Backer et al. (1998b) proposed the following empirical shape for the pressure dependency of the pump flow rate (= the pump flow correction factor C_{PF}):

$$C_{PF}(p) = c_0 \frac{1 + \sqrt{\frac{b}{p}}}{1 + \sqrt{\frac{b}{p_0}}} \tag{6}$$

with c_0 the ground calibration factor determined with a calibrated ozone source ($320\,\mu g m^{-3}$ running through the ozone sensor during 10 minutes) before launch and b a parameter depending on the performance of the sensor, determined in such a way that the integrated amount of ozone in the profile (increased with the residual amount of ozone), is equal to the total ozone measured with a spectrophotometer at the same site. In other words, the pump flow correction factor C_{PF}, determined after the temperature dependency correction of the pump flow rate in Eq. 4, is adjusted for each pump individually as to match both the single point calibration of the ozone sensor at the laboratory and the total ozone column value measured on site. For

20   completeness, we add that the residual amount of ozone is calculated with either the constant mixing ratio assumption or the McPeters and Labow (2012) satellite climatology, depending on the balloon burst altitude, as prescribed by WMO (1987). When the ground calibration factor c_0 is not available (i.e. before May 1992), the value c_0 in Eq. 6 is estimated from a relation between c_0 and the total ozone scaling factor, depending on the quality of the pumps. Since the movement of the manufacturing company seemed to have resulted in an inferior quality of the pumps used after April 1989, two different relationships have

25   been determined. Applying this correction method to both BM and ECC ozonesondes, De Backer et al. (1998a) could lower the ozone differences of 26 dual soundings at Uccle to within 3 % over almost the entire altitude range, while Lemoine and De Backer (2001) could reduce the drift between SAGE II and ozonesondes from $-0.51\,\%\text{yr}^{-1}$ to $-0.07\,\%\text{yr}^{-1}$ between 17 and 22 km with non-significant values at the 2σ level at all altitudes.

The above described procedure for the pump efficiency correction and total ozone normalisation makes up the largest differ-

30   ence with the so-called standard corrections or the O3S-DQA corrections (for ECC sondes). However, other smaller differences exist and some additional corrections, especially for BM sondes, have been developed at Uccle and are applied operationally

there. First, the **background current**, measured in the laboratory before exposure to ozone since May 1992, is subtracted from the measured cell currents over the whole altitude range for the ECC ozonesondes. For the BM sondes, no correction for the BGC is made ($I_B = 0$). However, before October 1981, the ozone concentrations imposed to the sensor during the preconditioning phase in the laboratory were much lower than recommended (WMO, 1987), causing too low ozone concentrations in

5   the lower tropospheric ascent profiles, as found by comparing the ascent to descent ratios of ozone profiles before and after that date (De Backer, 1994). Therefore, a pressure-dependent amount of ozone partial pressure is added to the ascent profiles from ground to 70 hPa, as proposed by De Backer (1999), which can be interpreted as a correction for "a negative BGC caused by impurities in the sensor".

The introduction of Vaisala radiosondes in 1990 allowed to measure the temperature in the Styrofoam box containing the

10 ozone sensor pump ("box temperature"). Since December 1998, the **pump temperature** T_P is measured with a thermistor in a hole in the pump. For the ECC sondes, this measured value (either box or pump temperature) is used in Eq 2. For the BM sondes launched after 1990, we use the measured box temperature as an approximation of T_P, instead of a recommended fixed value of 300 K (WMO, 1987), that is known to produce an overestimation of the ozone partial pressure of about 8 % near the burst altitude. From May 1989 to December 1989, the mean box temperature as derived from the soundings during 1990 and

15 1991 is used. As the insulating capacity of the Styrofoam boxes used before 28 April 1989 was higher, a modified average box temperature profile is used for this period, with a slower temperature decrease adjusted to reach the measured 7 °C at 10hPa (instead of 3 °C thereafter).

With the replacement of the VIZ radiosondes by Vaisala radiosondes in 1990, the accuracy of the pressure measurements increased substantially, which has an impact on the (BM) ozone profile measurements. At Uccle, between 1985 and 1989,

20 more than 450 soundings were used to calculate the differences between the altitudes from the VIZ radiosondes and the altitudes deduced from the tracking of the balloon train with a primary wind-finding radar (De Muer and De Backer, 1992). They showed that a systematic bias of up to 1.5 km was present near the top of the soundings, caused by the slow response time of the VIZ pressure sensor. Furthermore, the differences seemed to have changed during the campaign period, probably due to an additional calibration error in the period 1985−1988. Consequently, different **altitude corrections** have been made

25 for these different periods, see De Backer (1999). For the period before 1985, when no radar information is available, the more conservative altitude correction of the period 1988−1989 is applied. Although smaller differences between the radar and Vaisala altitudes were observed during a small campaign in the period September−December 1989, no altitude correction is made for this type of sondes.

The electrochemical sensors of both BM and ECC ozonesondes are also sensitive to other atmospheric trace gases, such as

30 SO_2. The decrease in the ozone readings of BM sondes is proportional to the SO_2 concentration with a proportionality factor of 1, within the limits of uncertainty (De Backer, 1999, and references therein). But also the total ozone measurements with a Dobson spectrophotometer, used for the total ozone normalisation of the ozone profile when available (before 2009), suffer from interference with SO_2 (De Muer and De Backer, 1993). Since Uccle is located near the large urban area of Brussels, it has been affected by SO_2 contamination in the 1970s (the beginning of the time series), but the SO_2 levels in the lower troposphere

35 decreased rapidly in the 1970s and to a lesser extent in the 1980s. Therefore, without a correction for the **SO_2 interference**,

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a fictitious (Dobson) total ozone trend has been induced (De Muer and De Backer, 1993) and the lower tropospheric ozone trends calculated from the BM sondes would be overestimated. Therefore, for Uccle, corrections for the SO_2 interference on the BM ozone soundings (and on the Dobson spectrophotometer) are applied (De Backer, 1994, 1999), making use of the in situ measurements of the SO_2 density near the ground in the urban area of Brussels (and even at Uccle itself). Since the Z-ECC

5 sondes were not used in Uccle before 1996, when the SO_2 concentrations in Brussels had already stabilized at low values, the impact of these concentrations on the ozone soundings is negligible and no correction for SO_2 needs to be applied to the ozone profiles obtained with this type of sensor.

As already mentioned, this complete set of corrections, operationally applied at Uccle, will be referred to as the PRESTO corrections.

10 ### 2.2.3   The De Bilt corrections

The focus of the ozonesonde programme of the De Bilt station, now 22 years long, lies more on the satellite validation and the Match campaign for the determination of stratospheric polar ozone losses[3], rather than the creation of a homogeneous long term data record. As a consequence, small changes in their procedures and data processing have occurred several times. However, the data from the ozone sensor has been digitised on board the sonde, and all original raw data are still available.

15 It is not our purpose to discuss here all changes that have been made over time, but concentrate on the ones that affect the homogeneity of the data series, also presented in Table 2.

The most significant changes took place in late 1998, when the participation of De Bilt in the Match campaign started, and an agreement on standardisation of operating procedures and data processing was reached among the participating ozonesonde stations. Therefore, from November 1998 onwards, the environmental conditions in the laboratory were recorded, the pump

20 temperature instead of the box (or sensor) temperature was measured, another pump efficiency correction table was used (Komhyr et al. (1995) instead of Komhyr (1986)), the background current value was reduced by adopting a new measurement strategy (see above), and the constant BGC subtraction was applied.

