

Dear editor,

please find hereafter our responses to the reviewers on the general and specific comments that they have brought up.

5 We have discussed and answered all their comments, and tried our best to implement them in the manuscript. However, the major concern of reviewer 1, about the current design of the manuscript and the suggestion to resubmit the manuscript in three separate pieces of work, also brought up in his/her quick review report, we could not implement. After this suggestion in his/her quick review, we asked you for feedback and the current organization and design was proposed/accepted by you. Therefore, 10 we decided to continue on the track chosen at that time, and not to go on a completely new track. In our response to this reviewer, we informed her/his about this communication between you and us. However, as a compromise and as a sign of our good will towards this reviewer, we moved some of the sections that were suggested by the reviewer to be treated in a separate work, to two appendices. Appendix A now describes the total ozone column trends of the co-located spectrometers at Uccle, to 15 which the ozonesonde profile data is normalized, and Appendix B treats the Uccle surface ozone trends from measurements by an ozone monitoring instrument and the ozonesondes.

We thank you for your time and look forward to the next step in the peer-review process!

20 Sincerely,

Roeland Van Malderen, on behalf of the authors

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Response to Reviewer 1 of Van Malderen et al. – 50 years of balloon-borne ozone profile measurements at Uccle, Belgium: short history, scientific relevance and achievements in understanding the vertical ozone distribution.

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In this response, we included the reviewer comments in black. Specific comments are numbered. Our response are written in red, with the modifications in the manuscript in *red italic*.

General Comments

40 In this manuscript, the authors provide an overview of the 50 year history of the ozonesonde program at Uccle.

The dataset is certainly extremely valuable to the field of atmospheric science, and everyone responsible should be congratulated on maintaining a high quality measurement program over such a long period of time. This is an impressive achievement!

45 The manuscript describes, in a medium level of detail, previous work that has been done to adjust for inhomogeneities and contamination, after which the authors present trends in total ozone at Uccle measured by ground-based instruments, a height resolved trend analysis of the Uccle ozone record in the stratosphere based on multi-linear regression, and separately a trend analysis for the troposphere, a trend analysis of tropospheric folding events and a trend analysis of surface ozone. The analyses also make use of a wide variety of other data sources to compare to the Uccle ozonesonde record. In
50 the final section satellite overpasses are compared for three different instruments (or series of instruments) – GOME2, AURA-MLS and AURA-TES.

In my view, these components in themselves are all very worthwhile and very appropriate subjects for ACP.

55 Unfortunately I must state however, that I have major concerns with the design of the manuscript in its current form. Because of the very wide scope, none of the subjects are presented in sufficient detail to be of much use to the specific communities who are interested in them. In my view it would be preferable to resubmit the manuscript as three separate pieces of work, each one with a more specific focus allowing greater depth. Alternatively, the number of topics could be reduced to allow a more satisfactory level of detail to be given.

60 I appreciate that the authors have deliberately chosen to present something more along the lines of a summary of the many applications of the dataset.

Unfortunately, in my opinion, this decision is not optimal for the benefit of the readers of ACP.

(Please be clear I am not intending to be in any way negative about the value of the dataset or the scientific competence of the authors, merely the structure of the submitted manuscript).

65 Old work is covered in too much detail, but still not in sufficient detail to enable it to be understood without reference to the original papers anyway. On the other hand, new work is "described" without enough detail provided however for the reader to make a proper assessment of its value. This is most evident in the discussion of tropospheric fold occurrence.

70 The major contribution of the work I see as being the assessment of long-term trends in the stratosphere and free troposphere (sections 4.2 and 4.3), at very high vertical resolution.

I would prefer these sections to be significantly expanded and sections 1, 2 and 3 shortened. I would also consider removing 4.1 and 4.4 and treating these topics in better detail in separate works. In my view section 4.1 is too superficial and section 4.4 too inconclusive in their current forms. The use of other data sets for comparison (eg De Bilt ozonesondes, Frankfurt IAGOS, surface ozone monitoring) is very interesting but does tend to clutter the analysis and obscure the conclusions.

Thank you very much for your honest review report, and for pointing your finger to some very important issues on specific analyses.

80 Concerning your remarks about the design of the manuscript. We are aware that, already in your quick review report, you raised the same concern of having too much specific topics, which had **partially** been picked up by the handling associate editor. Therefore, we contacted first the editor for additional advice before starting a complete reorganization of the manuscript. We repeat here again our arguments and inspiration behind the manuscript.

First of all, the two following ACP papers inspired us for our approach (presentation and organisation of the manuscript):

- 85 • Staehelin, J., Viatte, P., Stübi, R., Tummon, F., and Peter, T.: Stratospheric ozone measurements at Arosa (Switzerland): history and scientific relevance, *Atmos. Chem. Phys.*, 18, 6567–6584, <https://doi.org/10.5194/acp-18-6567-2018>, 2018.
- 90 • De Mazière, M., Thompson, A. M., Kurylo, M. J., Wild, J. D., Bernhard, G., Blumenstock, T., Braathen, G. O., Hannigan, J. W., Lambert, J.-C., Leblanc, T., McGee, T. J., Nedoluha, G., Petropavlovskikh, I., Seckmeyer, G., Simon, P. C., Steinbrecht, W., and Strahan, S. E.: The Network for the Detection of Atmospheric Composition Change (NDACC): history, status and perspectives, *Atmos. Chem. Phys.*, 18, 4935–4964, <https://doi.org/10.5194/acp-18-4935-2018>, 2018.

100 In this sense, we believe that this manuscript has only one focus or topic, i.e. the Uccle ozonesonde measurements, with the aim to demonstrate the scientific relevance of and the major achievements with this dataset. As ozonesondes are still the only technique able to measure the ozone concentrations all the way up from the surface to the middle stratosphere with very high accuracy and vertical resolution, they have many application areas in which they are crucial: (i) long-term variability in stratospheric and tropospheric ozone, (ii) backbone for satellite validation, with the satellites mostly measuring ozone only in stratosphere or upper troposphere, (iii) process studies in stratospheric-

105 tropospheric exchange, and chemical production/destruction of ozone. We believe that the strength and uniqueness of the ozonesonde measurements, and in particular of our long-term and very dense Uccle dataset, lies precisely in combining all those different aspects of ozone research. To our opinion, focusing on only one aspect (e.g. trends) really underrate the relevance of our dataset. And therefore we believe that it is better to see this manuscript as a whole, rather than break it in pieces.

These were the 2 options we proposed to the editor for a new organization of the paper:

- 1) The current one, which was already a reorganization and more focused one as the original manuscript (to which your quick report was addressed).
- 110 2) In case the proposed option one still contains too much topics according to you, we propose to split up the paper in two (and not three) parts, namely “50 years of balloon-borne ozone profile measurements at Uccle, Belgium. Part 1: Major achievements for stratospheric ozone” & “50 years of balloon-borne ozone profile measurements at Uccle, Belgium. Part 2: Major achievements for tropospheric ozone”. The exact titles of the different parts still need to be better determined, but the idea behind is to keep the focus on the measurement dataset by publishing the analysis as a series of two papers. The stratospheric part would then contain most of the short history, the total and stratospheric ozone trends and the AURA-MLS (stratospheric ozone satellite retrieval) comparison. The tropospheric part would contain some aspects of the short history (e.g. correction for SO₂), the tropospheric and surface ozone trends (with increased frequency of tropopause folds), and the AURA-TES (tropospheric ozone satellite retrieval) comparison. In this sense, both the stratospheric and tropospheric ozone community are served separately, the two papers will be shorter and lighter than the combined one and we keep only two topics in each of the more focused papers. If this option is the preferred one, we will submit the two papers at the same time, because they belong together. Of course, there will be inevitably cross-references between both manuscripts.

125 The editor had a preference for the current organization (option 1) of the manuscript, which was then accepted in ACPD.

130 From your comments, we understand that you believe that the assessment of long-term trends in the stratosphere and free troposphere (sections 4.2 and 4.3), at very high vertical resolution, is the major contribution of the work. However, you mention that this contribution is cluttered by taking the total and surface ozone trends at Uccle and the De Bilt ozonesonde and IAGOS Frankfurt measurements into account. We think that the opposite is true, and that these measurements underline the broader perspective, relevance and (spatial) representativeness of the Uccle ozonesonde measurements. A study presenting the vertical ozone trends at one single location only is less relevant than when these trends are compared with nearby, independent measurements. Nevertheless, we moved the Dobson/Brewer total ozone trends, making use of the LOTUS multiple linear regression model, at 135 Uccle to Appendix A, and the surface ozone trends at Uccle to Appendix B.

We also do not fully agree with you that sections 1, 2, 3 contain too much detail and section 4 (the trends) not enough. For instance, in the section on the history, we only shortly describe the three challenges the Uccle time series has been faced with (among others) that made the fame of the Uccle time series (frequency response analysis, SO₂ correction, BM to ECC transition) and are still very relevant for current new developments in ozonesonde data processing (frequency response analysis) or essential when calculating trends with the ozonesonde record over the entire time series (BM to ECC transition) or over subperiods (SO₂ correction). As all those analyses have already been published elsewhere, we give all the needed references for a reader that wants to have all the details. We only shortly describe the research and the main outcome of these past studies for this paper. We also do not fully understand the criticism that new work (the trends in section 4) is only described without enough detail and that the analysis is inconclusive or too superficial, especially taking into consideration that you proposed not including the De Bilt and IAGOS Frankfurt measurements. Every trend result is put in a broader perspective and compared to the most recent findings in other studies and trend assessments (often based on satellite ozone retrievals), while the interpretation is fed by references to recent model results. To give some examples: for the explanation of all specific features in the total ozone time series, we refer to Fig. 2 in Weber et al. (2018), we compare the lower-stratospheric ozone trends with the Ball et al. (2018, 2019, 2020) studies, we refer to all relevant surface and tropospheric ozone studies above Europe and confront their findings with ours. Indeed, some findings are inconclusive (why is the total ozone recovery at Uccle almost fully accomplished in contrast to global total ozone recovery? Why do we see the opposite lower-stratospheric ozone trends as in the Ball et al. papers? What processes are responsible for the surface/tropospheric ozone trends at Uccle?), but this is related to the fact that we only treat with great detail one station (first two questions raised), and that findings are still inconclusive on the global scale (third question raised). However, in answer to your specific comments, we tried to make the analysis more detailed, conclusive, and in-depth, where possible.

Specific comments

1. Line 49: You should also quote WMO 2018 Chapter 3 (and chapter 4 if you want to talk about recovery of Antarctic ozone, seeing you have mentioned it just earlier).
Done as suggested.
2. Line 53 A paper about SHADOZ is not appropriate for a statement about the radiative forcing of ozone – the best reference for this would be the IPCC or the major papers they have used.
We changed it to “estimated to have contributed ~20% as much positive radiative forcing as CO₂ since 1750 (IPCC, 2013)”, with text directly taken from the IPCC report.
3. Lines 50-52 You should be careful about the radiative effect of ozone at different levels in the atmosphere.
We specified that ozone can act as a greenhouse gas at certain altitudes.

- 180 4. Lines 61-64 This is confusing for the reader, because in the first sentence it sounds like "electrochemical ozonesondes" is being used broadly in a way which includes Brewer-Mast, but then in the second sentence you distinguish ECC ozonesondes from Brewer-Mast.
We believe the text is correct. Both BM and ECC sondes are electrochemical sensors: BM uses electrodes of different metal, and ECC (electrochemical concentration cell) sondes use different concentrations. We specified this further in the text.
- 185 5. Lines 67 Explain to the reader why you say "nowadays".
We added "*Before the digital sounding systems era the vertical resolution was less due to the manual sampling technique by the operator, providing only measurements at significant levels.*"
- 190 6. Lines 68-70 I am sure my Australian colleagues would also like you to mention Aspendale, which has data in WOUDC from as early as 1965 and has continued to the present, but with the launching site having moved somewhat (Aspendale – Laverton – Broadmeadows).
Thanks for pointing this out. We added this information, and we also included the Japanese sites Tateno (Tsukuba) and Sapporo, that initiated ozone soundings in 1968 and 1969, respectively. We changed the text into "*Regular measurements with ozonesondes started in the second half of the 1960s at a few sites: in 1965 at Aspendale (Australia, but moved to other suburbs of Melbourne thereafter, i.e. Laverton and Broadmeadows), in 1966 at Resolute Bay (Canada), in 1967 at Hohenpeissenberg (Germany), in 1968 at Payerne (Switzerland) and at Tateno (Tsukuba, Japan), in 1969 at Uccle (Belgium) and Sapporo (Japan), and in 1970 at Wallops Island (USA).*"
- 195 7. Line 82 I don't like "our" here – this makes it appear the work is intended for an internal audience.
We agree, we changed "*our*" to "*the*".
- 200 8. Line 82 This is probably out of scope for the current work, but I would be very curious to know if the high frequency of launches at Uccle can be shown to lead to better results, compared to weekly or fortnightly or monthly launches? (You could easily enough calculate the uncertainties on the trends using only a subset of the data).
Note that in the manuscript, we already made a reference to the recent work by Chang et al. (2020), who, based on a sensitivity analysis with IAGOS profiles above Europe, determined that an optimal sample frequency of 14 profiles per month is required to calculate tropospheric ozone trends with their integrated fit method (and about 18 profiles a month when this method is not used).
In the figure 1 here below, we calculated the vertical ozone trends (both of the troposphere and stratosphere) for the time periods 1969-2018 and 2000-2018 with the normal Uccle sounding frequency of 3 times/week and with reduced frequency (once a week). We used here a simple linear regression model for the fit, as both stratospheric and tropospheric ozone trends have been estimated. From this figure, it should be clear that the differences between both the trends estimates and their uncertainties are small when derived from the entire and reduced frequency datasets, if the 50 year time period is considered. However, for the ozone recovery time period (after 2000), large trend differences occur, especially in the important lower-stratospheric region.
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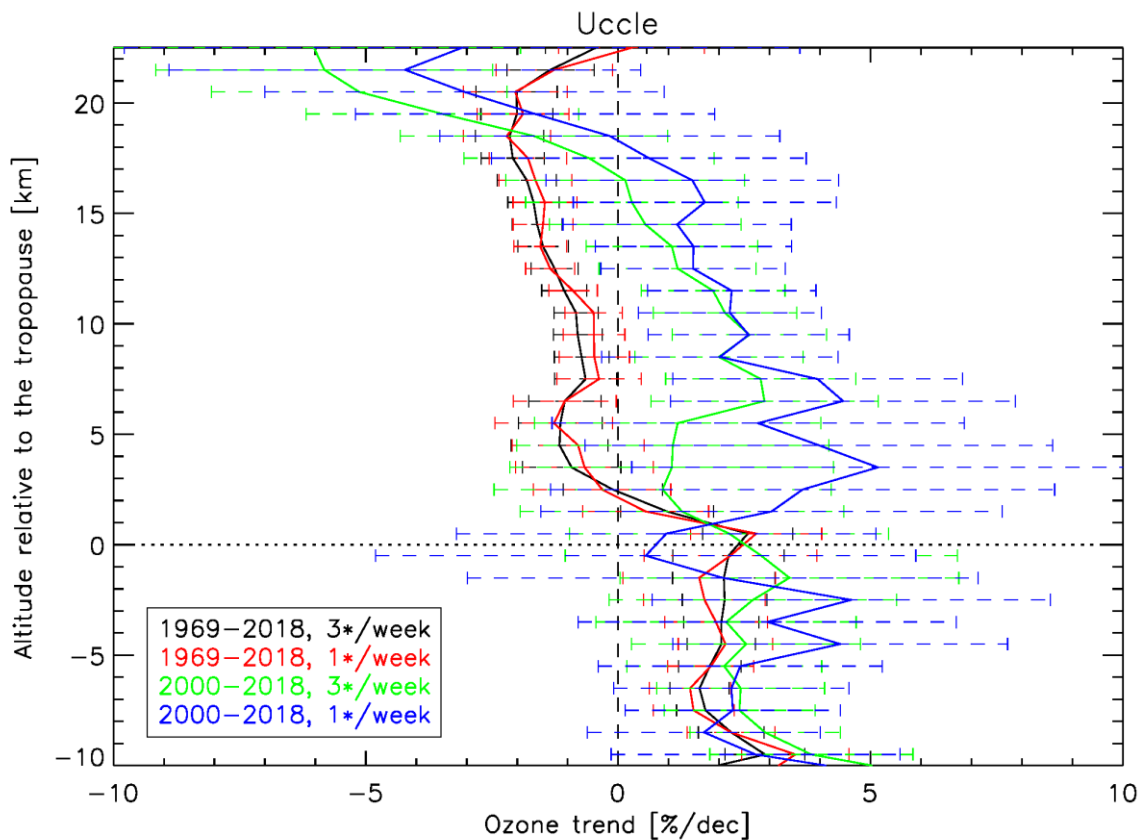


Figure 1: Vertical distribution of trends of ozone concentrations at Uccle for different periods and for different sounding frequencies (3 times a week and once a week). The trends and their 2-sigma error uncertainties are calculated a simple linear regression model.

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9. Line 91 Change "has been" to "was"
Done as suggested.

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10. Line 114 – It sounds like the ozone profile is adjusted to agree with the total ozone measurement - You should state this clearly to the reader. (Particularly because it is the less common practice at ozonesonde stations these days).

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This was (and is in case of Hohenpeissenberg) common practice for BM sondes, and is essential for these ozonesondes, as they are only sensitive to about 80-90% of the measured total ozone amount during a flight. This is explained in Sect. 2.2.3, but we specified this here as well by adding "(essential for BM ozonesondes, see Sect. 2.2.3)". In our operational correction/processing of the ozonesonde data, called PRESTO, we indeed apply a "pressure and temperature dependent total ozone normalization" for both the BM and ECC ozonesondes. A total ozone normalization is not needed for ECC ozonesondes (around 100% sensitive to the measured total ozone column amount during a flight), and it is advised not applying it (in e.g. the Ozonesonde Data Quality Assessment or O3S-DQA homogenization guidelines). However, when merging the BM and ECC time series for e.g. estimating trends, we rely on the PRESTO

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245 corrections, that have proven to ensure the homogeneity of the entire time series (see Sect. 2.2.3). A comparison between the PRESTO and O3S-DQA corrections for the Uccle ECC time series is available in Van Malderen et al. (2016). All this information is provided further in the manuscript.