Most critical for the homogeneity in the De Bilt dataset is the BGC. Before late 1998, the measured BGC values were too high (see Fig. 3), so that the BGC subtraction leads to an underestimation of the total ozone column from the integrated

25 profile with respect to the co-located Brewer instrument's value, which is even noted until the year 2000. Because a pressure-dependent correction subtracts a smaller BGC through the profile than the subtraction with a constant value – the subtracted BGC equals the measured one at ground pressure and then decreases with increasing pressure, see Sect.2.2 – the pressure-dependent correction with the measured BGC for the period before the end of 1998 is still preferred. As the BGC values in De Bilt decreased over time (see Fig. 3), this trend will have an impact on the calculated trends of (in particular tropospheric)

30 ozone, see Sect. 4. But also the change of the BGC subtraction method might generate an artificial trend in the ozone profile data series. Furthermore, as an ozone destruction filter is used for the BGC measurement, a seasonal dependent offset in the ozone profile is a distinct possibility if the efficiency of this filter is not equal to 100 %. This remark also applies to the Uccle dataset, where an ozone destruction filter is used too for measuring I_B.

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[3]http://www.awi.de/en/research/research_divisions/climate_science/atmospheric_circulations/expeditions_campaigns/ozone_loss_campaigns_match/

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3 Impact on the average ozone profiles

The different possible post-processing steps described in the previous section all have an impact on the final ozone profile. In this section, we will quantify these impacts on the average ozone profiles, first for Uccle and De Bilt separately. Thereafter, we will compare the resulting average ozone profiles of both stations.

3.1 Uccle

As two types of ozonesondes have been used at Uccle, we will treat them separately in this section.

3.1.1 The BM 1969-1996 time series

To visualize the influence of the different steps in the Uccle PRESTO corrections on the average ozone profile obtained by BM sondes, we show in Fig. 4 the relative differences to the profile obtained by applying only the correction of the pump efficiency decrease with the standard correction factors (Komhyr and Harris, 1965). A first thing to note is that applying the total ozone normalisation by multiplying the profile with a scaling factor (gold dotted curve in Fig. 4) causes a relative ozone increase of $20-25\,\%$ throughout the profile, compared with the reference average ozone profile. This number is in agreement with the fact that BM sondes of Uccle are known to have a typical response equivalent to about 80% of the actual ozone amount with total ozone scaling factors in the range $1.1-1.3$ (De Backer, 1999).

The combination of the correction for the pump efficiency decrease with decreasing pressure and the total ozone normalisation leads to a smaller relative difference in the troposphere (around $10-15\,\%$), and higher relative differences above the ozone maximum (see black curve in Fig 4). This can be explained by the fact that the used pump correction factors, determined in the vacuum chamber at Uccle, are higher than the standard correction factors on one hand, and on the other hand by the redistribution of the total ozone amount over the entire profile. With our combined method, layers contributing hardly to the total ozone amount, like e.g. the troposphere, will be exposed to smaller ozone normalisation scaling factors, as should be obvious from the figure. Additionally, also the poorer performance of the pump for decreasing temperatures is corrected for at Uccle. However, as without any box temperature correction a constant value of $300\,\mathrm{K}$ is assumed for BM soundings before 1990, we show in Fig. 4 the combined effect of both contributions (red curve). These pump temperature effect corrections seem to have the largest impact on tropospheric ozone, if we compare with the previous described correction (black curve). At first sight, this seems contradictory, because it is in the upper parts of the atmosphere that the pump efficiency is most affected by the lower temperatures and the box temperature deviates most from the $300\,\mathrm{K}$ standard value. Once more, we should keep in mind that these effects have been smeared out over the entire profile by the redistribution of the total amount of ozone. Because of a detected change in quality of the Styrofoam boxes and pumps used after April 1989, alternative corrections for both the box temperature and the pump efficiency were extrapolated for the period before April 1989. As can be seen from Fig. 4 (cyan curve), the effect of these corrections is quite large, especially in the upper part of the profile, where their impact is largest (see also the comparison with SAGE II data in Lemoine and De Backer, 2001).

The following two additional corrections for BM ozonesondes that are investigated, especially affect the tropospheric ozone. The first one, the correction for a negative background current because of the sensor being exposed to too low ozone concentrations during the preconditioning before October 1981, enhances the tropospheric ozone by about 10 to 15 % (magenta curve in Fig. 4). The correction for SO_2 interference adds another $5-10\%$ of ozone in the boundary layer (see green curve in Fig. 4), and

5 even around 25 % at the surface (not shown here). When we finally add the altitude correction for radiosondes launched before 1990, which affects especially the ozone profile at and above the ozone maximum (see grey curve in Fig. 4), the complete set of the PRESTO correction is in use (blue curve in Fig. 4). With respect to the standard pump correction, all these corrections give a roughly 30 % ozone increase in the free troposphere and even between 30 and 40 % in the lower troposphere/boundary layer. The impact of the PRESTO correction is lowest in the lower stratosphere (around 20% ozone increase), and increases

10 again from the ozone maximum to reach again 30% in the upper parts of the sounding. The PRESTO post-processing steps have been developed based on simulation chamber tests, double soundings, the comparison of ascent and descent profiles, etc. and have been validated against reference satellite data (SAGE II).

3.1.2 The ECC 1997-2014 time series

For the ECC time series, we again chose to confront the corrected profiles with the standard pump corrected (average) Uccle

15 ECC profile in Fig. 5. The alternative correction methods at Uccle produce average profiles within $\pm 2\%$ of this reference profile, a number even smaller than the estimated uncertainties for the Uccle ECC profiles (see Fig. 2). These smaller relative differences compared to the average BM profiles shown in Fig. 4 are due to the nearly 100 % response equivalent of the actual ozone amount of ECC sondes. Indeed, the total ozone normalisation by simple linear scaling increases the ozone relatively by less than 1 % throughout the profile (see gold dotted curve in Fig. 5). Consequently, the relative differences for the average

20 profiles processed by the Uccle pump efficiency correction method, with a pressure-dependent total ozone normalisation (in black in Fig. 5), are within the same range. They increase with decreasing pressure, because the measured pump efficiency correction factors in the vacuum chamber in Uccle are higher than the standard correction factors, see e.g. Fig 2 in Stübi et al. (2008). Introducing the temperature dependence of the pump efficiency in the corrections (to complete the PRESTO correction, blue curve in Fig. 5) adds another 1 % relative difference in the troposphere and the upper stratosphere. A similar

25 vertical behaviour of this temperature dependency correction as in Fig. 4 is observed. For ECC sondes, the relative differences only due to this correction (hence applying only Eq. 4) increase from around 0 % in the troposphere to a maximum of 4 % at balloon burst altitudes.

The Uccle O3S-DQA corrected profile is also included (grey curve in Fig. 5), and resembles the chosen reference most (within $\pm 1\%$), as could be expected from methods using the same standard pump efficiency correction factors (and applying

30 no total ozone normalisation). The difference is largest at high altitudes, due to the pump temperature correction applied in the O3S-DQA corrections. The difference between the two correction methods at Uccle (PRESTO and O3S-DQA) is largest in the troposphere (about 2 %), which can be explained by the redistribution of the total ozone amount over the entire profile interacting with the pressure and temperature dependent pump efficiency correction. Especially the temperature dependency

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correction blows up the differences with the O3S-DQA correction (compare the black and blue curves in Fig. 5), also at altitudes above the ozone maximum.

3.2 De Bilt

Now we compute for the entire observation period of De Bilt (1993-2014) the average profiles of the two different correc-

5 tion strategies: one generated according to the O3S-DQA guidelines, and another one corrected by the De Bilt operational algorithms (see Table 2). In Fig. 6 (green line), we compare the vertical profile of the relative differences between both those average profiles. A first important note is that the O3S-DQA average profile has smaller ozone amounts at all altitude levels. The relative differences between both corrections are smallest at the surface and the ozone maximum (around 2 %), and largest at the tropopause (about 6 %). Above the ozone maximum, the relative differences increase to a 4 % at burst altitude. The vari-

10 ation of these relative differences in altitude is caused by the differences in the correction and operating procedures before the end of 1998 (black curve in Fig. 6). From November 1998 on, the MATCH standard operating procedures were applied in the operational chain at De Bilt, resulting in an average profile differing by only 2 % at all altitudes with the O3S-DQA corrected profile for the same period (red curve in Fig. 6). Before 1998, the large relative differences in especially the free troposphere (even more than 15 %) can be ascribed to the different background current correction strategies applied in the O3S-DQA and

15 operational dataset. In both cases, the same (relatively high) value for the BGC is used, but this (constant) value is subtracted at all pressure levels for the O3S-DQA correction and a pressure-dependent BGC subtraction is applied for the operational correction. Because the subtracted BGC value decreases with increasing pressure in the latter case, the O3S-DQA correction results in lower ozone partial pressures at all pressure levels. The relative differences between the two average profiles are therefore largest in this period at those levels where the impact of the BGC on the measurements is highest (the free troposphere, see

20 Fig. 2) and the difference between the subtracted BGC values is largest (at the lowest pressures, see Fig. 6). However, because the measured background current values are so high before 1998 (see Fig. 3), BGC values in the range $0.1-0.2\,\mu$A correspond to about $3.5-7$ ppbv of ozone at surface and about $25-30$ ppbv in the upper troposphere at 200 hPa, which easily can introduce differences of 15 % or larger when either using a constant or pressure-dependent BGC correction. The comparison with the Uccle ozone profile in Fig. 6 (blue curve) illustrates the lower tropospheric ozone concentrations observed at De Bilt by both

25 BGC corrections. Therefore, the O3S-DQA guidelines (Smit et al., 2012) recommend to use a climatological value of 0.045 μA for the BGC after exposure of ozone.

3.3 Comparison of Uccle and De Bilt

As already mentioned, in the previous figure 6, the relative differences of the average Uccle PRESTO corrected ozone profile with the operational De Bilt profile are also added. For the $1993-2014$ period, higher tropospheric ozone amounts (by about

30 2 to 5 %) are measured at Uccle, while in the lower stratosphere the reverse is true by about the same ozone amounts. From the ozone maximum to burst altitudes, the Uccle ozonesondes measure increasingly higher ozone amounts than at De Bilt with a maximum relative difference around 8 %. In this section, we elaborate more on those Uccle-De Bilt average ozone profile

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differences. Because the Uccle data series covers two different types of ozonesondes in $1993-2014$, we make the comparison with De Bilt for each type separately.

3.3.1 Uccle BM versus De Bilt ECC: 1993-1996

For the comparison of the average profiles of Uccle, gathered by BM ozonesondes, and De Bilt, we first look at the relative

5 differences between the operationally corrected De Bilt average profile and the Uccle profile corrected with the standard pump efficiency factors (see cyan curve in Fig. 7). These profiles have in common that the recommended standard pump efficiency factors for the different ozonesonde types are applied. However, this Uccle BM average profile have lower ozone values than the De Bilt average profile at all altitudes, and even by 30 to 50% above the ozone maximum. This comparison demonstrates again clearly the need of applying a total ozone normalisation for BM sondes. For this time period, the BM sondes underestimate the

10 total ozone amount by about 25% on average (see the dotted gold curve in Fig. 7). Moreover, it should also be obvious from this example that the developed correction algorithms in Uccle are able to reduce the relative stratospheric ozone differences with De Bilt to less than 5%. After applying the operational corrections at both sites, the Uccle and De Bilt average profiles show a very similar vertical ozone distribution in the stratosphere, but with higher ozone partial pressures (by $2-5\%$) at all levels at De Bilt (compare the blue and cyan curves in Fig. 7). Due to the constant BGC subtraction, the O3S-DQA corrected

15 De Bilt average profile (in green) shows lower ozone concentrations at all pressure levels with respect to the PRESTO corrected Uccle profile, except at the layers just below and at the ozone maximum, where similar ozone concentrations are measured at both sites. In the other stratospheric regions, the O3S-DQA De Bilt profile is lower by about 5%.

In the troposphere, both the De Bilt corrections result in lower ozone amounts compared to the Uccle PRESTO corrected ozone partial pressures, ranging from $10-25\%$ for the O3S-DQA, and $2-10\%$ for the operational corrections. With the

20 high measured background currents at De Bilt, especially during this time period, these large differences can be expected with either BGC correction method. For the Uccle BM sondes, no BGC correction was applied. Another reason for the higher ozone amounts in the troposphere above Uccle could be the more urban area at Uccle (Brussels) and consequently higher emissions of ozone precursors like NO_x, methane, CO, Volatile Organic Compounds (VOCs), etc. The correction for SO_2 interference for BM sondes now affects only marginally the average profile in the boundary layer (at most 2%, compare the blue and red

25 dashed curves in Fig. 7). The other BM corrections included in the PRESTO method are not valid for the 1993-1996 time period.

3.4 Uccle (Z-ECC, 0.5%) versus De Bilt (SPC-ECC, 1%): 1997-2014

We now concentrate on the comparison of the average profiles during the time period in which both stations launched ECC ozonesondes, although different types and with different sensing solution strengths. We therefore look back at Fig 5. The most

30 striking feature in this figure are the large relative ozone differences at upper stratospheric altitudes. At burst altitudes, the relative differences between the Uccle and De Bilt ozone partial pressures can amount to up to 10%, independently of the used correction method. This number also exceeds substantially the quoted 3% difference at 5 hPa between the standard pump efficiency factor used at De Bilt and Uccle for the O3S-DQA corrections. The origin of the large relative ozone differences in

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the upper stratosphere is not clear to us, in particular because the agreement in the lower stratosphere is fairly good, around 5 % at most. In any case, measuring the ozone concentrations above 25 km is the most challenging for ozonesondes due to e.g. the pump efficiency decrease and the evaporation of the sensing solutions, but the relative differences found at those layers are also well above the quoted relative uncertainties of 5−6 % in Fig. 2. A relative difference around 5 % is achieved for the

5   troposphere, somewhat less for the boundary layer (2−5 %), and somewhat more for the upper troposphere (5−8 %).

   Finally, we investigate whether or not the uniform O3S-DQA corrections for the Uccle and the De Bilt stations result in a closer agreement of their average profiles. The relative differences between the two correction methods at De Bilt (in cyan and green in Fig. 5) vary between 2 and 4 %, with the larger value for the upper troposphere - lower stratosphere (UTLS) region. Based on Fig. 5, we can conclude that only for the lower stratosphere, the layers below the ozone maximum, the O3S-DQA

10   corrections effectively reduce the relative differences between the Uccle and De Bilt ozone partial pressures. In the troposphere, the O3S-DQA corrections enhances the relative differences, compared to the operational correction methods at Uccle and De Bilt. But, different tropospheric ozone concentrations at Uccle and De Bilt are to be expected due to different environmental conditions. In the upper stratospheric layers, the O3S-DQA correction at De Bilt increases the differences with Uccle, but the opposite is true for the O3S-DQA correction at Uccle.