250 11. Lines 119-135 Without more detail, it is not very convincing that this approach has worked very well.

255 We included some specifications (see next point). In the paper about the frequency response of BM sondes, the whole procedure about the measuring procedure for the determination of the frequency response by means of a Fourier transform as well as the deconvolution procedure are explained in detail. The example of the ascent and descent data of an ozone sounding before and after deconvolution shows the effectiveness of the method. Therefore it is hard to see what additional detail could make it more convincing that this approach works very well, especially with the more recent publications you are referring to in your next point, in mind. The De Muer and Malcorps (1984) paper was really the pioneer for the current ideas about convolution and deconvolution of the ozonesonde measurements (Vömel et al., 2020, and Tarasick et al., 2021).

260 12. Lines 119-135 I would also be curious to know if the results agree with more recent work, eg Tarasick 2020

265 We rewrote large parts of this section to make a more direct reference to this recent work: *“They found three different time constants: (i) a first-order process with a time constant of about 17 to 25 s (depending on the solution temperature) caused by the formation of iodine in the solution, (ii) a time constant of 7s, likely to be caused by the diffusion of iodine molecules to the platinum cathode, and (iii) a time constant of about 2.8 min that was explained by another diffusion process, i.e. an adsorption and subsequent desorption process of ozone at the surface of the air-sampling system. The slow first-order process with a time constant of about 20-25 minutes (found by Salzman and Gilbert (1959) and taken up by Vömel et al., 2020, and Tarasick et al., 2021) could not be identified, probably because the impact of this process for a 0.1% KI solution would be too small (being 10% of the fast process for a 1% KI solution), as noted in De Muer and Malcorps (1984).”*

270 13. Lines 139 – 163 This was all very important work, but the problem is that the reader has to take everything on trust, namely the effect of SO₂ on the Dobson, the effect on the ozonesonde, and the assumptions about the SO₂ concentration. You don't even show, for example, that when you put all this together the Dobson and the ozonesonde are brought into agreement. Thus, I find this moderate level of detail quite frustrating. I would much prefer you be briefer, and just report the outcomes of the historic work (with full references of course).

280 Yes, you are right that we rely on the reader consulting the references (we provide full references here) if he/she wants to find out more details on the effect of SO₂ and how the correction works. But, as we focus here on the ozonesonde measurements, we do show the impact of the SO₂ on an ozonesonde profile in Fig.1 and Fig. S1, and on the associated vertical ozone trends in Fig. S3 (as well as the impact of the SO₂ correction). Also the time variability of the SO₂ amounts at Uccle is shown in Fig. S2. We restructured the text in this section, to better put the emphasis of the SO₂ impact on ozonesonde measurements, and presented the SO₂ correction for Dobson total ozone measurements as a “supporting tool” for the correct

290 evaluation of vertical tropospheric ozone trends for the 1969-1996 period, for which the
combination of Brewer-Mast ozonesondes and a Dobson spectrophotometer for the necessary
total ozone normalization of the BM ozonesondes, has been used. We also now mention the
main idea/principle of the SO₂ corrections.

As to the question about the agreement between Dobson and ozonesonde data:
295 after any correction applied to each ozone sounding (such as the SO₂ correction), for Brewer-
Mast sondes it was common practice to apply a final normalization to the integrated ozone
amount (Dobson). This is a standard procedure that makes the SO₂ correction not relevant for
the agreement between the two corrected data sets.

300 14. Lines 163-185 Again, I this was very important work (and I think Uccle should be congratulated
for studying the effects of the change away from Brewer-Mast compared to many other stations
around the world which were not as careful) , however, the method is not actually explained
here, you just show the results (and reproduce an old figure). As above, I think you should just
mention this work and give the references.

305 Yes, you are right that not all the details of the method are actually explained here, only its main
principles. The details of the method are already available in other papers (De Backer et al.,
1998, De Backer 1999, and more recently also in Van Malderen et al., 2016), so there is no
need to describe the method in full detail again. We moved the figure to the supplementary
310 material (Fig. S4 now). But, as it is actually this (operational) method that has been used to
process the 50 years of ozonesonde measurements analysed in this manuscript (see Section
3), it is essential to describe the reason behind and the rationale of the method. We made some
small specifications/clarifications in the text and added the De Backer 1999 reference.

315 15. Line 208 For comparison with other work, it would be helpful to give the size of the altitude
correction specifically (ie not just the resulting difference in ozone).

We added "*sonde altitudes were too low up to 1000m at an altitude of 30km*".

320 16. Lines 221-223 How confident are you of the homogeneity of this time series? If you are going to
show this figure (Figure 3), you need to discuss the calibration history of the Dobson
(particularly in the pre-1977 era) and the transition from Dobson to Brewer.

For your comments 16 to 18, please note that we decided to move this section to an appendix.
We however still find it very relevant to include this section, as the Uccle ozonesonde time
series are normalized (with a pressure and temperature dependent procedure) to the total
ozone measurements with the Dobson and Brewer. Describing and explaining the vertical
325 stratospheric ozone trends without mentioning the total ozone trends does not seem to be
scientific "fair" to our opinion.

For this specific comment, we added the following sentence (and references): "*The calibration
history of the Dobson instrument is documented in De Muer and De Backer (1992) and the
transition to the Brewer instrument is described in De Backer and De Muer (1991). Both Brewer
330 instruments were recalibrated against the traveling standard Brewer instrument no. 17 in 1994
(no. 16 only), 2003, 2006, 2008, and against the travelling reference Brewer no. 158 since 2010
every second year. The stability of the instruments is also continuously checked against the co-
located instruments (with the Dobson no. 40 from 1991 until May 2009, between both Brewers
since 2001). Internal lamp tests are performed on a regular basis to check whether a Brewer
335 instrument is drifting. When instrumental drift is detected, it is corrected for.*"

17. Line 228 How can you be so confident about the attribution to volcanic eruptions? There are other large dips and several peaks as well of comparable size.

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We changed the text to “*Indeed, the episodes of enhanced stratospheric aerosol-related ozone loss after those major volcanic eruptions are confirmed by model results (see e.g. Tie and Brasseur, 1995, Solomon, 1999, Aquila et al., 2013 for a description of the mechanism behind) and can clearly be identified in the time series.*” We also made some small changes when referring to Weber et al., 2018 in the following lines, so that it should be clear that those authors make the same attribution of those dips to volcanic eruptions in their analysis, based on the same arguments.

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We added a description of the attribution of other large dips and peaks at the end of the time series, based on the relevant literature: “*In 2010, the Uccle ozone levels were unusually high, as over the entire NH extratropics. An unusually pronounced and persistent negative phase of the Arctic Oscillation and North Atlantic Oscillation in 2010, with the co-incidence of northern winter 2009/2010 with the easterly wind-shear phase of the QBO have been identified as major contributors (Steinbrecht et al., 2011) of this excess ozone. The 2011 ozone low anomaly cannot be fully explained by including this Arctic Oscillation and other dynamical proxies (e.g. for the Brewer-Dobson circulation) in the used multiple linear regression model in Weber et al. (2018), but might be linked to the strong Arctic ozone loss in 2011 (Manney et al., 2011). The below-average annual mean Uccle and NH total ozone in 2016 is partly ascribed to the severe Arctic ozone depletion in the same year and related to the anomalous quasi biennial oscillation (QBO) induced meridional circulation changes (see references in Weber et al., 2018).*”

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18. Lines 233-241 This section is too superficial. Your trend line has returned to the 1980 level by 2018 which is much faster than expected from ODS emissions. You seem to suggest the increase is therefore due to a large increase in the Brewer-Dobson circulation but then note the seasonality doesn't match. It is hard for the reader to have any confidence that the trend lines are meaningful.

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Thank you for this feedback. We understand your concern. Therefore, we also applied the LOTUS MLR model to the total ozone time series at Uccle and included the fit of the LOTUS MLR regression model and its different contributing terms (or proxies) to the total ozone monthly anomalies in the appendix A (Fig. A2). However, the trend estimates from this LOTUS model also give a nearly full total ozone recovery at Uccle, which can also be seen from the monthly anomaly time series itself in Fig. A2. As the spectrophotometers at Uccle are regularly calibrated on-site, we have no doubts on the homogeneity of the time series. Additionally, those trends are not completely incompatible with the vertical ozone trends estimated from the ozonesondes (which are of course normalized, pressure and temperature dependent to the total ozone measurements). We added an extra discussion in the manuscript.

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19. Line 259 What is the justification for treating AOD as constant since 2012? Have there been no volcanic injections into the stratosphere since then that could have affected Uccle?

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This is the approach followed in the LOTUS regression model for the AOD, so we consistently followed it. This approach is of course only valid assuming that during those last extrapolated years, the mean AOD was representative of background values. We think that for a NH site like Uccle, this is a valid assumption, whereas for SH sites, e.g. the impact of the 2015 Calbuco

eruption in Chile should not be underestimated (as it had an impact on the Antarctic ozone hole recovery e.g. Solomon et al., 2016, 10.1126/science.aae0061, and Ivy et al., 2017 <https://doi.org/10.1002/2016GL071925>).

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20. Line 267 I think this is really interesting work and I would have loved for you to show your full regression model and how well the different proxies can account for ozone variability at different altitudes at a specific site like Uccle.

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Thank you for your appreciation of this discussion. The output of the LOTUS MLR regression model and the different contributing terms (or proxies) for the monthly anomaly ozone concentrations at the layer 10 km above the tropopause (close to the ozone peak) are shown in Fig. S5. However, we think that showing the results of another multiple linear regression model for Uccle, with other proxies, does not bring a lot of added value here. First, we think that the use of a community developed and accepted MLR model should be preferred above an in-house developed MLR model. Secondly, the LOTUS report itself contains a nice overview of the sensitivity of the MLR models to the proxies for determining long-term trends in stratospheric ozone (see Section 4.3 “Sensitivity tests”). Thirdly, if we apply a simple linear regression model to calculate trends for Uccle, the estimated values are not significantly different from those of the LOTUS MLR model, as can be seen in Fig. 2 here below. However we added “However, here, the analysis is limited to the LOTUS model *and the sensitivity of the estimated trends on the chosen (M)LR model is very low for the Uccle time series.*”

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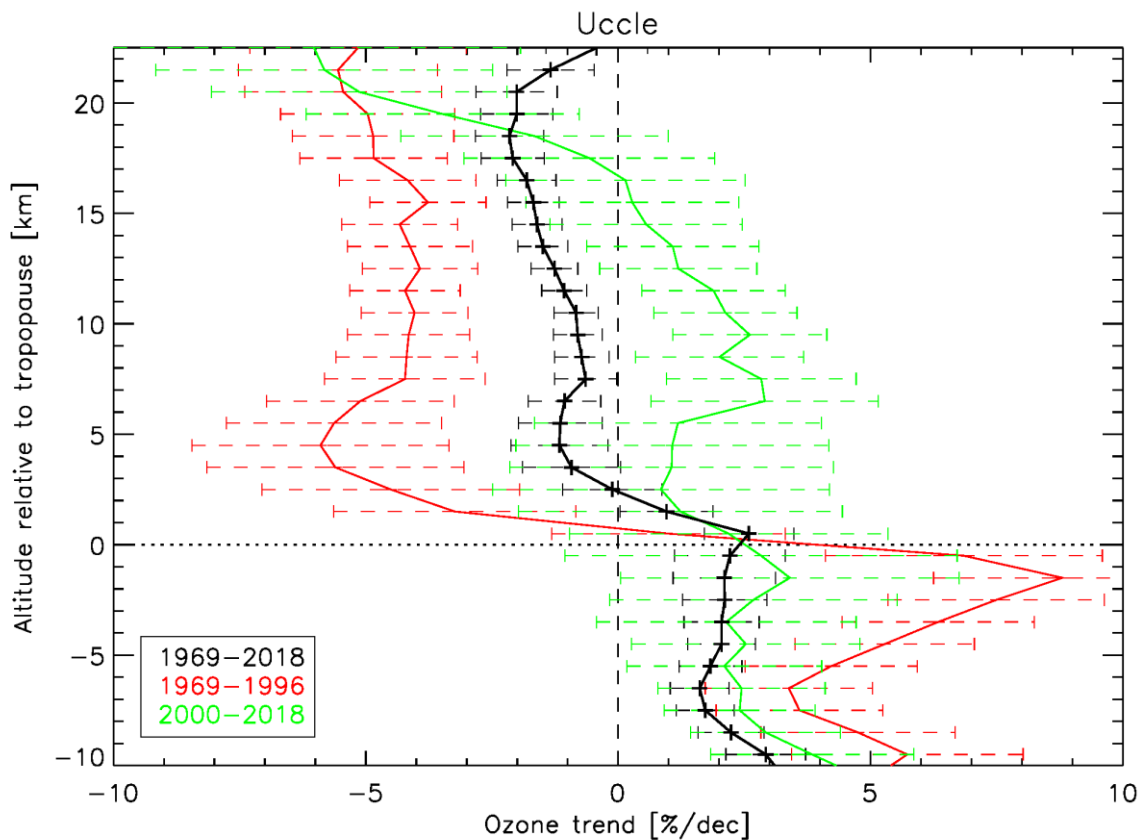


Figure 2: Vertical distribution of trends of ozone concentrations at Uccle for three different periods. The trends and their 2-sigma error uncertainties are calculated a simple linear regression model.

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21. Lines 274 I don't like the "etc" – you need to list the specific factors which have been identified.
 OK, we dropped the etc. All relevant factors are already summed up.

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22. Lines 276-277 Yes, this brings me to a question. Does it make sense to consider the relative height with respect to the tropopause once heights above 20 km say? Surely from 10 km above the tropopause, the absolute height is more relevant? This point might have been worth some consideration.

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In our opinion, it makes sense to still consider the relative height w.r.t. the tropopause at the higher pressure levels of an ozone sounding measurement, as we then also cancel out the seasonal variability of the ozone peak to some extent. In Fig. 3 here below, it can be seen that the ozone peak at Uccle lies at higher altitudes in summer (when the tropopause is also higher) than in wintertime (with a lower tropopause). For calculating long-term ozone trends at different pressure levels, we want to remove the seasonal cycle as much as possible, which leads to trend estimates which are more profile independent (i.e. more flat in the vertical component). To illustrate this, you can compare the vertical ozone trends from Fig. 2 and Fig. 4 in this response here. You can observe that the shape of the ozone profile is reflected in the vertical ozone

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trends with absolute altitude as vertical coordinate, while this is less the case for the trends as a function of the altitude relative to the tropopause.

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We added the following sentence to the text: “However, also for these altitudes, we prefer to calculate the vertical ozone trends in altitudes relative to the tropopause, to cancel out the seasonal variation of the ozone peak altitude, which roughly follows the tropopause height variation at Uccle: the ozone maximum peak is at its highest altitudes in summer (when the tropopause is also located higher), and lies at lower altitudes in winter (with the lowest tropopause). This approach gives in general vertical ozone trends that vary less over the different altitude levels.”

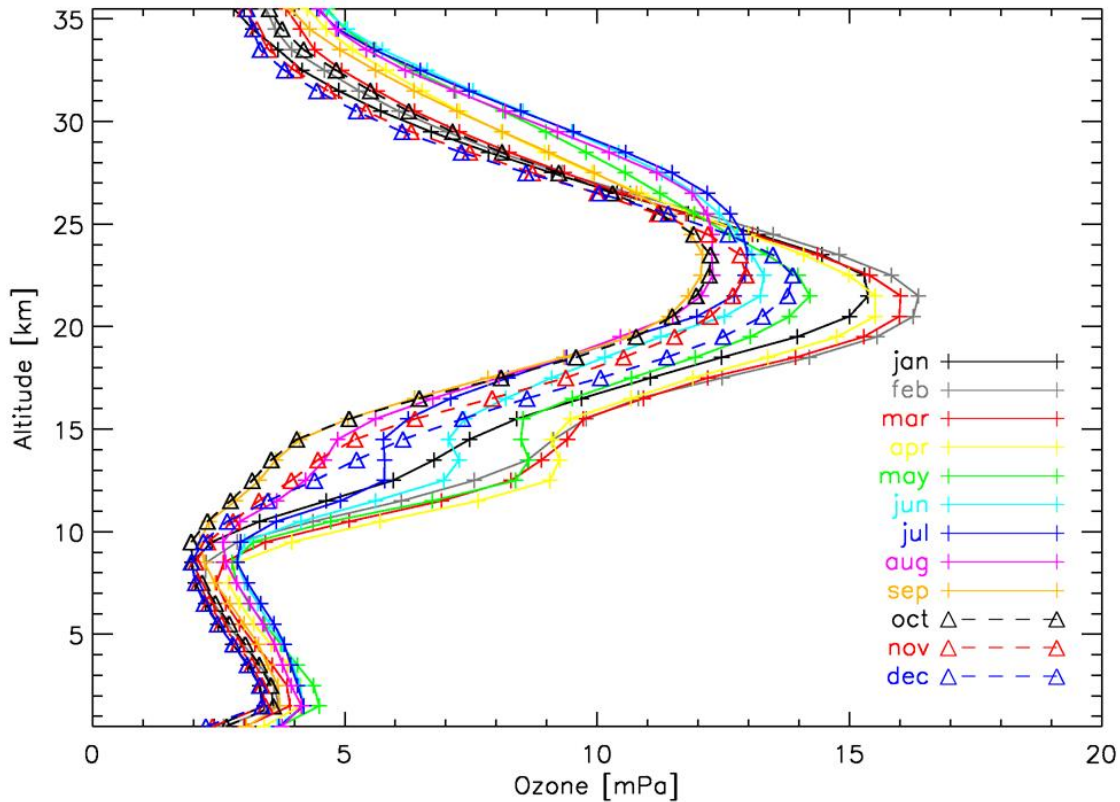


Figure 3: Monthly mean ozone profiles at Uccle.