15   4   Impact on the vertical ozone trends

Looking back at the ozone monthly means for three distinct (vertical) atmospheric layers in Fig. 1, the similar seasonal behaviour in the Uccle and De Bilt time series stands out. In this section, we will study the long-term time behaviour of the Uccle and De Bilt ozone series, which span different time periods. In particular, we will analyse the impact of the different correction strategies on the resulting vertical ozone trends, for different periods. To determine these trends, we first calculate

20   the monthly anomalies of ozone partial pressures in layers of 1 km height, relative to the tropopause height. Then, for each of these layers, (robust) trends are estimated from the monthly anomaly time series by simple linear regression. We did not apply a multiple linear regression model (e.g., Harris et al., 2015) to calculate trends, because the focus is here on differences between trends rather than on the trend values themselves. Compared to the average ozone profile calculation, we chose a lower vertical resolution, because the trend estimation is more sensitive to the number of available measurements per layer. Nevertheless, we

25   stress that the results are comparable when using the identical vertical resolution as for the average profiles.

4.1   The entire Uccle time series: 1969-2014

For the different correction steps present in Fig. 4, the estimated relative trends are shown in the same colour coding in Fig. 8. For the beginning of the time period, all PRESTO algorithms were in use and therefore have a large impact on a trend estimation based on simple linear regression. Indeed, this chain of corrections leads to a serious reduction of the overall positive

30   trends estimated from the profiles corrected only by the standard pump efficiency profiles (compare the blue and purple lines in Fig. 8). Whereas the altitude correction (in dashed grey) logically only reduces the trends in the upper stratospheric layers by about 1 tot 3 % dec^{-1}, the total ozone normalisation (gold dotted) is responsible for a relative trend decrease by 6−7 % dec^{-1}

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in the stratosphere, hereby inducing negative trends in the bulk of the stratosphere, and by around $5\,\%\,\mathrm{dec}^{-1}$ in the troposphere. Since this total ozone normalisation corrects for the lacking total ozone response equivalent by the BM sondes at the begin of the period, while the ECC ozonesondes have a nearly full total ozone response equivalent, it is clear that the ozone concentration trends will be smaller after this correction. With the introduction of a pressure-dependent total ozone normalisation

5   by combining it with the pressure and temperature dependency of the pump efficiency (red curve in Fig. 8), the trends are increased in the troposphere (by at most $2\,\%\,\mathrm{dec}^{-1}$) and decreased in the upper stratosphere (by about $3\,\%\,\mathrm{dec}^{-1}$). This follows immediately from the redistribution of the total ozone over the profile, illustrated in Figs. 4 and 5, which again is stronger for the BM sondes than for the ECC sondes.

The amendment for the quality change of the Styrofoam boxes and pumps in 1989 through alternative formulations of the
10   box temperature and pump efficiency corrections (cyan curve) also affects the trend estimates to a large extent, as it only applies to the BM sondes in the beginning of the period. Because this method is based on extrapolations, it causes a larger uncertainty in the trend estimates. These latter are reduced at all altitudes because of the total ozone redistribution throughout the profile, except above the ozone maximum, where the correction has the largest impact, and the ozone trends increased. The so-called negative background current correction for BM sondes preconditioned before October 1981 (in magenta) enhanced
15   the average tropospheric ozone profile by about $5\,\%$ (see Fig. 4) and therefore downsizes the tropospheric ozone trends for the entire period by 5 (tropopause) to 10 (boundary layer) $\%\,\mathrm{dec}^{-1}$. After applying the correction for the SO_2 interference on the ozone soundings at Uccle (green line), the calculated ozone trends in the boundary layer are reduced by about half. Changes in boundary layer ozone also propagate to trends to other altitudes because of the normalisation procedure, but we see that upper tropospheric and stratospheric trends are hardly affected by this correction.

20   The final trends, obtained after executing the whole set of PRESTO post-processing algorithms, are also shown in blue in Fig 8, together with their trend uncertainty estimates. These resulting trends are fairly constant and consistent over the two different atmospheric layers considered here: the ozone concentrations increased at a rate of 2 to $3\,\%\,\mathrm{dec}^{-1}$ in the troposphere, and they decreased at a rate of 1 to $2\,\%\,\mathrm{dec}^{-1}$ in the stratosphere from 1969 to 2014. Taking the calculated uncertainties into account, these trends are significant at almost all altitude levels.

4.2   The entire De Bilt time series: 1993-2014

Analogously to the previous time period, applying the Uccle pump efficiency correction method (in black in Fig 9), which is driven by the total ozone normalisation, leads to a significant reduction of the positive trends compared to the standard pump correction: by about $5\,\%\,\mathrm{dec}^{-1}$ in the troposphere, and by about 5 to $10\,\%\,\mathrm{dec}^{-1}$ in the stratosphere, with an increasing trend reduction with increasing pressure in the latter case. This significant change of trends after these corrections can be ascribed
30   to the larger impact on the BM sonde measurements, which were launched during the first 4 years of the considered time period, and thereby affecting to a large extent the linear trends. The resulting final relative trends at Uccle (in blue) remain positive at all altitudes, and are only in the boundary layer and the upper troposphere not significant, if we take into account the error bar ranges. They vary between $1-4\,\%\,\mathrm{dec}^{-1}$ in the troposphere, and between $2-9\,\%\,\mathrm{dec}^{-1}$ in the stratosphere. As the stratospheric ozone concentrations reached their minimum in 1993 at Uccle (see Fig. 1), because of the combination of the high

amount of ODSs in the atmosphere and the volcanic eruption of Pinatubo in June 1991, the increasing ozone concentrations in the stratosphere are not surprising. Compared to the entire time period trends, there is more variability of the calculated trends between different adjacent altitude levels, the trends are less consistent per atmospheric layer.

The De Bilt relative trends in ozone concentrations (cyan curve in Fig 9) are more modest than the corresponding Uccle trends, except in the boundary layer. Only there and in the lower stratosphere, the (positive) trends (of about $5\,\%\,\mathrm{dec}^{-1}$) in ozone concentrations are significant. At burst altitudes, even negative ozone trends are found, though not significant. The Uccle and De Bilt trends lie within each error bars, and could therefore considered as not significantly different, except at the highest and lowest altitudes of the range. If only the standard pump corrections should be applied for Uccle (in purple), the trend differences between Uccle and De Bilt would be very significant for this time period, ranging from 5 tot $20\,\%\,\mathrm{dec}^{-1}$ in the stratosphere.

The O3S-DQA corrections of De Bilt produced an average profile with lower ozone partial pressures at all altitudes compared to the operational corrections (see Fig. 6), with the largest differences in the beginning of the time period (especially before end 1998), see also Fig. 7. Consequently, the derived trend estimates for the O3S-DQA corrected profiles (in green in Fig 9) are larger at all altitudes, for both the absolute (not shown here) and relative trends. The largest differences in the trends occur in the upper troposphere and in the highest stratospheric layers, where also the corresponding average profiles diverged most during the period 1993-1998 (see Fig. 7). The O3S-DQA trend profile follows closer the Uccle trend profile in the stratosphere than the operational De Bilt profile.

4.3 The common ECC time series: 1997-2014

Now we concentrate on the time period in which both the Uccle and the De Bilt station are using ECC ozonesondes, although produced by a different manufacturer. First, we note from Fig. 10 that the different correction methods used at Uccle only have a minor impact on the calculated trends: the trend differences with the standard pump correction are within $\pm 1.5\,\%\,\mathrm{dec}^{-1}$, the largest differences occur at the stratosphere. The trends calculated from the O3S-DQA corrections (in grey in Fig. 10) are lowest at all altitudes, maximal by $3\,\%\,\mathrm{dec}^{-1}$ in the (upper) stratosphere in comparison with the operational corrections.

The two different correction methods for the De Bilt profiles (in cyan and blue in Fig. 10) now have very similar stratospheric trend profiles, also in the upper and lower stratospheric parts, in contrast to the period 1993-2014 shown in Fig. 9. The differences in trends are larger in especially the upper troposphere, even up to $4\,\%\,\mathrm{dec}^{-1}$, due to the differences in background current treatment before November 1998. If we consider only the period from 1999, the trend differences between the two corrections at De Bilt nowhere exceed $1\,\%\,\mathrm{dec}^{-1}$. For the 1997-2014 period, at all altitude levels, the O3S-DQA corrections result in higher relative trends for De Bilt, exactly the opposite as for Uccle. As a consequence, only in the lower stratosphere and in the lower part of the free troposphere, the O3S-DQA corrections bring the Uccle and De Bilt trend estimates closer to one another (compare the grey and blue lines in Fig. 10). However, throughout whole the vertical profiles, the O3S-DQA trends for Uccle and De Bilt lie within each error bars, and could therefore considered as not significantly different.

The O3S-DQA trends at Uccle and De Bilt are also only in the troposphere significantly different from zero for the considered time period. The PRESTO correction at Uccle on the other hand (in blue), lead to significant positive trends from about 5 to

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15 km above the tropopause (error bars are not shown here), hence also at the altitudes of the ozone maximum. The finding that the applied correction method determines if the ozone trend is significantly positive or not is important in the present day ozone research. Indeed, the beginning of the period 1997-2014 coincides with the mid-latitude stratospheric peak values of the Equivalent Effective Stratospheric Chlorine (EESC) abundance (see e.g. Figure 1-22 in WMO, 2014). The EESC is a sum

5   of chlorine and bromine derived from ODS tropospheric abundances weighted to reflect their potential influence on ozone. As, by the end of 2012, the EESC has already returned $38-41\%$ from its peak value (WMO, 2014), a major issue in current ozone research is if the onset of ozone recovery can be detected. Our study demonstrates that, at least for measurements with ozonesondes, caution is needed before qualifying an even significant ozone increase as the onset of ozone recovery.

5   Conclusions

10 For the nearby stations of Uccle and De Bilt, we calculated average profiles and vertical trend estimates from both the operational and internationally agreed O3S-DQA corrections. Because typical horizontal ozone correlation lengths are generally much longer than the distance between both stations, except in the boundary layer, and because the time separation between the launches at those stations is at most one day, the comparisons of the average profiles and trends enable us to investigate the impact of the correction strategies on the ozone profiles and resulting trends.

15   In Uccle, where the time series is built up with both BM and ECC ozonesondes, the main feature of the operational PRESTO correction is the combination of a pressure and temperature dependent pump efficiency correction with the total ozone normalisation. For the BM 1969-1996 time period, the operational corrections result in a relative ozone increase between 20-30 % in the average profile with respect to the standard pump efficiency corrections, due to the typical BM response being only equivalent to about 80% of the actual ozone amount. Because of the correction for SO_2 interference, this relative ozone partial

20 pressure difference even increases to about 40 % in the lower tropospheric layers. For ECC ozonesondes (1997-2014), the different correction strategies produce average profiles within $\pm 2\%$ of this reference. In particular, the O3S-DQA correction for ECC ozonesondes adds about 2 % ozone at the tropospheric levels compared to the operational correction, whereas above the ozone maximum, the reverse is true, but now with an amount around 1 %. For the De Bilt time series (1993-2014), the O3S-DQA average profile has smaller ozone amounts at all altitude levels. Here, the largest relative differences are obtained in the

25 UTLS (about 15 %) in the period before November 1998, when different background measurement operations and corrections have been applied for both corrections.

When comparing the average profiles of Uccle and De Bilt, we conclude that higher tropospheric ozone concentrations are measured at Uccle than at the Bilt, which might be ascribed to both natural (more polluted area at Uccle) and instrumental (higher background currents subtracted at De Bilt) origins. In the lower stratosphere, higher ozone amounts are present in De

30 Bilt, while the opposite is true above the ozone maximum. At burst altitudes, the relative differences between the Uccle and De Bilt ozone partial pressures can amount to up to 10 %, independently of the used correction method. This reason for these larger discrepancies is not clear to us. As a matter of fact, we found that the O3S-DQA corrections for ECC ozonesondes at both sites effectively reduce the relative differences between Uccle and De Bilt only in the lower stratosphere (below the ozone

maximum). On the other hand, the operational PRESTO correction method at Uccle is able to reduce the relative stratospheric ozone differences with De Bilt for the Uccle BM sondes (during the period 1993-1996) from about 20-30 % for the standard pump corrections to less than 5 %.

The used correction method has also a large impact on the derived trends. For the entire Uccle time period, the operational corrections result in a fairly constant and consistent trend over the troposphere ($+2$ to $+3\,\% \, \mathrm{dec}^{-1}$) and stratosphere ($-1$ to $-2\,\% \, \mathrm{dec}^{-1}$), which is a serious reduction of the overall positive trends estimated from the profiles corrected only by the standard pump efficiency profiles. In particular, the correction for the SO_2 interference is responsible for a reduction of the boundary layer ozone trends by about half. For the operational corrections during the entire De Bilt period, the De Bilt trends in ozone concentrations are more modest than the corresponding Uccle trends, except in the boundary layer. As larger (positive) trends emerge at all altitudes from the O3S-DQA corrections for De Bilt, the O3S-DQA trend profile follows closer the Uccle trend profile in the stratosphere. However, in the free troposphere, the O3S-DQA trends increased too much and now exceed the Uccle trends. Finally, for the period 1997-2014, the Uccle trends calculated from the O3S-DQA corrections are lower at all altitudes, maximal by $3\,\% \, \mathrm{dec}^{-1}$ in the (upper) stratosphere, in comparison with the operational correction trends. For De Bilt, the opposite is true, with the largest differences, by about the same amount, in the upper troposphere. Therefore, only in the lower stratosphere and in the lower part of the free troposphere, the O3S-DQA corrections bring the Uccle and De Bilt trend estimates closer to one another.

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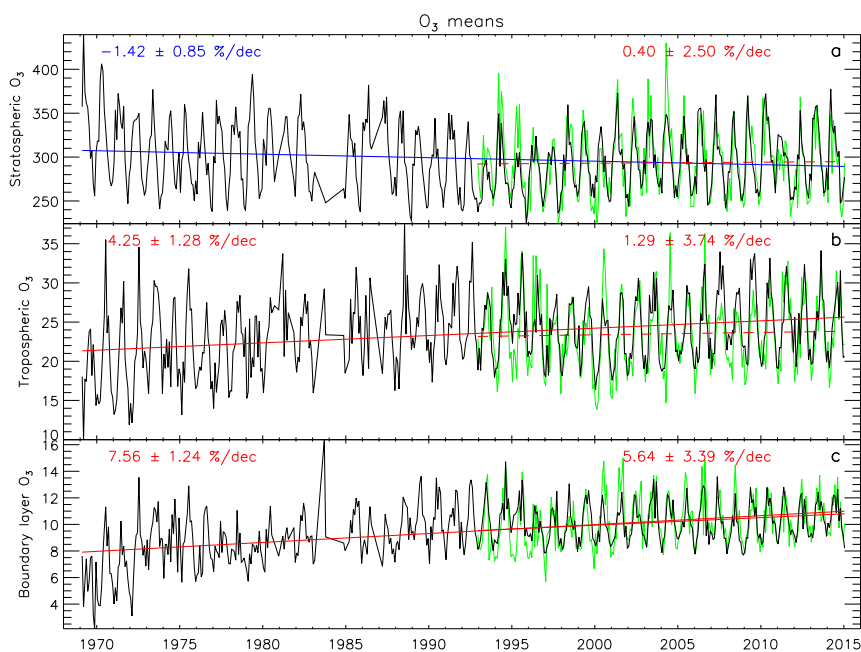
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Figure 1. Monthly means of integrated ozone amounts above Uccle (black) and De Bilt (green) for different parts in the atmosphere: (a) stratosphere ($h >$ tropopause height), (b) free troposphere (3 km $< h <$ tropopause height), (c) boundary layer ($0 - 3$ km). The linear regression lines are also drawn, and their slopes are used to calculate the trends (in $\%/\mathrm{dec}$, left for Uccle, right for De Bilt), together with their 2σ uncertainty estimates. Red lines are used for positive trends, blue lines for negative trends. A full line denotes a statistically significant trend. The statistical significance of the trends is investigated by Spearman's test.