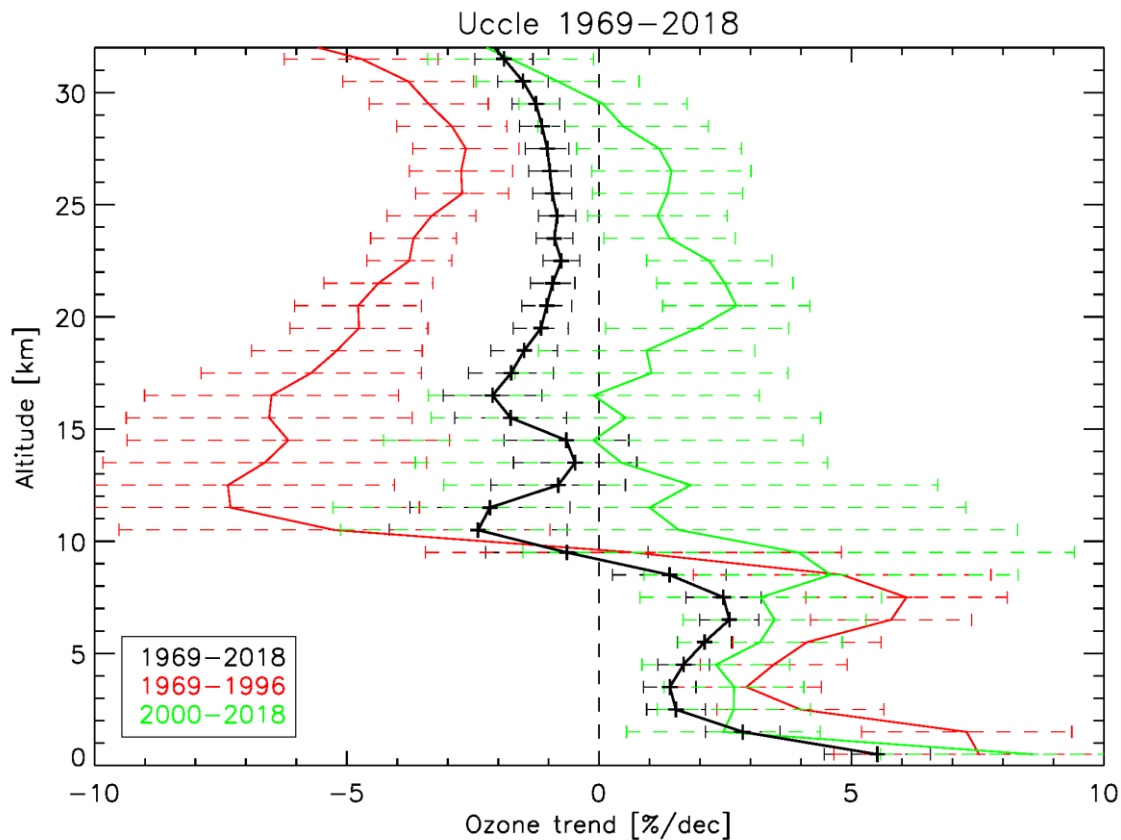


Figure 4: Vertical distribution of trends of ozone concentrations at Uccle for three different periods, but now with absolute altitude as vertical coordinate. The trends and their 2-sigma error uncertainties are calculated with a simple linear regression model.

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23. Lines 281-295 This is a very important result and deserves more attention.

After the submission of this manuscript, a couple of studies describing and explaining the lower stratospheric ozone decline in global observations have appeared. We updated and re-organized the discussion to those latest findings. However, it should also be noted that the lower-stratospheric ozone trends calculated from two nearby ozonesonde sites should also be put in perspective to the opposing trends from global satellite ozone retrievals and chemical climate model ensembles. Only a global assessment of the lower stratospheric ozone trends from different ground-based ozone instruments, could intervene with more weight in this discussion.

445

24. Line 297 (Figure 4). This is a very important part of the manuscript. The error bars for the recent period are very hard for the reader to assimilate. There does seem to be an offset between De Bilt and Uccle, not just larger uncertainty.

We increased the resolution of the figure. We have been considering other representations of the error bars (by symbols like triangles, circles) and other line styles for the lines connecting

450

455 the error bars, but the figure gets very busy with this non-standard representation. We also
want to keep this plot uniform with the vertical tropospheric ozone trends plot (now Fig. 3).
Yes, you are absolutely right that there is an offset. But taking into account the uncertainties of
both, you cannot state that this offset is statistically significant. Possible explanations between
460 the vertical ozone distribution and trends between Uccle and De Bilt have been given in great
detail in Van Malderen et al. (2016), we include a reference to these findings here. We added
the following text: *“The statistically insignificant offset between the Uccle and De Bilt trend
estimates is dependent on the used correction methods at both sites, but also differences in the
vertical ozone distribution (up to 5% in the stratosphere), of both geophysical and instrumental
465 origin, have an impact on the trend values (see e.g. Figs. 10a and 12 in Van Malderen et al.
(2016), in which a more detailed explanation of the differences in vertical ozone distribution and
trends between Uccle and De Bilt is given).”*

25. Lines 304-305 What level of "the stratosphere"?

470 As the text has been changed significantly, following your next comment, this sentence has
been replaced.

26. Lines 302-325 Unfortunately I find this discussion very "hand-wavy". There is a general
discussion of results from the literature but I find it hard to discern what the specific conclusions
are.

475 Independently from our study, Ball et al. (2020), based on CCMs, also looked at the imprints of
decrease ozone in the temperature variability, based on the same arguments. We included their
findings in this discussion and confronted them with ours. We also want to draw your attention
to the fact that the already cited Philipona et al. (2018) paper used the Uccle ozonesonde data,
480 averaged together with the Payerne and Hohenpeissenberg soundings, to also study the link
between stratospheric temperature and ozone trends. So, to our opinion, the literature
discussion is not so general and really tied to our dataset as well. The same argument is true
for the tropopause height variability: we first give the tropopause height variability at Uccle and
De Bilt, and compare these trend estimates from a recent, global literature study. Then, we
485 gave a general explanation of this global tropopause height variability, in which the Uccle and
De Bilt cases fit. However, we rewrote this discussion in lines 302-325 substantially.

27. Lines 345-350 Frankfurt and Munich seem a long way from Uccle if considering boundary layer
ozone – would you expect the trend to be correlated over this distance?

490 Not for boundary layer ozone, but the focus is here merely on free-tropospheric ozone, for
which a typical horizontal ozone correlation length is about 500 km (Liu et al., 2013). We added
this reference to the text. To be more specific about boundary layer ozone, we added to the text
“Near the surface, the De Bilt trend is in better agreement with the Frankfurt trend, but the local
surface ozone production and destruction and the boundary layer dynamics can vary
substantially between the three sites considered here, so that the boundary ozone distribution
495 and trends at the three sites are likely to be uncorrelated. However, comparing the lower-
tropospheric IAGOS measurements at Frankfurt with nearby (within 50-80 km) and more distant
(within 500 km) surface stations, Petetin et al. (2018) showed that the IAGOS observations in
the first few hundred meters above the surface at Frankfurt airport have a representativeness
typical of suburban background stations (like e.g. Uccle and De Bilt are), and as one moves
500 higher in altitude, the IAGOS observations shift towards a regional representativeness.”

28. Line 342 (Figure 5) This is a very important figure but the way you've drawn it is unintentionally misleading. The reader's eye sees that the black curve and the red curve agree very well, and agree better than the blue, and it's easy to miss the fact the black curve is for a completely different time period. I would suggest only showing the three 1995-2018 trends on the one plot. If you want to include the 1969-1994 Uccle trend it could be a different panel.

There is a clear legend in the figure, we think. Furthermore, we tried to use a consistent colour coding for different figures in the manuscript (Figs. 2, 3, S6): black for the entire Uccle period, green for the most recent Uccle sub-period (from 2000 for stratospheric ozone, from 1995 for tropospheric ozone), and blue for the De Bilt data. We added this information in the figure caption and explicitly referred to the colour of the vertical ozone trends in the text. We think it makes sense to include those vertical trends in one panel, to make it visually easier to directly compare their magnitudes. We do not want to include the 1969-1994 Uccle trend here (the 1969-1996 tropospheric ozone trends are already present in Fig. S3).

29. Line 342 (Figure 5) Wouldn't a lot of the height range shown in this plot be above the troposphere at Uccle, particularly in winter?

The tropopause heights at Uccle and De Bilt vary, in the mean, between 10.5 km in winter months and 11.5 km in summer months (with standard deviations between 1 and 1.5 km). So, some of the height range shown in this plot is showing variability of stratospheric ozone as well. We included this information in the text when discussing the upper-tropospheric trends: *"Also, at those altitudes, the trends do not represent the tropospheric ozone time variability only, as the mean tropopause height range between 10.5 km (winter time) and 11.5 km (summer time), with standard deviations between 1 and 1.5 km, both at Uccle and De Bilt. As a consequence, lower-stratospheric ozone concentrations will contribute to the estimated trends in the upper altitude levels in Fig. 3."*

30. Lines 399-403 This result seems extraordinary. Could the number of events really have increased by a factor of eight in only fifty years? It seems very implausible, and if you want to include it you need to show much more convincing evidence. (Otherwise the reader will assume the most likely explanation is that there's a mistake in your algorithm).

The result on the tropopause fold frequency increase is based on an algorithm, developed for ozonesonde profiles by Van Haver et al., 1996, that identifies tropopause folds based on 6 criteria (2 for ozone, and then 1 for relative humidity, stability, vertical gradient of wind speed and wind speed), see also Fig. S8 in the supplementary material for an visual illustration of those criteria in one profile. Only if those 6 criteria are fulfilled, a tropopause fold is identified. Van Haver et al. used this algorithm in automatic mode. The results shown in the manuscript are done in automatic mode as well. Here, two important remarks should be made:

1) As noted in the text, the higher vertical resolution of the sounding data in the more recent digital era (since 1990) might have an impact on the larger detected number of tropopause folds, although the amount of events has continuously increased since then. For instance, the increase in the digital era is $0.12 \pm 0.05\%$ per year. For the first 25 years of the time series (until 1995), Van Haver et al. (1996) mentioned an increase of $0.07 \pm 0.06\%$, while we obtain an increase of $0.10 \pm 0.06\%$ since 1995. So, the increase of the frequency seems to be independent of the vertical resolution of the sounding data, but the rate of increase might be affected by it.

2) We also used the algorithm in manual mode. This means that the vertical profiles of all relevant variables are plotted if at least 5 tropopause fold identification criteria are met. In this mode, one can ascertain visually if a tropopause fold is really present in the profile or not, and why criteria are met or not. In this manual/visual inspection mode, which is of course more subjective, more tropopause folds have been identified (344 instead of 290), and in particular in the beginning of the time series. The main reason is that the automatic method do not identify a tropopause fold when there is no relative humidity data, or when the humidity sensor was clearly iced at the tropopause fold location (following the icing recognition algorithm developed by Leiterer et al., 2005, doi:10.1175/JTECH-1684.1). Unfortunately, missing or iced humidity sensor data occur mostly for the older radiosonde humidity sensors (VIZ, Vaisala RS80). More recent radiosonde types in use in Uccle (since 2007: RS92 and RS41) do not suffer from iced sensors because they are equipped with 2 sensors, that are heated alternately. As a result, the time series of manually identified tropopause folds shows a lower increase rate of the frequency, of about $0.09 \pm 0.02\%$ per year for the entire 1969-2018 time period. After 1990, the increase rate amounts to $0.05 \pm 0.04\%$ per year.

So, to conclude, to be as objective as possible in the tropopause fold identification, we prefer to use the automatic detection mode of the algorithm, but we point to a possible overestimation of the trend of the frequencies due to the reasons summed up here above. However, given the sensitivity analysis for those two impacting factors, we believe that the increase in the trend frequency is very robust and an important result (see references in the manuscript). Moreover, the very high vertical resolution of ozonesonde profiles (and of the coupled radiosonde profiles of meteorological variables), makes this one of the most suitable datasets for identifying tropopause folds. Finally, the (global) climatology of tropopause folds is recently gaining importance, as (chemical) reanalyses like CAMS and climate models are used to analyse this (e.g. recent work by Akritidis et al., 2019 and 2021, resp. doi: 10.5194/acp-19-14387-2019 and doi: 10.1029/2020JD034115). Currently, we are also working on a similar analysis of ERA5, but results are too preliminary to already share at this moment. Based on these arguments, we would argue to keep this paragraph on board of the paper, but we are open to omit it if you insist on it.

The manuscript has been changed as follows: “*On one hand, the large increase over the entire time period might be explained to some technical aspects. First, the higher vertical resolution of the sounding data in the more recent digital era (since 1990) might have an impact on the larger detected number of tropopause folds (thinner layers might be detected), although the amount of events has continuously increased since then, at a slightly smaller rate of $0.12 \pm 0.05\%$ per year. Secondly, a visual inspection of all profiles fulfilling at least five of the tropopause fold detection criteria, led to a higher number of (manually) identified events (around 50), and (relatively) especially in the beginning of the time series. This is explained by the fact that the low humidity criterion was often not met in the automatic detection, because there were no humidity data or the humidity sensor was iced (following the icing recognition algorithm of Leiterer et al., 2005). More recent types of radiosonde humidity sensors (in use since 2007 at Uccle) prevent ice contamination by heating them during flight. However, this manual (and hence more subjective) mode of the algorithm still gives a $0.09 \pm 0.02\%$ increase per year of the tropopause fold events since 1969. Therefore, we believe that the significant increase, although possibly overestimated by the automatic procedure, is nevertheless a robust feature of the analysis here. On the other hand, a higher rate of tropopause folding events is expected*

595 *due to climate change (Tarasick et al., 2019, and references therein): climate change is projected to increase planetary wave activity and so cause an accelerated Brewer-Dobson circulation. This acceleration, along with stratospheric ozone recovery, will lead to increased transport of ozone from the stratosphere into the troposphere and hence more tropopause folding events. Akritidis et al. (2019) add to this that the degree of increase in the downward transport of stratospheric ozone is partially driven by the long-term changes in tropopause fold activity.”*

- 600 31. Lines 412-465 Unfortunately, I have to say my honest recommendation would be to delete section 4.4 altogether. For one thing, it is not clear that the ozonesonde record adds any benefit compared to the continuous surface monitoring. (Noting that the ozonesonde data before 1985 is of limited value). There is a small offset which you don't seem to explain. Secondly, although
605 you show an increasing trend, after a fairly lengthy discussion you are not able to reach any conclusions about its causes (in terms of changes in the precursors or meteorological conditions or some other factor).

610 *We moved section 4.4 to an appendix. We really think that this section should not be deleted altogether. Concerning your first point: we do not claim that the ozonesonde record adds any benefit compared to the continuous surface monitoring. As a matter of fact, it is the other way around: the agreement in trend with the surface monitoring device gives some credibility to the ozone sounding measurements near the ground, despite the impact of pre-launch procedures on the surface ozone measurements, which might be a reason for the small offset as well (explained in the text now). These two points have been raised respectively in the text as “In this appendix, we elaborate more on how representative and complementary the surface ozone trend derived from the ozonesonde data at Uccle is, compared to the one from a surface station at the same site. The ground network of (air quality) stations provides surface ozone measurements at higher temporal and horizontal resolution, and with higher accuracy than ozonesonde measurements, but these latter provide vertical ozone profiles in the lower troposphere as well, and sometimes even over a longer time span.” & “This offset might be explained by the difference in air masses for which the ozone concentrations are measured (surface vs. surface to 1 km above the ground), and by some Uccle pre-launch procedure of testing the ozonesonde-interface-radiosonde configuration by exposing the ozonesonde shortly (< 30 s) to (stratospheric) ozone concentrations between 15 to 30 minute prior to launch. Because of the slow time constant of 20-25 minutes in the chemical reactions in the cell, this pre-launch ozone exposure might still contribute to the measured cell current after launch, resulting in a positive bias in the boundary layer ozone measurements with the ozonesondes.”*

630 Concerning your second point, we agree that the discussion about the trends was rather lengthy and perhaps a bit too general. So we shortened and reorganized the discussion by concentrating on the ozone (mean) trends (only since 1986 for ozonesonde data as well) first, describe the general possible causes for them, and then concentrate on the link with the ozone precursor emission changes, because these measurements are available at the Uccle site (or nearby for CO). Finally, we describe the Uccle surface ozone trends for the lowest and highest
635 percentile data (Figure S10 in Supplementary Material), as these give additional information for explaining the increase in (mean) surface ozone amounts. Finally, we also want to draw your attention to the fact that a clear identification of which factors contribute most to the observed surface ozone trends is not straightforward at any site and has consequently hardly been for

640 any site in the literature so far. We think that the summarizing conclusion contains it all: *“To conclude, explaining the increasing mean surface ozone amounts in combination with the decreasing ozone precursor emissions at Uccle is less straightforward than the (opposing) trends in high and low level ozone concentrations due to the compression of the surface ozone distribution. The interpretation of the increasing mean surface ozone concentrations is hampered by the interplay of many factors such as meteorology and transport, the non-linear dependence of the ozone concentrations on the emissions of VOC and NO_x, the dual role of NO_x as ozone source or sink depending on the season, and the amount of NO_x emissions.”*

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32. Line 464 (Figure 7) Overall the figure is cluttered and difficult to read. Just from looking at the plot, while there appears to be a good agreement between the ozonesonde and the surface monitor, it is really just the existence of a consistent seasonal cycle. There seems to be a jump around the year 1998?
- 650

We decided to replace this Figure 7 by two figures. One figure shows only the surface ozone monthly means from ozonesondes and the surface monitor, so that the offset and the consistent seasonal cycle is highlighted. In the other figure (inspired by a similar figure that was originally in the supplementary material), we show the monthly anomalies of the surface ozone by the two devices (we do not identify a jump around 1998 in this figure and it should be clear that both time series agree also rather well after removal of the seasonal cycle), with in the lower panels the monthly anomalies from the co-located NO, NO₂ and CO concentrations. All monthly anomaly time series do not start earlier than 1986 and the figure has been enlarged as well, so we hope the figure is less cluttered and easier to read now.

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33. Line 484 Who is "us" in this context?

We removed “us” here.

34. Line 503 You should explain why applying the averaging kernels makes a much larger improvement in the lower stratosphere compared to other altitudes.
- 665

Yes, you are absolutely right. This is because the lower stratosphere is the region with the highest ozone variability, so that, smoothing the high vertical resolution ozonesonde data to correctly inter-compare both products at the same vertical resolution, will have the largest effect here by removing the details of the differences.

670

In the text, we added: “The lower stratosphere is the region with the highest ozone variability, so smoothing the high resolution ozonesonde profiles to the GOME-2 vertical resolution will have the largest effect here by removing details of the differences.”

35. Lines 505-509 This would be more convincing if you didn't just show the "before" and "after" plots – the reader is left wondering whether the degradation correction has been tuned to match the ozonesondes – perhaps you could show the degradation correction over time?
- 675

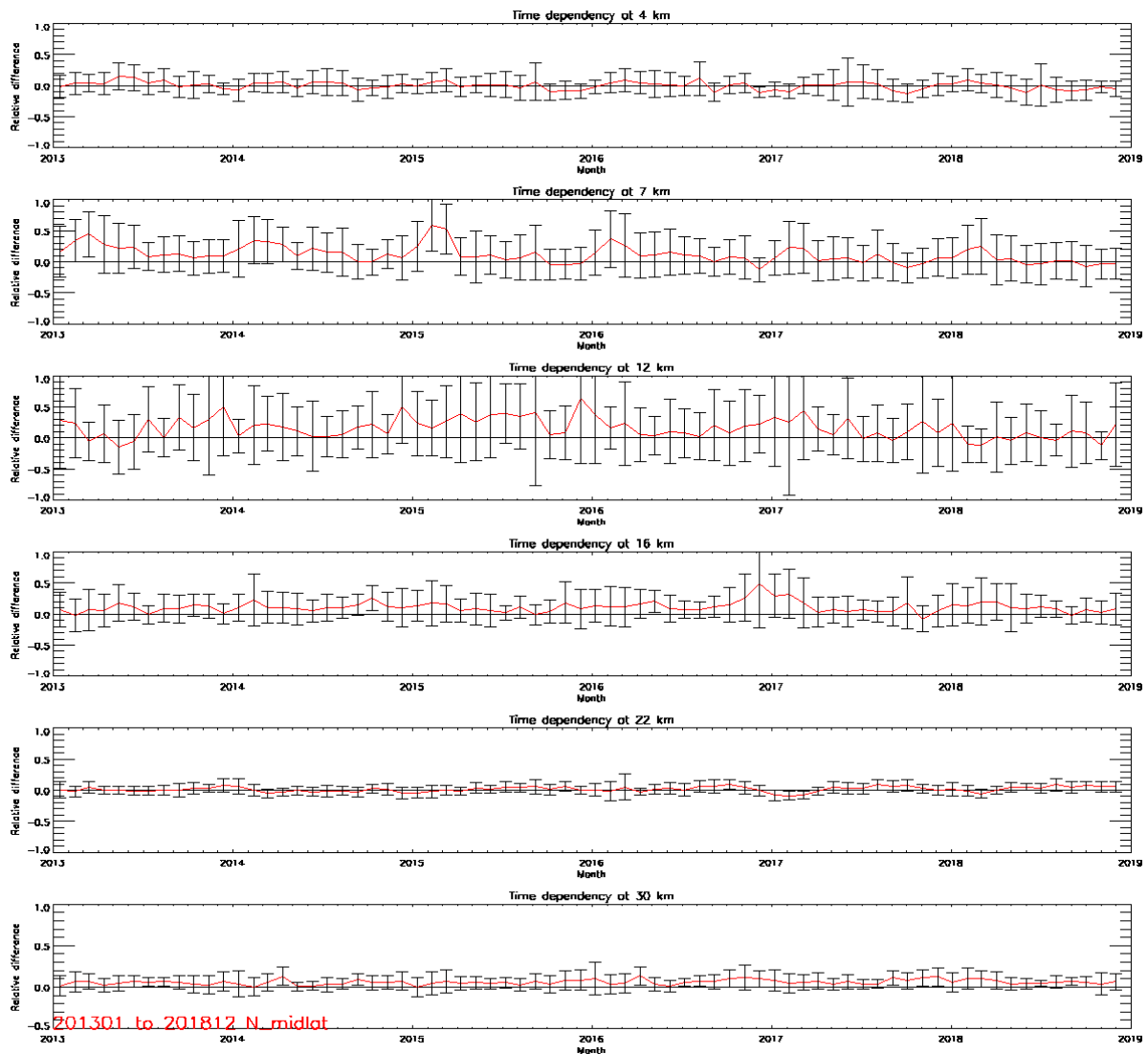
Thank you for your comment. The degradation correction has certainly not been tuned to match the ozonesondes. We gave in the manuscript some more information about the degradation and its correction: “For example, the measured values of the GOME-2A Irradiance in the UV (below 300nm) has reduced by roughly 80% in 2016 (since its launch in 2007). Since the vertical ozone profile retrieval algorithm depends on an absolute calibrated reflectance (sun normalised radiance) there is a need to correct for this temporal change of the (joint) radiance and irradiance. This method depends on the assumption that, taken as an average across the

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685 *globe, the atmospheric constituents (mainly ozone) will be close to the multiyear climatological*
value from McPeters and Labow (2012). The climatological ozone profile is then scaled with the
Assimilated Total Ozone columns to get the overall ozone absorption correct (Tuinder et al.,
2019)."

690 Figure 6.3 in the document shows the effects of the different methods that can be applied to re-
distribute an excess or shortcoming of the expected ozone column.

695 The relative differences between the GOME-2A retrievals and NH midlatitude ozonesonde
ozone measurements after applying the degradation error correction (Fig. 5) or not (Fig. 6) are
presented in the plots here below. It can be seen that the agreement between both datasets is
better, especially at but not restricted to the end of the time series when the degradation error
correction is applied. However, we believe that this is out of scope for the present manuscript
that focuses on the Uccle dataset, as the validation of this degradation error correction requires
a more global approach (as shown in the plots here below).



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Figure 5: Time series of relative ozone differences between GOME-2A and NH midlatitude ozonesonde at 6 different altitude levels for January 2013 - December 2018 for the GOME-2A time series that have been corrected for the degradation error.

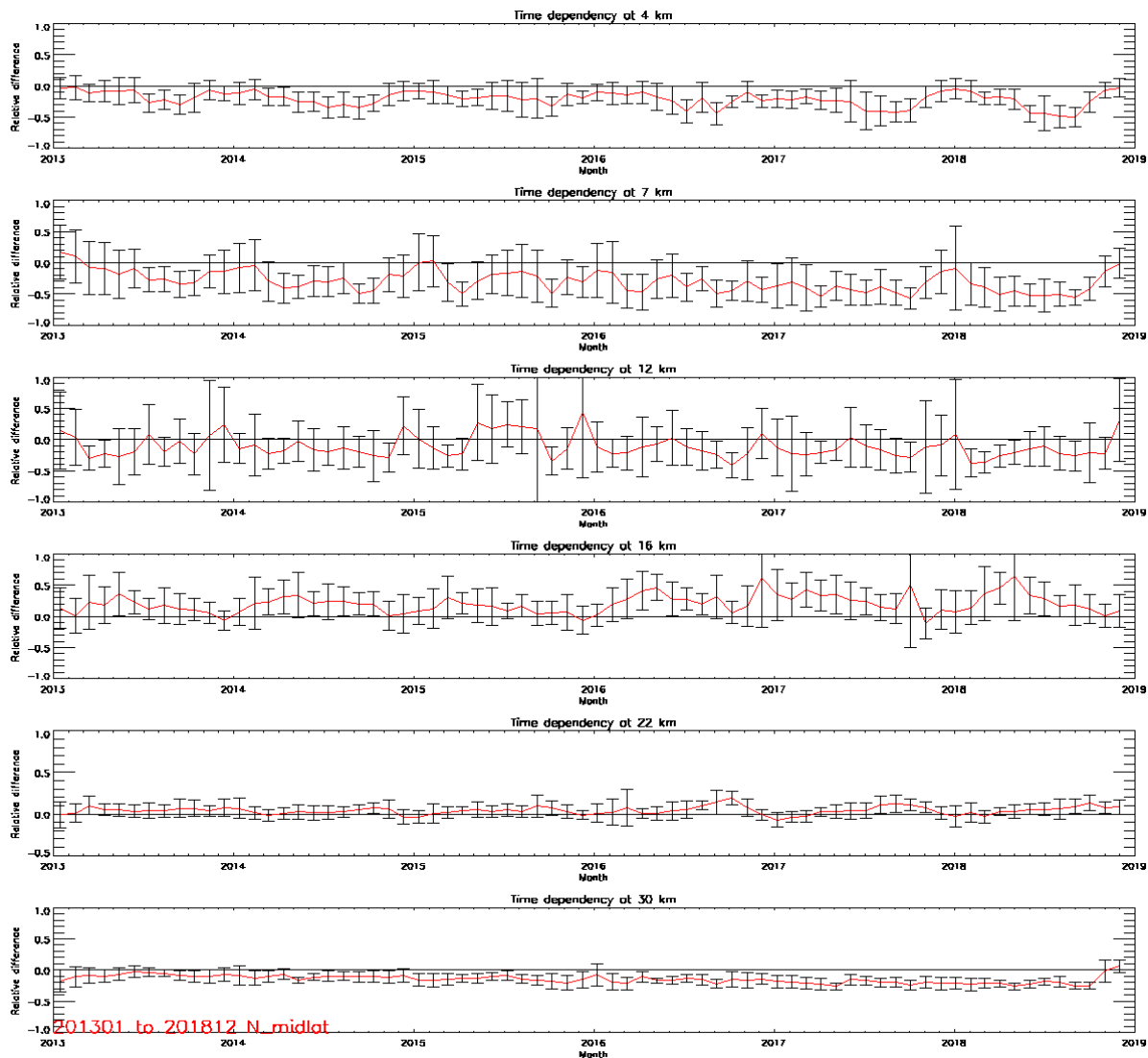


Figure 6: Time series of relative ozone differences between GOME-2A and NH midlatitude ozonesonde at 6 different altitude levels for January 2013 - December 2018 for the GOME-2A time series that have been not been corrected for the degradation error (operational processing).

36. Line 531 In section 5.2 you don't use the averaging kernels but in section 5.1 you said how important it was to apply them – why is this?

It really depends on the satellite ozone retrieval technique with which the ozonesondes are compared. In the case of MLS, “the relatively high vertical resolution due to the limb sounding technique, allows for many scientifically useful studies to be undertaken without reference to the averaging kernels” (see pag 7, Livesey et al., 2020). Moreover, from Fig. 3.18.8 (top) on page 128 in the same document, the dashed line (the FWHM of quasi-triangular functions) shows that the vertical resolution of MLS is rather stable around 2.5-3 km between 10-200 hPa

(vertical range of interest for comparison with Uccle ozonesondes). So, we simulated those averaging kernels by determining around each MLS pressure level a pressure range that coincides with half of this vertical resolution. For this pressure range, we then interpolated the ozone concentrations in the ozonesonde profile to the MLS pressure level, and this value was then compared with the MLS measurement at that pressure level. This interpolation is to be preferred above layer averaging (see again Livesey et al., 2020), but the differences between both approaches for Uccle are at most 8% at 200 hPa, and less than 3% between 100-10 hPa. Another drawback of using the averaging kernels for MLS is that MLS provides only one fixed averaging kernel for our latitude for all data, which is e.g. not the case for GOME-2 and TES.

We modified the text into: *“Thanks to the relatively dense and regular MLS vertical resolution of around 2.5 km in the 10-200 hPa pressure range, it is feasible to interpolate the Uccle ozonesonde data to the MLS pressure levels on a fine pressure grid of 2.5 km. Applying the time invariant MLS averaging kernel on the latitude of Uccle on the ozonesonde data did not have a large effect on the smoothing of the vertical ozonesonde profile, as compared to applying the identity matrix to the ozonesonde vertical profile (< 1%). This contrasts strongly with the GOME-2 and TES retrievals (see Sect. 5.3), where the spatio-temporal varying averaging kernels affect the vertical ozone profiles substantially, and as such should be used on the sonde data for pairwise comparison.”*

37. L 537-538 The figure shows a growing bias at altitudes above 10 hPa – it seems to be a systematic effect and not just that the ozonesondes are "known to be less accurate"

Yes, it is a systemic effect, in the ozonesonde measurements. You are completely right. Above 10 hPa, the sensing solutions either boil/evaporate and freeze, changing completely the stoichiometry of the chemical reactions, but mostly leading to an underestimation of the true ozone concentrations. However, at those pressure levels, the pump efficiency decrease of the ozonesonde is really large, with large uncertainties, which makes the ozone measurement not very accurate there as well. Both effects increase with increasing altitudes. Above 10 hPa, it is therefore no longer advised to use ozonesonde data, as simulation chamber experiments with a photometer as reference have shown (Smit et al., private communication). The same effect is seen in the composite ECC-MLS comparisons in Fig. 3 of Stauffer et al. (2020), for both manufacturers of ECC ozonesondes.

The text has been changed into *“At pressures smaller than 10 hPa, ozonesonde measurements are systematically underestimating ozone due to the evaporation or freezing of the sensing solutions (see also the composite ECC-MLS Fig. 3 in Stauffer et al., 2020), and they have a larger uncertainty due to increased pump efficiency uncertainty at low pressures.”*

38. Lines 545 (Figure 9) Overall I like this figure but the error bars seem strange. If the agreement from one year to the next is so good, it doesn't seem possible that the error bars could really be so large. The description in the caption is too brief and doesn't show what the error bars are really representing.

The caption has been changed into *“Relative ozone profile differences between MLS and Uccle ozonesondes. The different colours correspond to the different yearly averages, illustrating the large consistency among those. The black line represents the overall mean relative differences, with the error bars the one standard deviations due to the individual differences. Therefore, it could be noted that individual differences are relatively large at some pressure levels, but they are cancelled out in the yearly mean.”*

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39. Line 551 Here, you should also let the reader know that TES was decommissioned in early 2018.

Thank you! We added this information.

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40. Line 552 You have already described the orbit of Aura in the MLS section.

Thank you, we removed the Aura orbit description here.

41. Line 559 It seems curious that you apply a limit of 300 km in the troposphere but 100km in the stratosphere – is this a reasonable thing to do?

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It is common practice to use 300 km between the sonde launch location and the overpass of the satellite to compare ozone profiles in the troposphere for TES (see Nassar et al., 2008; Verstraeten et al., 2013, among others) to ensure a reasonable amount of pairs of ozone profiles. When we use the 100 km criteria we only have 33 pairs, otherwise 191, which is a substantial difference. For tropospheric profiles, the quality flag is determined by clouds (especially for Uccle which has many cloudy and rainy days) and retrieval errors, among others, reducing the amount of data passing the quality flag.

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42. Line 563 You should explain to the reader what is meant by the term "observation operator". Is it the same as an averaging kernel?

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Yes, indeed, the observation operator is the same as the averaging kernel. In order not to mix terminology we have changed it in the revised manuscript to averaging kernel.

43. Line 580 In this case, seeing there are only "1-2" degrees of freedom in the tropo-sphere, wouldn't it better to make a plot of the regression described in lines 581-590, rather than figure 10 as it currently stands? Then the reader could see the temporal stability.

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We agree. We have added the correlation plots for the lower and upper troposphere in the revised manuscript, as lower panels in what becomes Figure 7 now.

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Response to Reviewer 3 of Van Malderen et al. – 50 years of balloon-borne ozone profile measurements at Uccle, Belgium: short history, scientific relevance and achievements in understanding the vertical ozone distribution.

805

In this response, we included the reviewer comments in black. Specific comments are numbered. Our response are written in red, with the modifications in the manuscript in *red italic*.

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This manuscript provides a detailed overview of the ozonesonde measurements Uccle. This is one of the most important and longest records of profiles measured at a higher frequency (3/week) than nearly all other global ozonesondes sites. The manuscript covers the ozonesonde history, editing techniques applied to past data (homogenization) and includes data analysis of long-term tropospheric and stratospheric ozone. The fully homogenized data is used in the linear regression and well-documented LOTUS models to evaluate trends. Trends are evaluated and compared to MOZAIC/IAGOS commercial aircraft profiles. An additional interesting topic is the evaluation showing an increase in the frequency of tropopause fold events observed in the ozonesonde record. The manuscript also discusses and documents the changes in manufacturer sonde models used over the long record and operating procedures (changes were minimal making homogenization of long-term data much more straightforward). It also presents an important documentation of the homogenization method used for their long term records. The manuscript presents satellite comparisons/validations with the Uccle ozonesonde database, one of the most critical applications of ozonesonde data. This manuscript is a substantial contribution that shows the importance of 50 years of ozonesonde data records.

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Thank you very much for taking your time to review our manuscript and your positive feedback!

General Comment

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1. Scientific Question: Figures S6 shows an example of a tropopause fold event - a narrow high ozone layer at 600 hPa. I am not all that familiar with tropopause folds but have seen some examples showing massive ozone in broad layers near the tropopause. The RH is very low in the green line (very hard to see the RH scale in light green) which would indicate stratospheric source but wondering if anything else that shows this is a purely stratospheric ozone peak?

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This is an example of a tropopause fold illustrating that stratospheric air can penetrate deeply in the troposphere, not only near the tropopause. The very low RH is one indicator of the stratospheric source, but the detection algorithm looks especially for tropopause folds which occur in connection to upper tropospheric frontogenesis in the polar jet stream region, as those are considered to be responsible for a large part of the mass exchange across the tropopause (see Van Haver et al., 1996, and references therein). During a test period, those authors used cross sections of potential vorticity from ECMWF analysis to check the necessary folding of the dynamical tropopause, resulting in the tuning of the stability, wind speed and vertical shear conditions as used in the detection method. We added to the figure caption that “These criteria primarily focus on the detection of tropopause folds that occur in connection to upper

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tropospheric frontogenesis in the polar jet stream region, as those are considered to be responsible for a large part of the mass exchange across the tropopause (see Van Haver et al., 1996, and references therein)."

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We must admit that at our printed out copies, we have no issue with the visibility of the light green color, which has also been used in other figures in the manuscript (e.g. Figs. 3, S1, S6). But, we will keep an eye on it.