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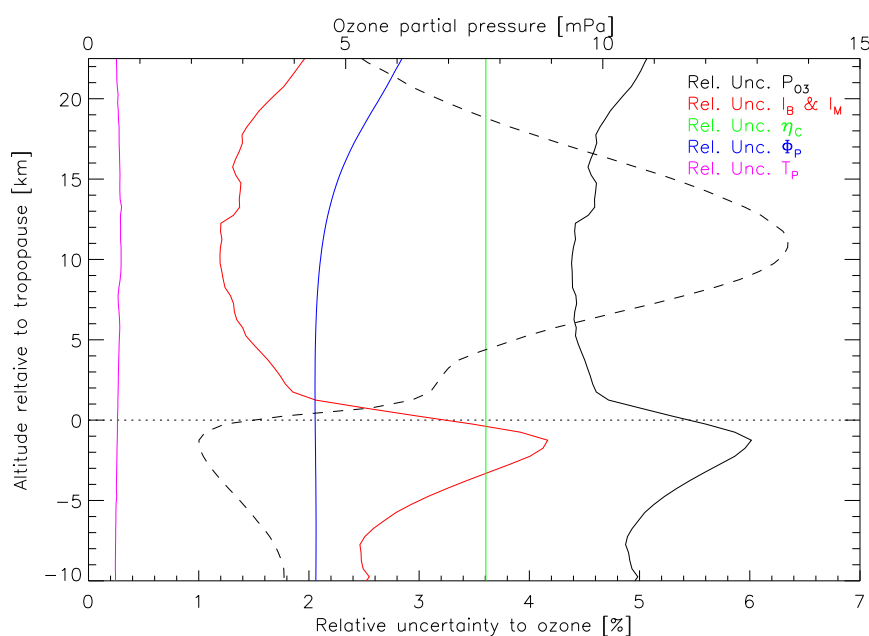
Figure 2. Relative uncertainty of the ozone partial pressure and the contributions from the individual uncertainties of the different instrumental parameters like measured cell current I_M, background current I_B, conversion efficiency η_c, pump flow rate Φ_p, and pump temperature T_P as a function of altitude relative to the tropopause. These vertical profiles are the average profiles of these uncertainties, calculated for all ECC O_3 profiles at Uccle. The black dashed line is the mean O_3 profile at Uccle, in units of mPa (see upper scale), for the same period $(1997-2014)$. This figure has been adapted from Fig. 3-1 in Smit et al., 2011 for the Uccle case.

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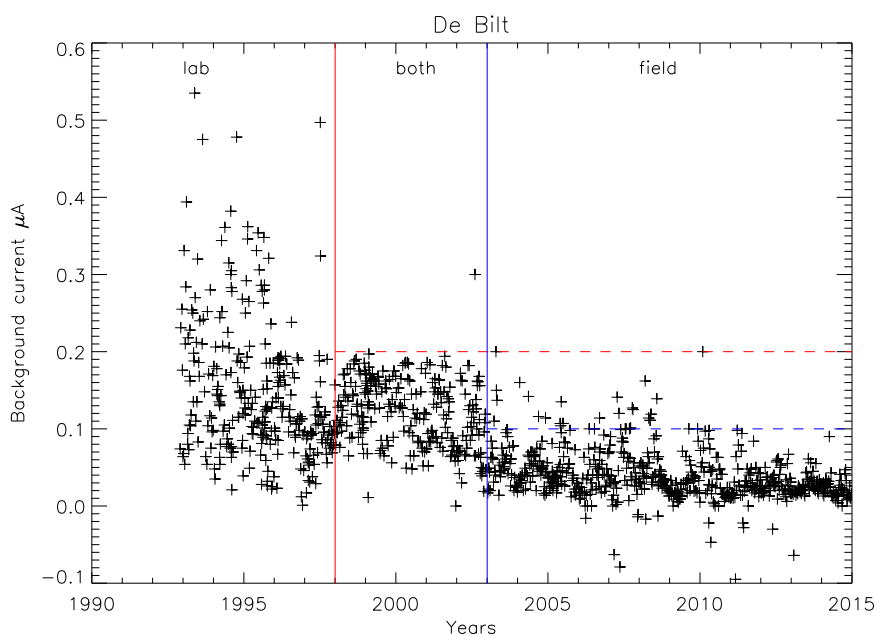
Figure 3. Time series of background current values measured after exposure to ozone at De Bilt. The vertical lines denote the periods from which the upper limits for the BGC (horizontal lines, same colour coding) were imposed. For more details, see the text.

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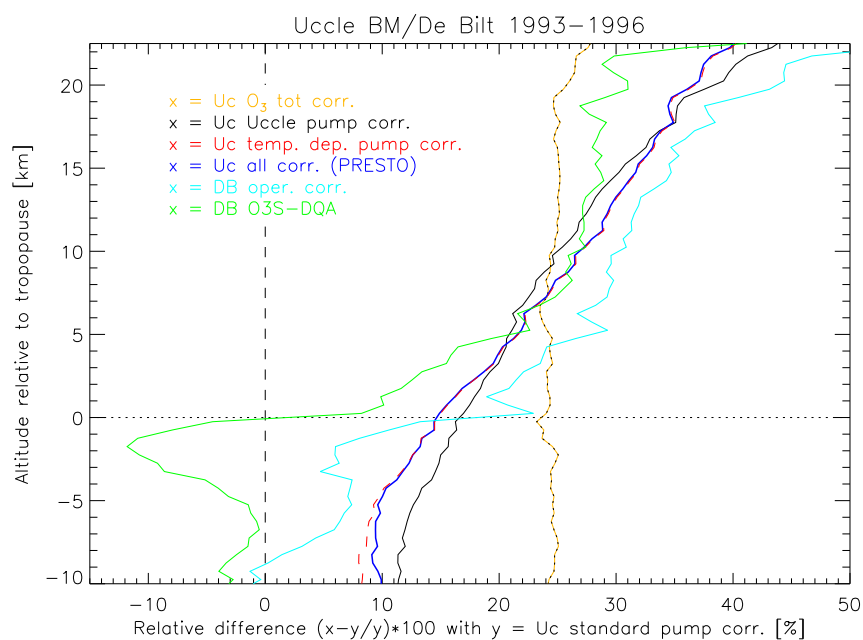Open Access

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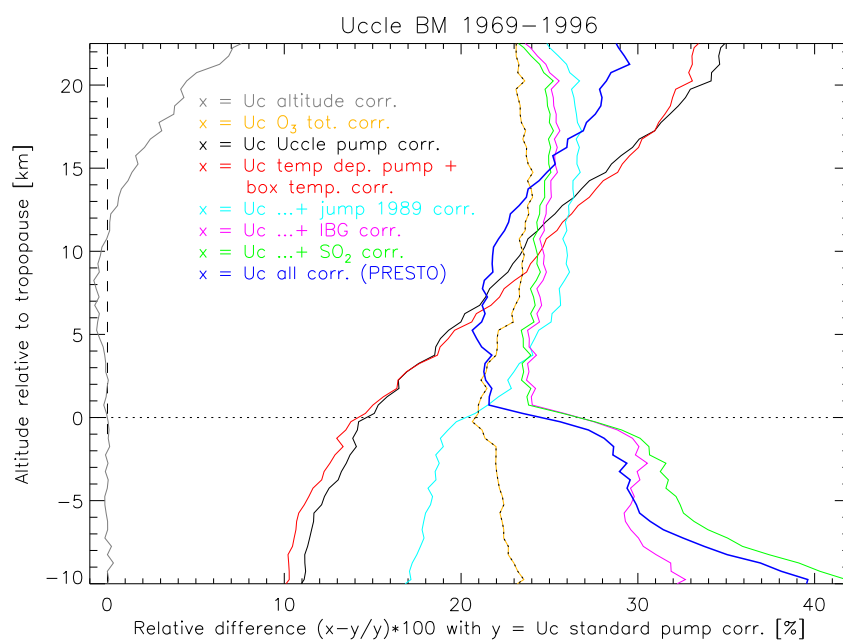
Figure 4. Relative differences of the average Uccle ozone profile calculated for different correction strategies with respect to the average ozone profile obtained by applying the standard pump efficiency correction factors. The average ozone profiles are calculated for the entire BM 1969-1996 observation period, and in layers of 0.5 km height, relative to the tropopause height. For more details, see the text.

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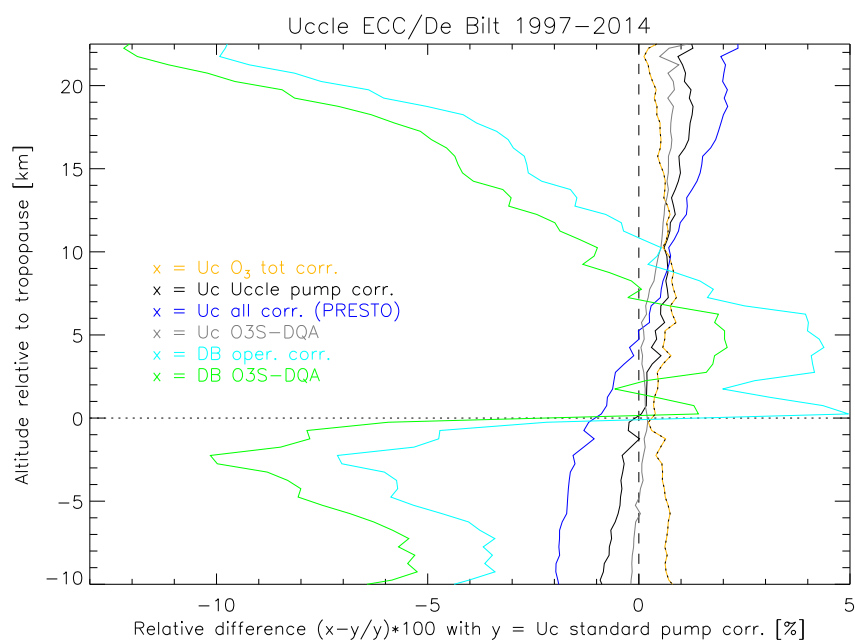
Figure 5. Relative differences of the average Uccle and De Bilt ozone profiles calculated for different correction strategies with respect to the average Uccle ozone profile obtained by applying the standard pump efficiency correction factors. The average ozone profiles are calculated for the 1997-2014 observation period, when both stations used ECC ozonesondes, and in layers of 0.5 km height, relative to the tropopause height.
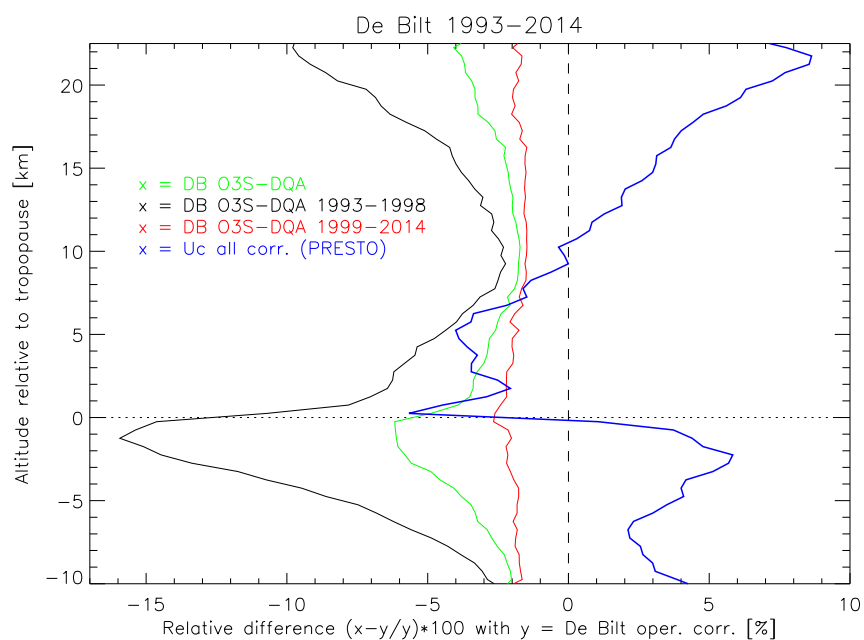
Figure 6. Relative differences of the average De Bilt O3S-DQA corrected ozone profiles calculated for different periods with respect to the average De Bilt ozone profile obtained by applying the operational corrections for the same periods. The relative difference of the average Uccle PRESTO corrected profile with the operationally corrected De Bilt profile for the 1993-2014 period is also shown.

Figure 7. Relative differences of the average Uccle and De Bilt ozone profiles calculated for different correction strategies with respect to the average Uccle ozone profile obtained by applying the standard pump efficiency correction factors. The average ozone profiles are calculated for the 1993-1996 observation period, when BM sondes were in use in Uccle, and in layers of 0.5 km height, relative to the tropopause height.