Technical Corrections/Suggestions:

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2. Line 43: First sentence states ozone is found mainly from surface to top of atmosphere (50km) which is true for all gases. Would be good to separate it out a little more and note that Ozone, O₃, is a key trace gas in the Earth's atmosphere, where is present in the troposphere but mainly resides in the lower to middle stratosphere (90%).

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Done, we added: "with the highest concentrations in the lower to middle stratosphere (90% of total column ozone amount)."

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3. Line 109: I would say something like "funding limitations or reductions" here rather than "financial problems"

Done, changed to "funding reductions".

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4. Line 151: reduction by 100% may sound like the ozone signal is zero, which can be the case in very high SO₂ that exceeds ozone concentration. It would be more clear to state that – in particular, SO₂ reduces the ECC cell response on a 1:1 basis for every SO₂ molecule.

You are right, we changed it to "In particular, one SO₂ molecule cause a reverse current of two electrons, reducing the electrochemical cell response on a 1:1 basis"

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5. Line 170: Change "double soundings" to "dual soundings" so it matches with text in Figure 2.

Done as suggested.

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6. Line 208: Change "From 1990" to "Since 1990"

Done as suggested.

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7. Line 213: SECTION 4 Note: This section title is "Temporal evolution of vertical ozone concentrations at Uccle" but the section begins with "Total ozone trends from Dobson and Brewer". Therefore, this section may be better titled as "Temporal evolution of total column and vertical ozone concentrations at Uccle"

As a response to the other review report, we moved the "Total ozone trends" subsection to an appendix, so the title of Section 4 now better covers the entire contents of this section.

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8. Line 227: "...on the Uccle total ozone concentrations pops up." to "...shows in the significant dips in Uccle total ozone."

Done, changed to "is shown in the significant dips in Uccle total ozone."

9. Line 231: "(e.g. the excess total ozone in 2010, the 2011 and 2016 low ozone anomalies)." to "(e.g. the excess total ozone in 2010, and the low ozone anomalies in 2011 and 2016)."

Done as suggested.

895 10. Line 351: Improve sentence to make clear if proxies were not used: “.....,as there is no consensus on the used proxies to account for natural variability” which implies that proxies were used or change to “....., as there is no consensus in using proxies to account for natural variability.”

Done, changed to “as there is no consensus in using (which) proxies to account for natural variability.”

900 11. Line 368: Need more clarity here: “(since 1995, but the post-2000 trends have the same magnitude)” Is a flat trend (zero slope) meant for same magnitude. Also, wondering if post-2000 trends are shown in one of the graphs.

905 *You are right. We changed the text in “The Uccle tropospheric ozone concentrations have been increasing at about the same rate since 1969 (in black in Fig. 3) as since 1995 (in green in Fig. 3), and also the post-2000 increase rate is very similar (not shown here, but could to some extent be noted from the tropospheric ozone column time series shown in Fig. S7).”*

910 12. Line 621: Replace...” is almost entirely compensated by the gain” with...” has nearly fully recovered by the +2%/decade gain between 1997-2019”

Done as suggested.

915 13. Line 645: The most recent update by Stauffer shows the ozonesonde drop-off in TCO ranges from 3-7% was observed at 13 of 53 global stations (25%) 1/4 rather than 1/3.

Yes, thank you, we are aware of this update (being member of the ECC Drop-off Task Team within ASOPOS) and even a more recent one (after re-processing of two Canadian sites). We added to the text: “a number now reduced to about 20% (12 of 60 global stations, Stauffer et al., 2021, private communication)”.

920 14. Line 651: The drop-off was mentioned in line 645 – suggest removing “, as the total column ozone drop-off in a third of the ozonesonde stations (Stauffer et al., 2020) paper made obvious”

Done as suggested.

925

50 years of balloon-borne ozone profile measurements at Uccle, Belgium: short history, scientific relevance and achievements in understanding the vertical ozone distribution

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Abstract. Starting in 1969, and with three launches a week, the Uccle (Brussels, Belgium) ozonesonde dataset is one of
longest and densest of the world. Moreover, as the only major change was the switch from Brewer-Mast ([BM](#)) to
940 Electrochemical Concentration Cell (ECC) ozonesonde types in 1997 (when the emissions of ozone depleting substances
peaked), the Uccle time series is very homogenous. In this paper, we briefly describe which efforts have been taken during
the first three decades of the 50 years of ozonesonde observations to guarantee the homogeneity between ascent and descent
profiles, under changing environmental conditions (e.g. SO₂), and between the different ozonesonde types. This paper
focusses on the 50 years long Uccle ozonesonde dataset and aims to demonstrate its past, present and future relevance to
945 ozone research in two application areas: (i) the assessment of the temporal evolution of ozone from the surface to the
(middle) stratosphere, and (ii) as backbone for validation and stability analysis of both stratospheric as well as tropospheric
satellite ozone retrievals. Using the Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS) multiple linear
regression model (SPARC/IO3C/GAW, 2019), we found that the stratospheric ozone concentrations at Uccle declined at a
significant rate of around 2% dec^{-1} since 1969, rather consistently over the different stratospheric levels. This overall
950 decrease can mainly be assigned to the 1969-1996 period with a rather consistent decline rate around -4% dec^{-1} . Since
2000, a recovery between +1 to +3% dec^{-1} of the stratospheric ozone levels above Uccle is observed, although not
significant and not for the upper stratospheric levels measured by ozonesondes. Throughout the entire free troposphere, a
very consistent increase of the ozone concentrations at 2 to 3 % dec^{-1} is measured since both 1969 and 1995, the latter
trend being in almost perfect agreement with the trends derived from the In-service Aircraft for a Global Observing System
955 (IAGOS) ascent/descent profiles at Frankfurt. As the amount of tropopause folding events in the Uccle time series increased
significantly over time, increased stratosphere-to-troposphere transport of recovering stratospheric ozone might partly
explain these increasing tropospheric ozone concentrations, despite the levelling off in (tropospheric) ozone precursor
emissions and notwithstanding the continued increase of mean surface ozone concentrations. Furthermore, we illustrate the
crucial role of ozonesonde measurements for validation of satellite ozone profile retrievals. With the operational validation

960 of Global Ozone Monitoring Experiment GOME-2, we show how the Uccle dataset can be used to evaluate the performance of a degradation correction for the MetOp-A/GOME-2 UV sensors. In another example, we illustrate that the Microwave Limb Sounder (MLS) overpass ozone profiles in the stratosphere agree within $\pm 5\%$ with the Uccle ozone profiles between 10 and 70 hPa. Another instrument on the same AURA satellite platform, Tropospheric Emission Spectrometer (TES), is generally positively biased with respect to the Uccle ozonesondes in the troposphere by up to ~ 10 ppbv, corresponding to
965 relative differences up to $\sim 15\%$. Using the Uccle ozonesonde time series as reference, we also demonstrate that the temporal stability of those last two satellite retrievals is excellent.

1 Introduction

Ozone, O_3 , is a key trace gas in the Earth's atmosphere, where it mainly resides between the surface and the top of the stratosphere (about 50 km), with the highest concentrations in the lower to middle stratosphere (90% of total column ozone amount). Ozone is mainly produced in the tropical stratosphere and transported to the lower stratosphere at high latitudes. Depending on its altitude, ozone is involved in different chemical reactions and therefore has a different impact on life on Earth. Stratospheric ozone absorbs the harmful solar ultraviolet (UV) radiation, hereby protecting life on Earth, and warming the stratosphere. This protective shield has been in danger due to anthropogenic emissions of ozone depleting substances (ODSs, such as chlorofluorocarbons or CFCs) since the 1970s, with the Antarctic springtime ozone hole as the most striking
975 signature. Thanks to the Montreal Protocol (1987, and subsequent amendments and adjustments), positive trends in the ozone concentrations in the upper stratosphere are observed since 2000 (WMO, 2018, Chapters 3 and 4, and SPARC/IO3C/GAW, 2019). Ozone is also an important absorber of infrared (terrestrial) radiation, mainly in the tropopause region, and therefore acts-can act as a greenhouse gas at certain altitudes, estimated to have contributed $\sim 20\%$ as much positive radiative forcing as CO_2 since 1750 (IPCC, 2013)~~estimated to be responsible for 1/4 to 1/3 of the Earth's warming over the past 200 years (Thompson et al., 2019)~~. Tropospheric ozone is also the main source of the OH free radical, the primary oxidant in the atmosphere, responsible for removing many compounds (including atmospheric pollutants) from tropospheric air. At the surface, ozone is an air pollutant that adversely affects human health, natural vegetation, and crop yield and quality (e.g. Cooper et al., 2014).

Because of the many roles of ozone, the knowledge and measurement of the vertical distribution of the ozone concentration
985 in the atmosphere – and its variability in time – is crucial. Vertical ozone profiles can be obtained from ground-based instruments (Dobson/Brewer Umkehr, lidar, FTIR, and microwave radiometer), balloon-borne techniques (ozonesondes), and satellite-based measurements (using solar/stellar occultation, limb emission/scattering and nadir-viewing techniques), (see e.g. Hassler et al. (~~2014~~) for details). In this research, we focus on ozonesondes, lightweight and compact balloon-borne instruments measuring the ozone concentration from the surface through the mid-stratosphere (about 10 hPa or 30
990 km). In electrochemical ozonesondes atmospheric ozone is measured via an electrochemical reaction of ambient air bubbling in a solution of potassium iodide (KI), by means of a stable miniature pump. In a Brewer-Mast sonde two electrodes of

different metal are immersed in a buffered KI solution (Brewer and Milford, 1960), while Electrochemical Concentration Cell (ECC) sondes consists of two half cells with different solutions of KI as electrodes (Komhyr, 1969). The ozonesonde is launched in tandem with a radiosonde that also transmits air pressure, temperature, humidity and wind data to a ground station. With a 20-30s response time of the ozone cells and an ascent rate of about 6 m s^{-1} , the effective vertical resolution of the ozone signal lies nowadays around 150 m. Before the age of the digital sounding systems era the vertical resolution was coarserless due to the manual sampling technique by the operator, providing which delivered only measurements at significant levelspoints selected by the operator.

Regular measurements with ozonesondes started in the second half of the 1960s at a few sites: in 1965 at Aspendale (Australia, but moved to other suburbs of Melbourne thereafter, i.e. Laverton and Broadmeadows), in 1966 at Resolute Bay (Canada), in 1967 at Hohenpeissenberg (Germany), in 1968 at Payerne (Switzerland) and at Tateno (Tsukuba, Japan), in 1969 at Uccle (Belgium) and Sapporo (Japan), and in 1970 at Wallops Island (USA). These ozone sounding stations provide the longest time series of vertical ozone distribution. at a single site. In the Southern hemisphere there are measurements in Australia since 1965, but from different locations. Up to an altitude of about 30 km, ozonesondes constitute the most important data source with long-term data coverage for the derivation of ozone trends with sufficient vertical resolution, particularly in the climate sensitive altitude region around the tropopause. Furthermore, ozonesondes are widely used to study photochemical and dynamical processes in the atmosphere or to validate and evaluate satellite observations and their long term stability (Smit and ASOPOS panel, 2014, and references therein).

In this paper, we focus on the ozonesonde measurements at Uccle, covering 50 years, demonstrating its scientific relevance and the major achievements. Ozonesondes are still the only technique able to measure the ozone concentrations from the surface all the way up to the middle stratosphere with very high (absolute) accuracy and vertical resolution. Therefore, they have many application areas in which they are crucial: (i) quantifying the long-term variability in stratospheric and tropospheric ozone, (ii) as backbone for satellite validation, with satellites mostly measuring ozone only in stratosphere or upper troposphere, and (iii) for process studies in stratospheric-tropospheric exchange, and chemical production/destruction of ozone. The strength and uniqueness of the ozonesonde measurements, and in particular of ~~our~~ the long-term and very dense Uccle dataset, lie in combining all those different aspects of ozone research. In this paper, we will first give a description of the ozonesonde measurements at Uccle from a historical point of view (Sect. 2) and describe briefly which data processing has been applied to the ozonesonde measurements used in this paper (Sect. 3). In Section 4, we assess the time evolution of ozone at Uccle at different vertical layers against the background of recent findings in ozone variability. The fifth section illustrates the important role of the Uccle data for the validation of satellite ozone retrievals. Finally, in Section 6, concluding remarks and perspectives are given.

2 The Uccle ozone measurements: a historical overview

1025 In this section, we give a brief overview of the history of the ozone measurements at Uccle (Brussels, Belgium, 50°48'N, 4°21'E, 100 m asl). We explain why the ozone sounding program [has been was](#) initiated more than 50 years ago and discuss the presence of a period of gaps in the time series (Sect 2.1). We also describe which efforts have been taken during this time period to guarantee the homogeneity of the time series of ozonesondes between ascent and descent profiles (Sect. 2.2.1), with changing environmental conditions (Sect. 2.2.2), and between different ozonesonde types (Sect. 2.2.3). [We only give a brief description here, and refer to all the relevant earlier publications for more details.](#)

2.1 The start of the ozone observations

1030 The ozone sounding program at the Royal Meteorological Institute of Belgium (RMI) at Uccle was initiated by Prof. Jacques Van Mieghem, director of RMI from 1962 to 1970. Initially the ozone soundings were not performed out of a concern for possible human influence on the ozone layer, but rather to use ozone as a tracer to study the general air circulation in the troposphere and the lower stratosphere. Therefore, from the beginning it was planned to perform regular ozone soundings three times per week (on Monday, Wednesday, and Friday).

1035 In 1965 and 1966 the first few soundings were performed with Regener chemiluminescent ozonesondes, [and these data are still available at the World Ozone and Ultraviolet Radiation Data Centre \(WOUDC\)](#). A well-known effect of this sonde type is that it shows changes in sensitivity during the ascent trajectory (see e.g. Hering and Dütsch, 1965). For that reason it was decided to switch to Brewer-Mast electrochemical ozonesondes (developed by Brewer and Milford (1960) and commercially produced by the Mast Development Company at Iowa, USA) at RMI from November 1966 onwards. Based on a number of criteria such as continuity of the measurements and how well the preparation of the sondes was documented, it was decided to use the ozone soundings for scientific studies only from 1969 onwards, when Dirk De Muer took over the ozone research at RMI (in July 1969).

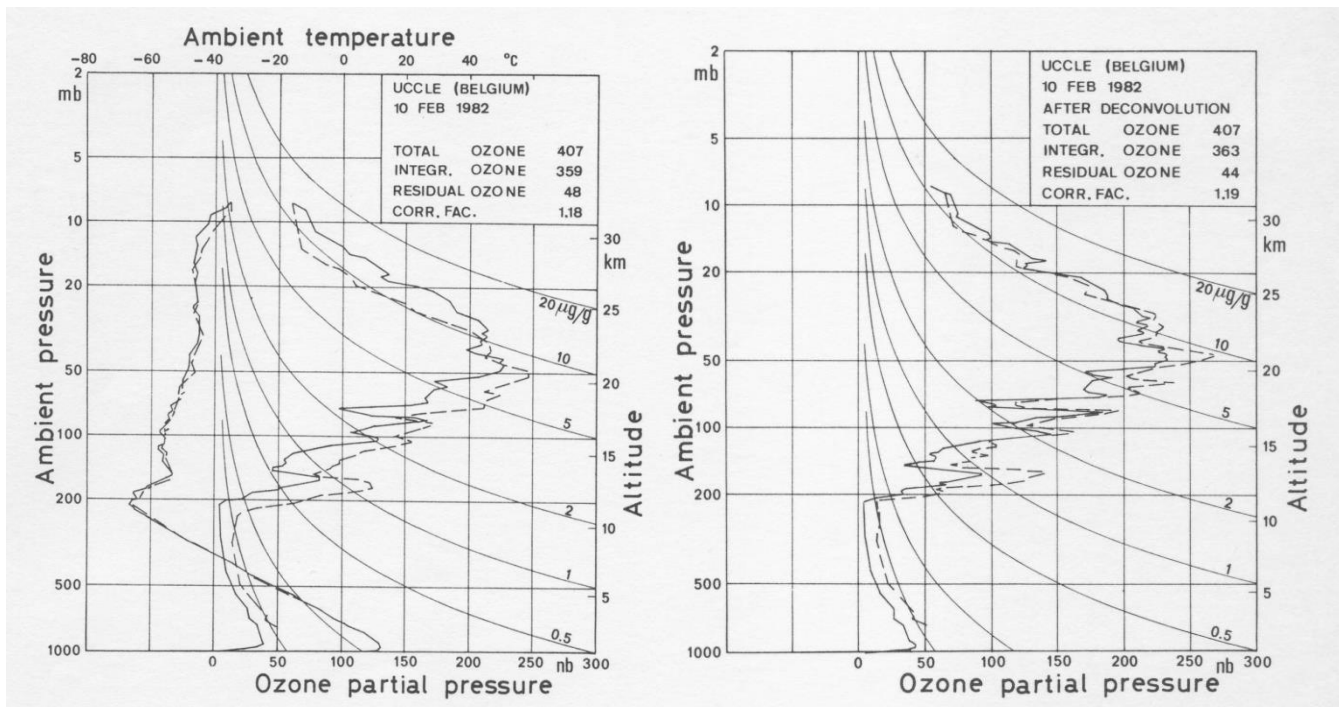
1040 In the period from February 1983 to January 1985 there were only a few ozone soundings. This gap in our time series was due to [financial problemsfunding reductions](#). Later on, when the Uccle time series of ozone soundings had proved its scientific value and with the growing concern of a human influence on the ozone layer, the continuation of the soundings became less an issue. In the course of time different radio sounding systems have been used. A major change occurred in 1990 when digital data transmission at high sampling rate was introduced, which allowed a higher vertical resolution of the profiles (not only at significant and standard pressure levels).

1045 To normalize the integrated ozone amount of the ozone soundings [\(essential for BM ozonesondes, see Sect. 2.2.3\)](#), the Dobson spectrophotometer (no. 40, D40) at Uccle was used since July 1971; before that date an interpolation of values from other Dobson stations in the European network was employed. In 1984, the Uccle site was equipped with a single Brewer UV spectrophotometer (no. 16), and with a double Brewer instrument (no. 178) in September 2001, to provide total ozone column measurements.

2.2 Challenges

1055 2.2.1 Frequency response of the electrochemical ozonesonde

In 1970 the ozone sounding program was adapted to gather also the data during the descent of the sonde after balloon burst. De Muer (1981) found that the measured ozone concentrations in the lower stratosphere and the troposphere were systematically higher during the descent than during the ascent (see Fig 1, left panel). ~~Two~~As possible explanations were mentioned: (i) a contamination of the ozonesonde at the ascent (e.g. by reducing constituents in the atmospheric boundary layer, see Sect. 2.2.2) and/or (ii) the response time of the sensor. ~~were mentioned. Therefore,~~To investigate the latter, De Muer and Malcorps (1984) analysed the frequency response of the combined ozone sensor and air sampling system of Brewer-Mast ozonesondes by means of a Fourier analysis. They found three different time constants: (i) a first-order process with a time constant of about 17 to 25 s (depending on the solution temperature) caused by the formation of iodine in the solution, (ii) a time constant of 7s, likely to be caused by the diffusion of iodine molecules to the platinum cathode, and (iii) a time constant of about 2.8 min that was explained by another diffusion process, i.e. an adsorption and subsequent desorption process of ozone at the surface of the air-sampling system. The slow first-order process with a time constant of about 20-25 min (found by Salzman and Gilbert, 1959, and taken up by Vömel et al., 2020, and Tarasick et al., 2021) could not be identified, probably because the impact of this process for a 0.1% KI solution would be too small (being 10% of the fast process for a 1% KI solution), as noted in De Muer and Malcorps (1984). ~~They concluded that the measured frequency response can be represented by a first order process and a diffusion process in series. A study of the dependency of the global transfer function of the sensor system as a whole on solution temperature and on background ozone level provided better insight into the performance of electrochemical ozonesonde, so that~~With these findings and time constants, a method for deconvolution of the ozone profiles through a process of Fast Fourier transform ~~could be~~was developed, ~~and:~~ aAn example of an ozone profile before and after deconvolution is also shown in Figure 1. After deconvolution the observed ozone values during the descent are still larger than the ascent values in the troposphere and the lowest layers in the stratosphere, which was then attributed to the effect of SO₂ on the ozonesonde measurements in the boundary layer.



1080 Figure 1: Ozone sounding at Uccle on 10 February 1982 with a Brewer-Mast ozonesonde before (left) and after (right) deconvolution of the ozone profile for both ascent (solid line) and descent (dashed line) of the sonde. In the left panel, the vertical profile of the air temperature is also shown (figure taken from De Muer & Malcorps, 1984).

2.2.2 The impact of the boundary layer SO₂ concentrations on the ozone measurements

1085 As SO₂ has even stronger absorption bands than ozone in the UV 305–340 nm wavelength range used for the total ozone determination with a Dobson spectrophotometer, total ozone amounts might be overestimated with this instrument in case of a considerable total vertical SO₂ column amount, as discussed by Komhyr and Evans (1980). In the suburban area of Uccle, the SO₂ densities near the ground were quite elevated at the start of the ozone measurements, but showed a steep decrease from the late 1960s to the early 1990s (Fig. S1). A comparison between quasi-simultaneous total ozone observations at Uccle with a Dobson and a Brewer spectrophotometer showed that there was no drift in the difference between the two datasets if the effect of SO₂ was taken into account (De Backer and De Muer, 1991). It was also found that the SO₂ correction of the Dobson spectrophotometer D40 data had a significant effect on the calculated total ozone trend (De Muer and De Backer, 1992), and made this trend consistent with the one derived from reprocessed Total Ozone Mapping Spectrometer (TOMS) satellite data for a sub-period in which both datasets were available.

1090 Ozonesonde measurements by the KI method are sensitive to interference by oxidizing or reducing agents (e.g. Tarasick et al., 20210, and references therein). In particular, one SO₂ molecule cause a reverse current of two electrons, reducing the electrochemical cell response on a 1:1 basis~~can cause an important reduction in the ozone detected of 100% (one SO₂~~

~~molecule cause a reverse current of two electrons~~), and excess SO₂ can accumulate in the cathode solution, affecting ozonesonde measurements well above the polluted boundary layer (Komhyr, 1969, De Muer and De Backer, 1993, see also Fig.1 and Fig. S1) or near volcanic sites (~~–~~Morris et al., 2010), see also Fig. 1 and Fig. S2. Furthermore, in case of a considerable total vertical SO₂ column amount, the Dobson total ozone amounts might be overestimated as SO₂ has even stronger absorption bands than ozone in the UV 305-340 nm wavelength range used for the total ozone determination (Komhyr and Evans, 1980). As a matter of fact, in the suburban area of Uccle, the SO₂ densities near the ground were quite elevated at the start of the ozone measurements, but showed a steep decrease from the late 1960s to the early 1990s (Fig. S2). As a consequence, the variation of SO₂ density near the ground has a twofold effect on ozone soundings with electrochemical sondes: (i) the integrated ozone amount of the (BM) soundings is normalized by means of spectrophotometer data, so that a trend in the latter data will lead to an effect on ozone trends from soundings, and (ii) due to the SO₂ interference with the ozonesonde cell reactions, any trend of SO₂ causes a distortion of ozone profile trends as a function of altitude.

To minimize this double impact of SO₂ on the ozonesonde ozone measurements, two corrections were developed. Based on the comparison between quasi-simultaneous total ozone observations at Uccle with a Dobson and a Brewer spectrophotometer (De Backer and De Muer, 1991), a model connecting SO₂ column readings with long-term surface SO₂ monitoring measurements was able to subtract a fictitious trend in the Dobson. Applying this correction made the Dobson total ozone trend consistent with both the Brewer trend and the one derived from reprocessed Total Ozone Mapping Spectrometer (TOMS) satellite data for the sub-periods in which both datasets were available (De Muer and De Backer, 1992). Furthermore, a

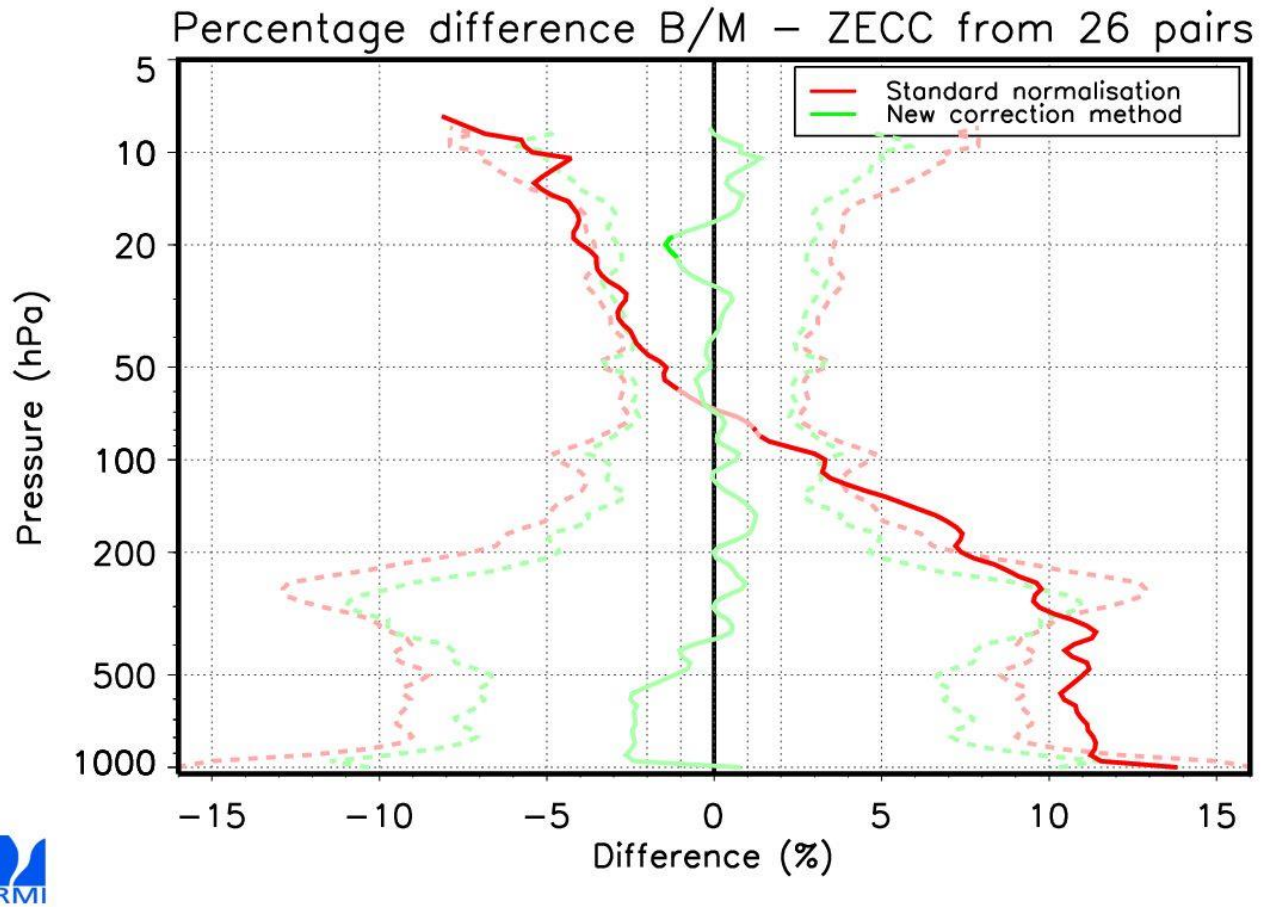
~~As a consequence, the variation of SO₂ density near the ground has a twofold effect on ozone soundings with electrochemical sondes: (i) the integrated ozone amount of the soundings is normalized by means of spectrophotometer data so that a trend in the latter data will lead to an effect on ozone trends from soundings, and (ii) due to the SO₂ interference with the ozonesonde cell reactions, any trend of SO₂ causes a distortion of ozone profile trends as a function of altitude. A~~

method to calculate the vertical SO₂ distribution associated with each ozone sounding was developed, based on two assumptions: a constant SO₂ mixing ratio from the ground to the mixing layer height, and an exponentially decreasing mixing ratio above the mixing layer balancing the integrated SO₂ amount to the reduced thickness of the SO₂ layer (De Muer and De Backer, 1993). The effect of ~~those two~~this correction~~s~~ for SO₂ interference on the vertical ozone trends in the 1969-1996 Brewer-Mast period is illustrated in Fig. S3. It shows that those corrections are essential in assessing the trends in tropospheric ozone at Uccle until the mid nineties.

2.2.3 The transition from BM to ECC sondes

As mentioned before, at the start of the operational ozone sounding series, the Brewer-Mast sensor type ~~of sensor~~ was used. This type of ozonesonde had several issues at that time: (i) a strong reduction of the efficiency of the pump at low pressure

1130 (De Backer et al., 1998a), (ii) the loss of ozone in the sensor itself, causing a relatively high (up to 20%) underestimation of
the integrated ozone from a sounding profile with respect to the total ozone measured with a Dobson or a Brewer
spectrophotometer, and (iii) a variable response in the troposphere, depending on preparation (Tarasick et al., 2002).
Therefore, in the middle of the 1990s, RMI investigated the switch from the BM sondes to the ECC sensors (Komhyr, 1969),
which seemed to perform better and were easier to prepare before launch. To document this transition, ~~double-dual~~
1135 soundings were launched about twice a month during one year. The comparison between both sensor types on those dual
soundings is shown in Fig S42. If standard correction methods for both sensors are used, large statistically significant
differences appear: Brewer-Mast sensors overestimate tropospheric ozone and underestimate stratospheric ozone, mainly due
to the standard normalisation by linear scaling of the vertical ozone profile for BM sondes. ~~Therefore~~. De Backer et al.
(1998a,b) developed ~~onea PRESSsure and Temperature dependent Total Ozone normalization correction method~~ (now called
1140 PRESTO, ~~for PRESSsure and Temperature dependent Total Ozone normalization~~, see Van Malderen et al., 2016) correction
method for both ozonesonde types, based on (i) measurements of the pump efficiencies of both ozonesonde types ~~y~~ in a
pressure chamber at Uccle, (ii) ~~and on~~ a pre-flight comparison of every ozonesonde with a calibrated ozone source in the lab,
and (iii) the comparison with the total ozone column measured with ~~thea~~ co-located ozone spectrophotometer (full practical
details are available in De Backer, 1999). ~~This method is still the operational one at Uccle and has been used to process all~~
1145 the ozonesonde data here (see Section 3). By applying this method, the differences between the dual ozone sounding profiles
are reduced below 3% almost throughout the whole profile and below the statistical significance level (see again Fig. S42).
The impact of this new pump correction method on the vertical ozone trends is also significant, especially for the 1969-1996
BM period (see Fig. S3 and also for other periods in Van Malderen et al., 2016).
Further validation of the method was done by comparing the profiles with measurements from the SAGE II satellite
1150 instrument (Lemoine and De Backer, 2001). This study showed that the PRESTO correction removed the jump, caused by
the BM to ECC transition, in the difference time series with SAGE II at low pressures (compare Fig. 1 and 2 in Lemoine and
De Backer, 2001).



1155 **Figure 2: Mean percentage differences between ozone profiles obtained with Brewer-Mast and (En-Sci model Z) ECC sensor during dual soundings with standard correction method (red) and with the PRESTO method (green). Thick lines denote the pressure levels where the differences between the mean profiles are statistically significant. The dashed lines are the one sigma variance of the distribution of the differences [fig. adapted from De Baeker et al, 1998b].**

3 The Uccle ozonesonde dataset

1160 In this paper, the PRESTO correction has been applied to the entire ozonesonde dataset, i.e. to both the BM and ECC
 ozonesonde types, but with the appropriate different measured pump efficiency coefficients at Uccle for ~~the different~~both
 types, to ensure consistency over the entire data record of 50 years. Although a total ozone normalization is not required for
 the ECC sonde measurements (Smit and ASOPOS panel, [1994](#)[2014](#)), it is applied for the entire Uccle time series within the
 PRESTO correction. To calculate the residual ozone above the balloon burst level, we use a combination of the constant
 1165 mixing ratio approach and the climatological mean obtained from satellite ozone retrievals (McPeters and Labow, 2012). An
 alternative, homogenized, Uccle ozonesonde corrected dataset is available by request from the authors for the ECC time
 series since 1997 (Van Malderen et al., 2016), following the principles of the Ozonesonde Data Quality Assessment (O3S-

DQA) activity (Smit et al., 2012), but is not used here to maintain the consistency over the entire time series. Differences between both versions of corrected Uccle ECC ozonesonde data, in comparison with [the nearby ozonesonde site De Bilt](#) (the Netherlands, 175 km north of Uccle), are highlighted in Van Malderen et al. (2016).

For the BM ozonesondes, the applied PRESTO corrections include (i) a correction for SO₂ interference on the ozone soundings (imperative to have reliable lower-tropospheric ozone trend estimates for the period 1969-1996, see Fig. S3), (ii) a correction for a negative background current caused by impurities in the sensor before October 1981, (iii) a correction for box temperatures depending on the insulating capacity of the Styrofoam boxes (a short discussion of those additional corrections and the proper references are given in the appendix A of Van Malderen et al., 2016), and (iv) an altitude correction for VIZ/Sippican radiosonde pressure measurements based on comparisons with wind-finding radar. Without this altitude correction, ~~(calculated~~ sonde altitudes were too low [up to 1000m at an altitude of 30km, De Muer and De Baeker, 1994](#)), so that the calculated ozone concentrations with VIZ radiosondes were too low by 7.5 to 14%, depending on the manufacturing series of radiosondes (De Muer and De Baeker, 1994). ~~From~~ [Since](#) 1990, the ozonesondes were combined at Uccle with Vaisala RS80 radiosondes, which showed a much smaller difference of the calculated altitude with respect to wind-finding radar data. Therefore, for the digital era period since 1990, no radiosonde pressure sensor bias corrections have been applied, although they have been identified in different studies (e.g. De Baeker, 1999; Steinbrecht et al., 2008; Stauffer et al., 2014; Inai et al., 2015).

4 Temporal evolution of the vertical ozone concentrations at Uccle

As ozonesondes are the only devices that are able to measure ozone concentrations from the surface up to the middle stratosphere with high vertical resolution, they are very suitable to study and relate the temporal variability of ozone in different atmospheric layers. The evaluation of the temporal variability of the ozone measurements at Uccle is therefore organized in different sections. We first describe ~~the total ozone temporal evolution (Sect. 4.1), continue with~~ the stratospheric (Sect. 4.1.2) and tropospheric (Sect. 4.2.3) ozone trends. [The relation to other co-located ozone measurements is described in the appendices. Total ozone trends are treated in Annex A and the, and we wrap up with the](#) temporal behaviour of surface ozone and several ozone depleting substances [is discussed in Annex B\(Sect. 4.4\)](#).

~~4.1 Total ozone trends~~

~~The total column ozone amounts at Uccle, available since 1971, are retrieved with a Dobson UV spectrophotometer (no. 40, 1971-1989), a single Brewer UV spectrophotometer (no. 16, 1990-current, but used in the time-series until the end of 2001), and a double Brewer UV spectrophotometer (no. 178, 2002-current). The calibration history of the Dobson instrument documented in De Muer and De Baeker and the transition to the Brewer instrument is described in De Baeker and De Muer, 1991. The time-series of total ozone measurements is shown in Fig. 3, but has been smoothed by applying a low pass Gaussian filter with a width at half height of 12 months, to filter out variations with frequencies higher than one year. With~~

1200 this representation, the impact of the major (strato)volcanic eruptions of Fuego (Guatemala, Oct 1974), El Chichon (Mexico,
Mar/Apr 1982), and Pinatubo (the Philippines, Jun 1991) on the Uccle total ozone concentrations pop-ups illustrates the
significant dips in Uccle total ozone. Indeed, the episodes of enhanced stratospheric aerosol related ozone loss after those
major volcanic eruptions are confirmed by model results (see e.g. Tie and Brasseur, 1995, Solomon 1999, Aquila et al., 2013
1205 for a description of the mechanism behind) can clearly be identified in the time series. The inter annual variability in Fig. 3
is very similar to the Northern Hemisphere (NH) annual mean total ozone time series of five bias corrected merged datasets
in the 35–60° N latitude band in Weber et al. (2018; their Fig. 2). We refer to this paper for the discussion of several features
(e.g. the excess total ozone in 2010 and the 2011 and 2016 low ozone anomalies in 2011 and 2016).