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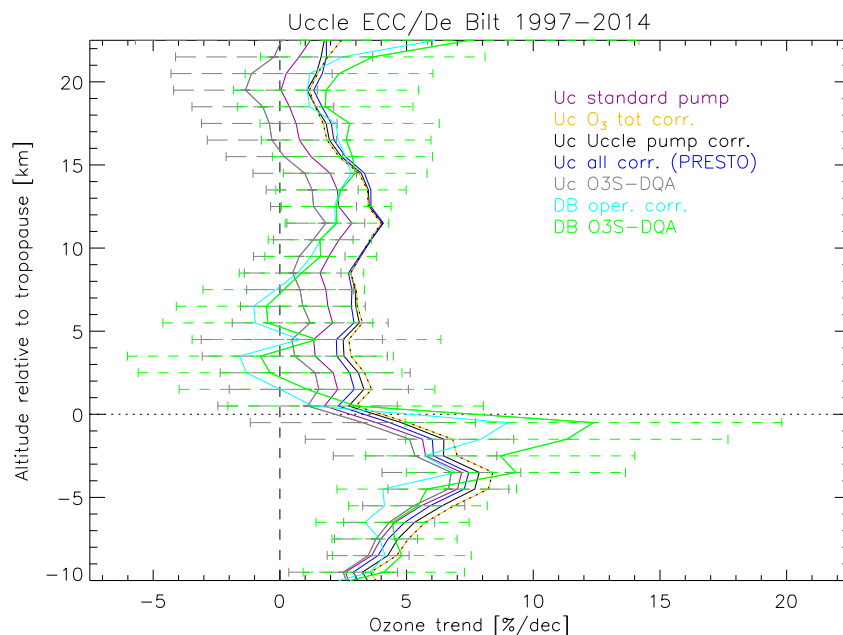Techniques

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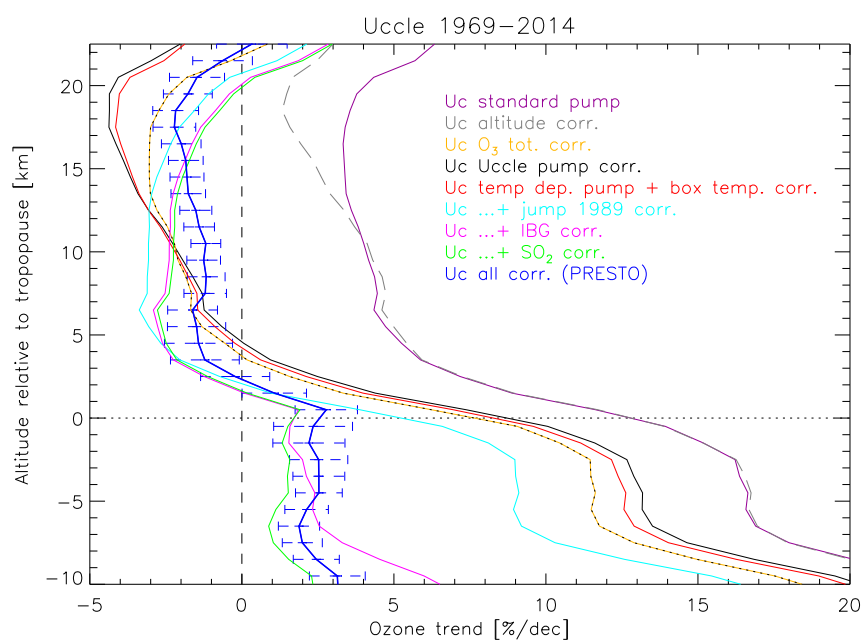
Figure 8. Vertical distribution of the linear relative trends for different correction strategies applied to the Uccle ozone data for the entire time record. The trends are estimated for layers of 1 km height, relative to the tropopause height. The error bars denote the 2σ standard errors of the linear regression slope determination after applying all profile corrections and can be considered as a rough estimate of the trend uncertainty.

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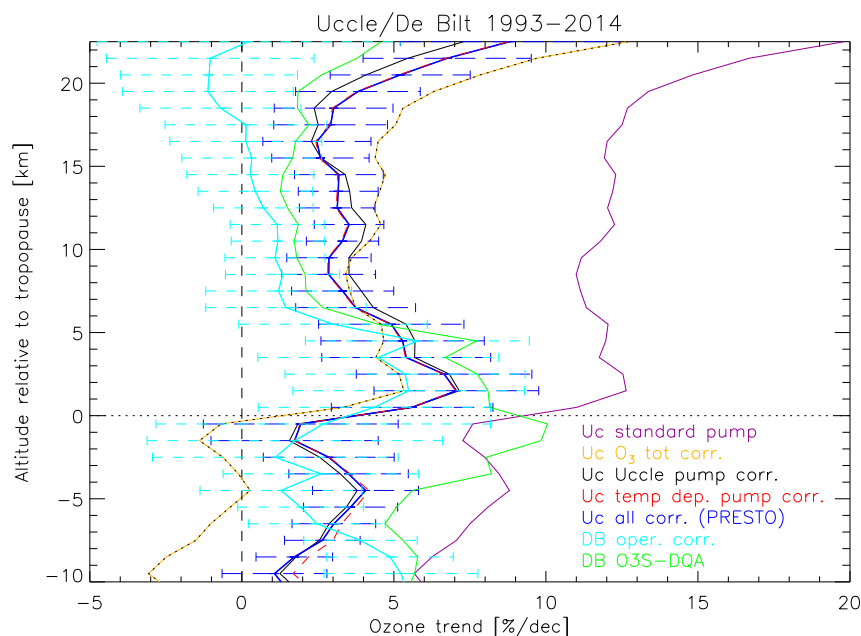
Figure 9. Vertical distribution of the linear relative trends for different correction strategies applied to the Uccle and De Bilt ozone data for the entire De Bilt time record. Trends and error bars are calculated as in Fig 8.

Figure 10. Vertical distribution of the linear relative trends for different correction strategies applied to the Uccle and De Bilt ozone data for the 1997-2014 time period, in which ECC sondes were used at both stations. Trends and error bars are calculated as in Fig 8.

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Table 1. Overview of the properties of the ozonesonde measurements at Uccle and De Bilt. This table is adapted from Van Malderen et al. (2014).

	Uccle	**De Bilt**
coordinates	$50°48'$ N,$4°21'$ E, 100 m asl	$52°10'$ N,$5°18'$ E, 4 m asl
first launch	Jan 1969	Nov 1992
average frequency	3/week	1/week
sonde type	Brewer-Mast ENSCI ECC Z	SPC ECC 5A SPC ECC 6A
switch date	1 Apr 1997	24 Jul 1997/1 Mar 2001
RS type	VIZ/RS80/RS92	RS80/RS92
switch dates	Jan 1990/Sept 2007	Nov 2005
ECC SST	0.5	1.0
solution amount	3.0 cc	2.5 cc 3.0 cc (from 23 Nov 1994)
location T_p sensor	in the box (from 1 Jan 1990) in the pump (since Dec 1998)	in the box in the pump (from 19 Nov 1998)
I_B measurement	in laboratory before exposure to O_3	in laboratory/at launch field after exposure to O_3

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Table 2. Overview of the most important properties of the different correction strategies applied at Uccle and De Bilt. More details can be found in the text and in Smit et al. (2012) for the O3S-DQA corrections.

	Uccle		De Bilt	
	operational (PRESTO)	O3S-DQA (only ECC)	operational	O3S-DQA
η_C correction	no (= 1)	no (= 1)	no (= 1)	yes, for 2.5 cc solution
Φ_p efficiency correction	T, p dependent see Eqs. 4 and 6	p dependent	p dependent	p dependent humidification effect corr.
correction profiles C_{PF}	measured in-house De Backer et al. (1998b)	Komhyr et al. (1995)	Komhyr (1986) Komhyr et al. (1995) (from Nov 1998)	Komhyr (1986)
T_p value used	(estimated/measured) T_{Box} (BM) measured T_{Box}/T_p (ECC)	measured T_{Box}/T_p	measured T_{Box}/T_p	measured T_{Box}/T_p
corrections		$T_{Box} \longrightarrow T_p$ $T_p \longrightarrow$ "truest" T_p		$T_{Box} \longrightarrow T_p$ $T_p \longrightarrow$ "truest" T_p
I_B subtraction	constant	constant	pressure-dependent constant (from 5 Nov 1998)	constant
total O_3 normalisation	yes, pressure-dependent	no	no	no
SO_2 correction	yes(BM)/no(ECC)	no	no	no
altitude correction	yes(BM:VIZ RS)/no(ECC)	no	no	no