The long term temporal variability of the total ozone amounts at Uccle is dominated by the 1980–1997 ozone decline (at a
rate of 2.5%/decade) due to the anthropogenic production of ozone depleting substances (ODS), transported into the
stratosphere, with peak concentrations in 1997. Subsequently, in the late 1990s, the annual mean total ozone started to
1210 increase again (at a rate of 2%/decade at Uccle for the period 1997–2018). This increase was due to the combination of the
slow decrease in ODSs and of atmospheric dynamics, notably ozone transport via the Brewer–Dobson circulation, causing
also the interannual variability described in Weber et al. (2018). It should also be noted that the strongest increase of the total
ozone amounts since the beginning of this century took place in Uccle in late winter—early spring (Feb–Apr), at a rate of 3–
4%/decade, while the ozone transport by the Brewer–Dobson circulation from its tropical source region poleward and
1215 downward into the lower stratosphere is strongest during wintertime (e.g. Butchart, 2014; Langematz, 2019).

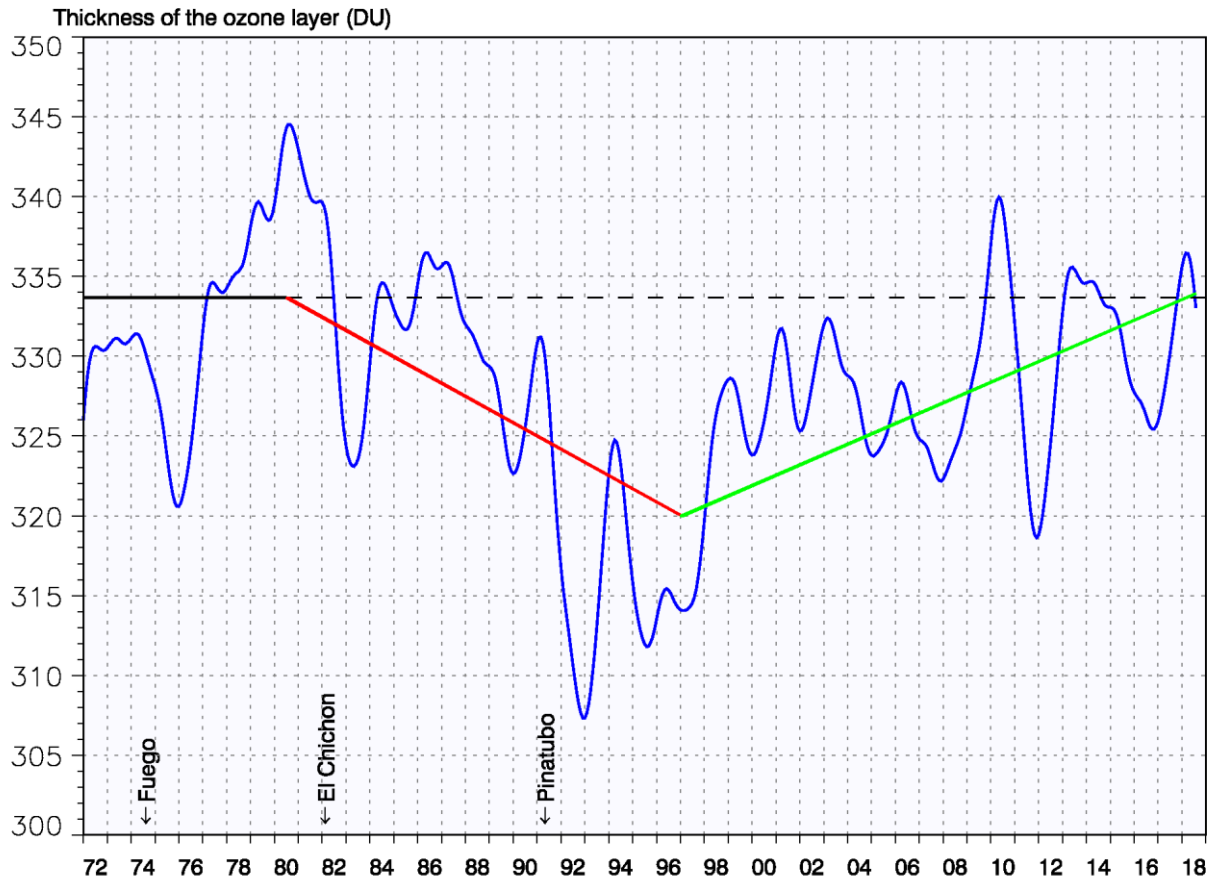


Figure 3: Evolution of the total ozone column at Uccle as observed with Dobson D40 (1972-1989), Brewer 16 (1990-2001), and Brewer 178 (2002-present). Linear trends during the periods 1980-1997 and 1997-2018 are shown respectively in red and green. The horizontal black full line marks the 1972-1980 total ozone average, extended until the end of the time series by the dashed horizontal line. The periods of major volcanic eruptions affecting the ozone layer are indicated on the time axis as well.

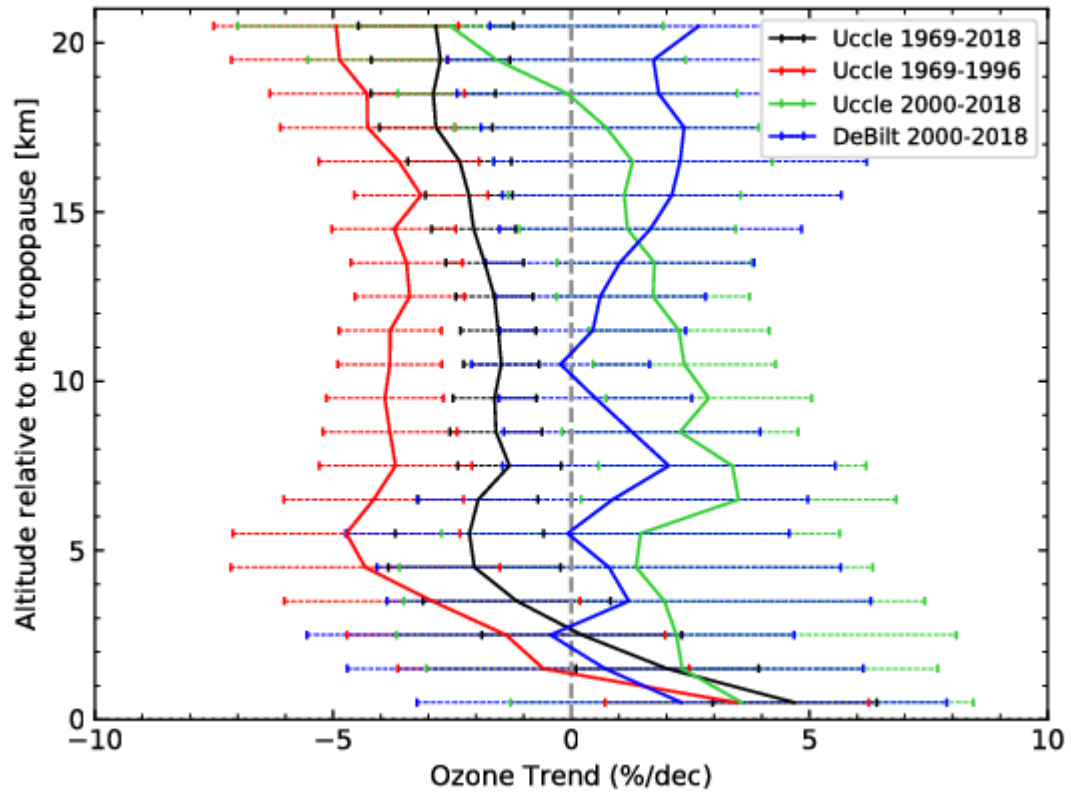
4.24.1 Stratospheric ozone trends

For calculating the vertical distribution of trends in the stratospheric ozone concentrations from the Uccle ozonesonde data, we use the altitude relative to the tropopause height as the vertical coordinate. The tropopause applied here is the standard (first) thermal tropopause (WMO, 1957), and is derived from the vertical temperature profiles measured by the Uccle radiosondes, as described in Van Malderen and De Backer (2010). The implemented statistical model to calculate trends is the Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS) multiple linear regression (MLR) model (SPARC/IO3C/GAW, 2019). This model uses an independent linear trend (ILT) method as a trend term, which is based on two different, independent, trends to describe the ozone decrease until 1997 (ODS increase) and the slow ozone increase since the early 2000s (after the turnaround in ODS concentrations). These two periods have been used since WMO (2014)

1230 and their use avoids endpoint anomalies near the turnaround in 1997 for the two independent linear trend terms in the ILT method. Additionally, the LOTUS regression includes two orthogonal components of the quasi biennial oscillation (QBO), the 10.7 cm solar radio flux, El Niño Southern Oscillation (ENSO) without any lag applied, and the Aerosol Optical Depth (AOD, extended past 2012 by repeating the final available value from 2012 as the background AOD, which should be a valid assumption for Uccle). Four Fourier components representing the seasonal cycle are also included, unless (relative) monthly anomaly series are used as input ozone data. The output of the LOTUS MLR model and the different contributing terms (or proxies) for the monthly anomaly ozone concentrations at the layer 10 km above the tropopause (close to the ozone peak) are shown in Fig. S5. The final choice of those proxies (and possible lags) in LOTUS was based on retaining the optimal regression for global analysis of satellite data and broad latitude band analyses. Therefore, proxies describing rather local or small-scale phenomena might not have been included in the general "LOTUS regression" model. In particular, using an alternative stepwise multiple linear regression model for the Uccle stratospheric ozone amounts, we found that the Uccle tropopause pressure and the Arctic Oscillation are significant proxies as well (contributing statistically significant, i.e. at the 95% significance level of the t -test, to the regression coefficient). However, here, the analysis is limited to the LOTUS model and the sensitivity of the estimated trends on the chosen (M)LR model is rather limited for the Uccle time series.

1240 The vertical profile of stratospheric ozone trends is shown in Fig. 24. From 1969 to 1997, stratospheric ozone concentrations decrease almost uniformly (and significantly) at a rate around $-4\%/\text{dec}^{-1}$ ade, except at the layers just above the tropopause. Since 2000, the stratospheric ozone concentrations increase with about $\pm 2\%/\text{dec}^{-1}$ ade, but only significantly at the layers below and at the ozone maximum (from 6 to 13 km above the tropopause, or 17 to 24 km for an average tropopause height of 11 km at Uccle). The insignificant negative trend of the Uccle ozone concentrations at the upper levels of Fig. 24 should be treated with caution, as the reliability of the ozonesonde instrument at those levels (above 30 km) is reduced. This is due to the increasing uncertainty in the pump efficiency at low pressures, the different stoichiometry of the chemical reaction due to a reduced amount of sensing solutions by evaporation, and frozen solutions, etc. Additionally, an increase of the burst altitude in the Uccle ozonesonde time series in recent years and inhomogeneities due to changing pressure sensors with different radiosonde types might have an impact on the ozone trends at these very low pressures. In fact, the negative ozone trends are also less pronounced if calculated for absolute altitude levels. However, also for these altitudes, we prefer to calculate the vertical ozone trends in altitudes relative to the tropopause, to cancel out the seasonal variation of the ozone peak altitude, which roughly follows the tropopause height variation at Uccle: the ozone maximum peak is at its highest altitudes in summer (when the tropopause is also located higher), and is located at lower altitudes in winter (with the lowest tropopause). This approach gives in general vertical ozone trends that vary less over the different altitude levels. When we compare the post-2000 trends with those from the ozonesondes launched at De Bilt, the overall stratospheric positive insignificant trends apply for both stations, also at the higher altitude levels at De Bilt. The larger trend uncertainties for the De Bilt time series can be explained by the smaller frequency of launches (once a week versus three times a week at Uccle). The statistically insignificant offset between the Uccle and De Bilt trend estimates depends on the used correction methods at both sites, but also differences in the vertical ozone distribution (up to 5% in the stratosphere), of both geophysical and

instrumental origin, have an impact on the trend values (see e.g. Figs. 10a and 12 in Van Malderen et al. (2016), in which a more detailed explanation of the differences in vertical ozone distribution and trends between Uccle and De Bilt is given). The lower-stratospheric ozone trends deserve more discussion here, as Ball et al. (2018, 2019) reported a significant decline in lower stratospheric (13-24 km) ozone amounts for the periods 1998-2016 and 1998-2018 respectively, from multiple (merged) satellite measurements in the lower stratosphere between 60°N and 60°S. Also the latest Scientific Assessment of Ozone Depletion (WMO, 2018), largely based on the LOTUS final report (SPARC/IO3C/GAW, 2019), concluded that “there is some evidence for a decrease in lower stratospheric ozone from 2000 to 2016”, although not statistically significant in most analyses. This decline, contradictory to the decline of ozone-depleting substances since 1997, is surprising and the current state-of-the-art chemical climate models (CCMs) used in Ball et al. (2020) and Dietmuller et al. (2021) do not show a decrease, but rather an increase of the lower-stratospheric mid-latitude ozone, although they confirm the lower-stratospheric ozone decline in the tropics in the observations. Using the Modern-Era Retrospective Analysis for Research and Applications Version 2 (MERRA-2) ozone output fields, Wargan et al. (2018) found a discernible negative trend of -1.67 ± 0.54 Dobson units dec^{-1} in the 10-km layer above the tropopause between 20°N and 60°N, and attributed the trend to changes driven by dynamical variations (as Chipperfield et al., 2018), in the form of enhanced isentropic mixing between the tropical (20°S–20°N) and extratropical lower stratosphere in the past two decades. In a follow-up study, Orbe et al. (2020), demonstrated that in the NH, this mid-latitude ozone decrease is primarily associated with changes in the advective circulation rather than changes in mixing. In this study, both the Uccle and De Bilt time series do not show a significant decline in lower stratospheric (13-24 km) ozone amounts. On the contrary, –as reported by Ball et al. (2018, 2019) for the periods 1998–2016 and 1998–2018 respectively, from multiple (merged) satellite measurements in the lower stratosphere between 60°N and 60°S. Using the Modern Era Retrospective Analysis for Research and Applications Version 2 (MERRA-2) ozone output fields, Wargan et al. (2018) found a discernible negative trend of -1.67 ± 0.54 Dobson units per decade (DU/decade) in the 10 km layer above the tropopause between 20°N and 60°N. Also the latest Scientific Assessment of Ozone Depletion (WMO, 2018), largely based on the LOTUS final report (SPARC/IO3C/GAW, 2019), concluded that “there is some evidence for a decrease in lower stratospheric ozone from 2000 to 2016”, although not statistically significant in most analyses. This decline, contradictory to the decline of ozone depleting substances since 1997, is attributed to changes driven by dynamical variations (Chipperfield et al., 2018), in the form of enhanced isentropic mixing between the tropical (20°S–20°N) and extratropical lower stratosphere in the past two decades (Wargan et al., 2018). However, although never significant, we found that the positive Uccle ozone trends in the lower stratosphere are rather robust, independent of the starting date (1997/1998/2000), the used vertical coordinate system (absolute or relative to the tropopause), and the trend model used (LOTUS MLR or simple linear fit). The lower stratospheric ozone trends derived from the De Bilt time series show a larger variability between positive and negative statistically insignificant values, especially in the ten lowest kilometres.



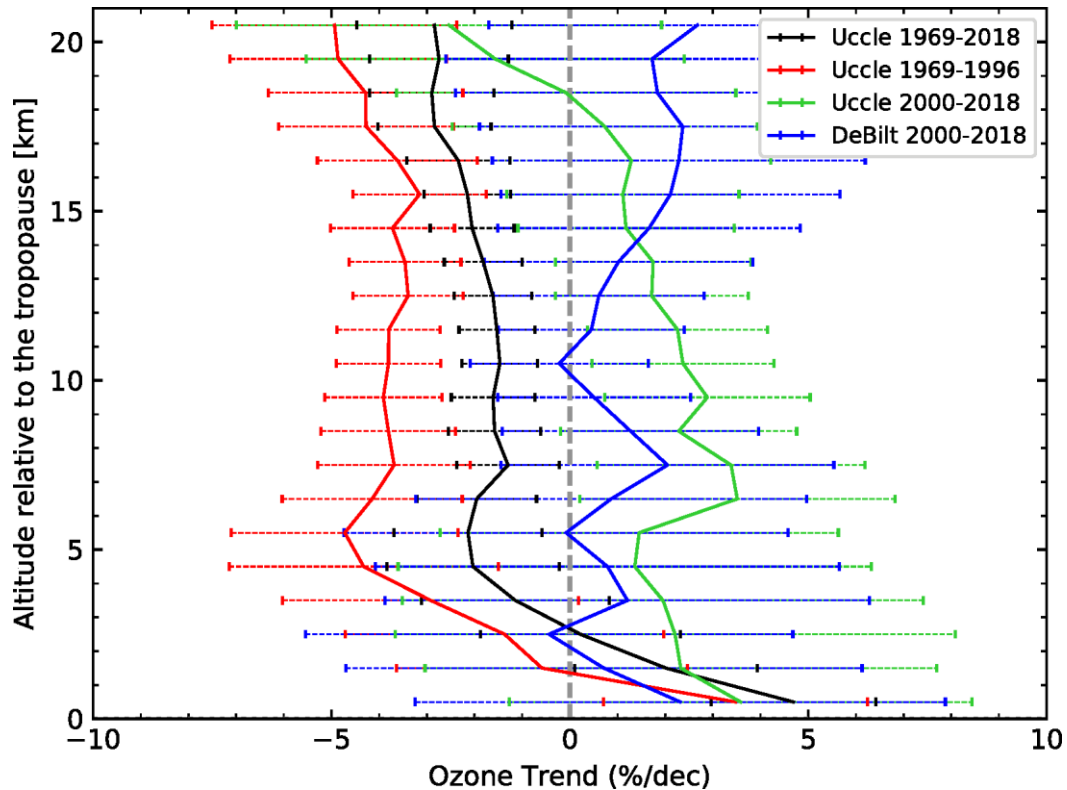


Figure 24: Vertical distribution of trends of stratospheric ozone concentrations at Uccle for different periods (see text) and at De Bilt (2000-2018). The trends and their uncertainties are calculated with the LOTUS multiple linear regression model (see text and SPARC/IO3C/GAW, 2019), including an independent linear trend term. The 2-sigma error bars represent the trend uncertainty estimated by the regression model (using the fit residuals). For the Uccle 1969-2018 time series only, one linear trend term is included in the model instead. The output of the LOTUS MLR model and the different contributing terms for the monthly anomaly ozone concentrations at the layer 10 km above the tropopause are shown in Fig. S5.

Ball et al. (2020) investigated if the aforementioned changes in ozone and transport are also found in other stratospheric variables like the temperature. Globally, a reduction in lower stratospheric ozone should lead to reduced radiative heating and a decrease in observed temperature (see references in Ball et al., 2020). Quasi-global lower stratospheric temperatures from observations and in CCMs indeed decreased, with the post-2000 negative temperature trend being smaller compared to pre-1998, mimicking the observed lower-stratospheric ozone trends (Ball et al., 2020, but also Maycock et al., 2018), although not the modelled ozone increase after 2000. On a smaller (European) scale, Philipona et al. (2018) found very similar seasonal and annual changes for temperature and ozone when averaging the Payerne, Hohenpeissenberg and Uccle ozonesonde measurements. With the exception of the fall season, annual and seasonal profiles switch from negative to positive trends before and after the turn of the century, for both ozone and temperature (see Fig. 4 in Philipona et al., 2018). Here, on the local scale of Uccle and De Bilt, we also investigated the link between the lower-stratospheric ozone and temperature trends (see Fig. S6). Before 1997, the entire stratosphere above Uccle cooled significantly by -0.9 to -0.5 $^{\circ}\text{C dec}^{-1}$, in line with the decreasing stratospheric ozone concentrations. After 2000, the stratospheric cooling at both Uccle and De

1320 Bilt ceased at the altitudes where ozone concentrations peak (see Fig. S6), and where their radiative impact on stratospheric temperatures is largest. Above and below the ozone maximum, the sign of the post-2000 temperature trends at Uccle (respectively positive and negative) and De Bilt (respectively negative and positive) are reversed. As such, there is no direct imprint of the slightly positive lower-stratospheric ozone trends since 2000 in the temperature variability, in particular for Uccle. However, this might not be expected on a local scale, and in addition to ozone, stratospheric temperatures are affected by radiative effects from CO₂, N₂O, CH₄, as well as stratospheric water vapour, and chemical changes in these gases (Ball et al., 2020). These authors point to the increasing stratospheric water vapour amounts in the CCMs since 1996 in the mid-latitudes, cooling the lower stratospheric, to reconcile the increasing lower-stratospheric ozone concentrations in the models with their stratospheric cooling over the same period and latitudes.

1325 ~~To understand the differences in the lower stratospheric ozone trends since the end of the nineties, we also consider the temperature trends in the stratosphere here (see Fig. S4), as the evolution of stratospheric ozone in a changing climate also depends on the cooling of the stratosphere due to increases in greenhouse gases (GHGs). Before 1997, the stratosphere cooled significantly by 0.9 to 0.5 °C/decade, corresponding with both the tropospheric warming due to the increase of GHGs and the decreasing stratospheric ozone concentrations (Philipona et al., 2018). After 2000, the stratospheric cooling at both Uccle and De Bilt ceased at the altitudes where ozone concentrations peak and where their radiative impact on stratospheric temperatures is largest. This stratospheric warming is however not significant. Above and below the ozone maximum, the sign of trends between Uccle (respectively positive and negative) and De Bilt (respectively negative and positive) are reversed. As such, there is no clear similar covarying behaviour between ozone and temperature changes in the lower stratosphere at these two sites. However, when averaging the Payerne, Hohenpeissenberg and Uccle ozonesonde measurements, Philipona et al. (2018) found very similar seasonal and annual changes for temperature and ozone. With the exception of the fall season, annual and seasonal profiles switch from negative to positive trends before and after the turn of the century, for both ozone and temperature.~~

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Finally, as we use the altitude relative to the tropopause as vertical coordinate, we should also mention the time variability of the tropopause height ~~here, which might impact the lower-stratospheric ozone trends~~. The tropopause height is increasing at both Uccle and De Bilt for all considered periods, but with different magnitudes: for Uccle, these are $6.98 \pm 1.12 \text{ m}_{\text{dec}}^{-1}$ (1969-2018), $13.81 \pm 3.00 \text{ m}_{\text{dec}}^{-1}$ (1969-1996), and $11.62 \pm 79.42 \text{ m}_{\text{dec}}^{-1}$ (2000-2018), while for De Bilt the post-2000 trend magnitude is $25.73 \pm 19.23 \text{ m}_{\text{dec}}^{-1}$. These increases in tropopause altitudes are consistent with results from the global study in Xian and Homeyer (2019) based on radiosondes and reanalyses, although with smaller magnitudes (they found increases of 40–120 m ~~per~~ dec^{-1} for the period 1981-2015). ~~With climate model experiments, Santer et al. (2003) ascribed the simulated rise in tropopause altitude over 1979–1999 to cooling of the stratosphere (caused by ozone depletion) and warming of the troposphere (caused by well-mixed greenhouse gases). The thermal expansion of the troposphere and the associated increase in tropopause height have been proposed as robust fingerprints of anthropogenic climate change based on multiple observational and model evidence (Santer et al., 2003, Seidel and Randel, 2006, Lorenz and DeWeaver, 2007).~~

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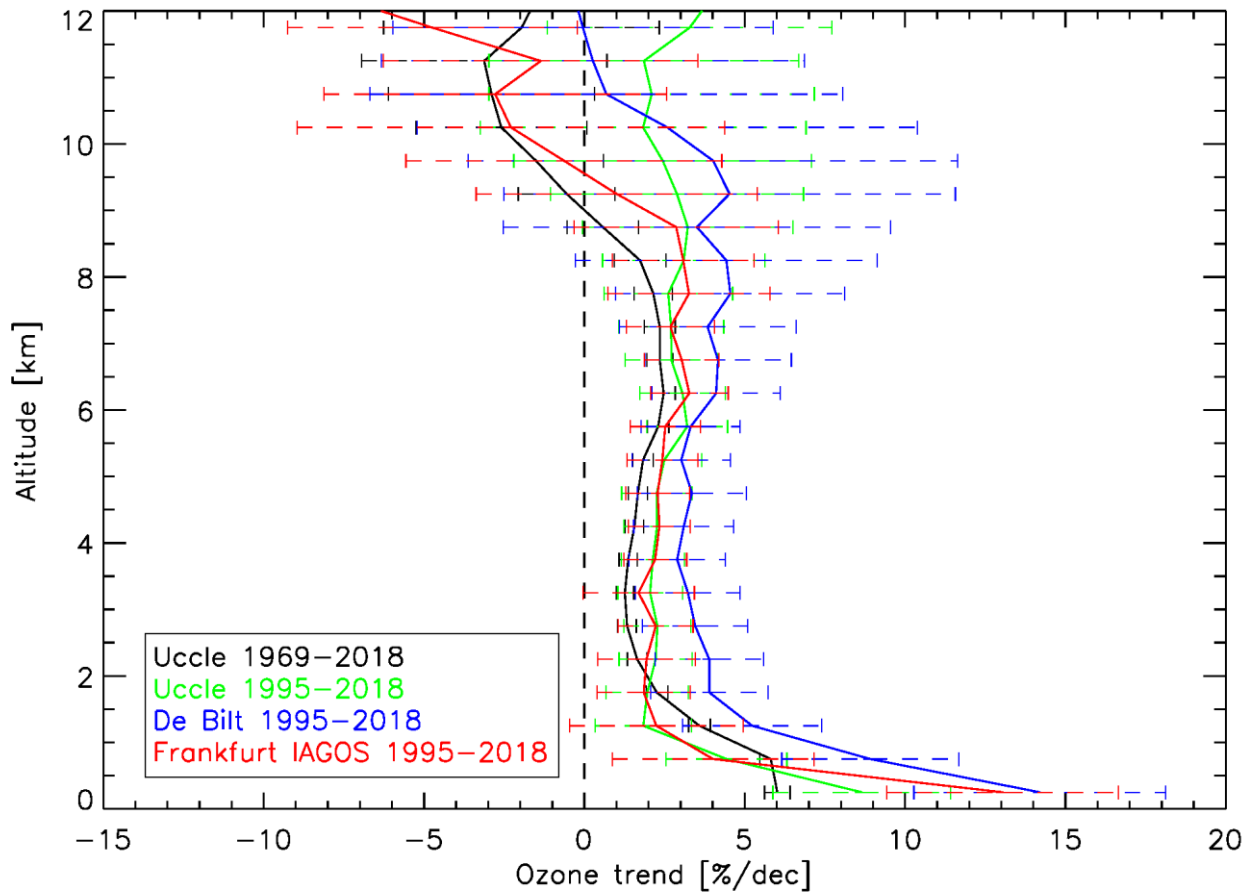
1345

1350 We can conclude here that the Uccle stratospheric ozone trends before 1997 are well understood, but that the behaviour after
2000 is harder to explain, especially for the lower stratosphere, because of ~~due to its interaction with~~ the lack of a clear link
with stratospheric temperature variability and the impact of the ~~and~~ tropopause variability, ~~hence due to dynamics, induced
by increasing GHG concentrations.~~ The link between the Uccle stratospheric ozone trends and these from the total ozone
column measured with co-located spectrophotometers is discussed in Annex A.

1355 4.34.2 Tropospheric ozone trends

Ozone in the troposphere is affected by many processes. Stratosphere-troposphere inflow and photochemical formation by
interaction with sun light and ozone precursors (NO_x, CO and Volatile Organic Compounds) increase the ozone levels, while
photochemical destruction of ozone in low NO_x conditions (e.g. marine boundary layer and free troposphere, through OH-
HO₂ cycle) or at high NO_x concentrations (urban regions under titration, i.e. via reaction with NO), and dry deposition on the
1360 ground removes ozone from the troposphere. Its short lifetime causes highly variable ozone concentrations in space and
time, which complicates the understanding of the processes at play at all relevant spatio-temporal scales (Young et al., 2018).
Moreover, the production of ozone in the troposphere is sensitive to variations in air temperature, radiation and other
climatic factors (Monks et al., 2015).

Tropospheric ozone is measured with ozonesondes, by commercial aircraft, with different types of ground-based remote
1365 sensing instruments and with satellite instruments. Besides clear regional differences, the distribution and trends of ozone in
the troposphere are not always consistent between these different datasets, and even not between different retrieval methods
of the same satellite (e.g. Cooper et al., 2014, Gaudel et al., 2018). In fact, measuring the vertical profile of tropospheric
ozone concentrations from satellites remains very challenging and relies on ground-based retrievals of ozone for validation
(see Sect. 5).



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Figure 35: Vertical distribution of trends of tropospheric ozone concentrations at Uccle for different periods, at De Bilt (ozonesonde data) and Frankfurt (IAGOS data) for 1995-2018. Simple linear trends are calculated for monthly ozone anomalies in 1 km altitude ranges and the error bars are the 2σ standard deviations. The same colour coding is used as in Fig. 2: trends for the most recent Uccle sub-period (here 1995-2018) are in green, for the entire Uccle time series trends, we use black and the De Bilt trends are shown in blue. The red line denote the Frankfurt IAGOS trends in this figure.

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Here, we calculated the tropospheric ozone trends from the Uccle and De Bilt ozonesonde time series and the MOZAIC (Measurement of Ozone and Water Vapour by Airbus in-service Aircraft) and IAGOS (In-service Aircraft for a Global Observing System) ascent and descent profiles at Frankfurt airport, at about 320 km from Uccle. This MOZAIC-IAGOS dataset consists of more than 27600 profiles, starting in August 1994, and is combined with the data from Munich airport, approximately 300 km southeast of Frankfurt, between 2002 and 2005 (about 4200 flights), to fill a large data gap in 2005 (also done in e.g. Petetin et al., 2016). With typical horizontal ozone correlation lengths of about 500 km in the troposphere (Liu et al., 2013), some correlation of especially free-tropospheric ozone trends between Uccle on one hand, and De Bilt and Frankfurt on the other hand, is expected. We used simple linear trends based on the monthly anomalies at different altitude levels (see Fig. 35) ~~for the period 1995-2018~~, as there is no consensus ~~in the used using (which)~~ proxies to account for

1380

1385 natural variability ~~in a multiple linear regression model as for stratospheric ozone (e.g. LOTUS)~~. First, for the 1995-2018
period, the extremely good agreement between the Uccle (in green in Fig. 3) and IAGOS (in red) vertical ozone trends in the
free troposphere (3-8 km) ~~for the 1995-2018 period~~ is striking. Although the integrated tropospheric ozone amounts for this
altitude range are lower for the region above Frankfurt (14.9 DU) than above Uccle (16.2 DU), the overall relative trends are
similar (resp. $2.09 \pm 1.01 \text{ \% /dec}^{-1}$ ~~ade~~ and $2.47 \pm 1.01 \text{ \% /dec}^{-1}$ ~~ade~~, see Fig. S75). The De Bilt trends (in blue in Fig. 3) are
1390 larger in the free troposphere, with also larger uncertainties, ~~probably~~ due to the lower launch frequency. In this context, we
mention ~~Note also~~ the sensitivity analysis of IAGOS profiles above Europe by Chang et al. (2020), ~~who determined which~~
concluded that an optimal sample frequency of 14 profiles per month is required to calculate trends with their integrated fit
method (and about 18 profiles a month when this method is not used). Near the surface, the De Bilt trend is in better
agreement with the Frankfurt trend, but the local surface ozone production and destruction and the boundary layer dynamics
1395 can vary substantially between the three sites considered here, so that the boundary ozone distribution and trends at the three
sites are likely to be uncorrelated. However, comparing the lower-tropospheric IAGOS measurements at Frankfurt with
nearby (within 50-80 km) and more distant (within 500 km) surface stations, Petetin et al. (2018) showed that the IAGOS
observations in the first few hundred meters above the surface at Frankfurt airport have a representativeness typical of
suburban background stations (like e.g. Uccle and De Bilt), and as one moves higher in altitude, the IAGOS observations
1400 shift towards a regional representativeness. A detailed description of the surface ozone trend at Uccle and its relation with
ozone precursor trends is provided in Appendix B. -

~~In the upper troposphere, the ozone concentration trends deviate more between the different datasets, both in magnitude and
sign, with larger trend uncertainties. At these altitudes, the aircraft could be very distant from Frankfurt (or Munich) airport,
as the ascent/descent profiles stop/start at about 400 to 500 km from the airport. The measurements at these altitudes are
hence representing large areas. Therefore, the closer agreement between the Uccle and De Bilt trends above 8 km compared
to the IAGOS trend might be attributed as well to a similar source region. Also, at those altitudes, the trends do not represent
the tropospheric ozone temporal variability only, as the mean tropopause height range between 10.5 km (winter time) and
11.5 km (summer time), with standard deviations between 1 and 1.5 km, both at Uccle and De Bilt. As a consequence,
lower-stratospheric ozone concentrations will contribute to the estimated trends in the upper altitude levels of Fig.
1405 3. Therefore, the closer agreement between the Uccle and De Bilt trends above 8 km compared to the IAGOS trend might be
ascribed as well to a similar source region.~~

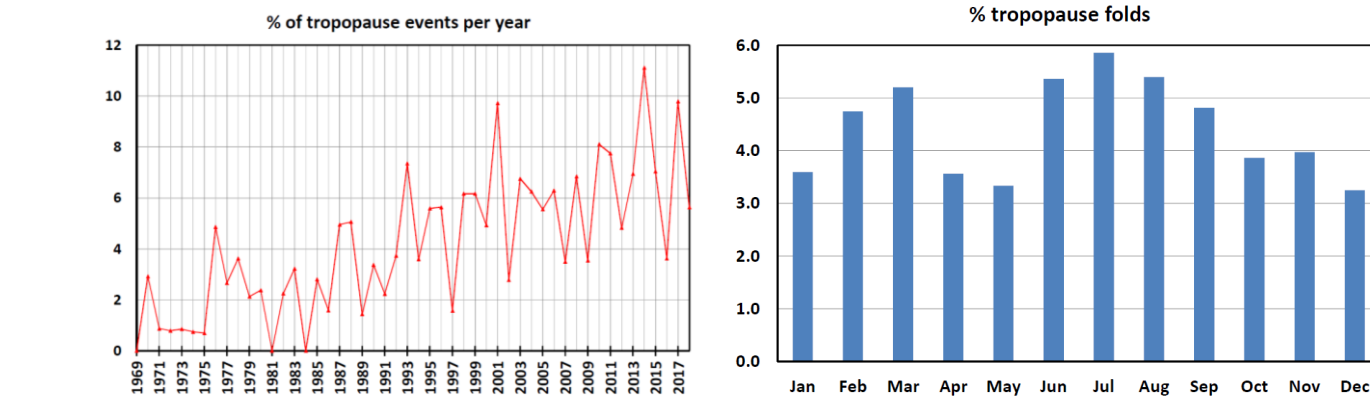
The Uccle tropospheric ozone concentrations have been increasing at about the same rate since 1969 (in black in Fig. 3) ~~as in~~
since 1995 (in green in Fig. 3), and also the most recent considered period here (since 1995, but the post-2000 increase rate
trends is very similar ~~have the same magnitude~~) (not shown here, but is also suggested in the tropospheric ozone column time
1415 series shown in Fig. S7). The increase in (free) tropospheric ozone concentrations above Uccle until the early 2000s is
consistent with the findings reported above (Western) Europe in the literature review of Cooper et al. (2014). Over the 2000-
2014 period, the emissions of the key ozone precursor, nitrogen oxides (NO_x), declined in North America and Europe due to
transportation and energy transformation (Hoesly et al., 2018). Therefore, the overall increase in ozone concentrations has

1420 flattened, but resulted in spatially and seasonally varying tropospheric ozone trends over North America and Europe, without consistency in even the sign of the ozone trends (Gaudel et al., 2018, and references therein). However, Cooper et al. (2020) concluded, based on the IAGOS observations, that the Western Europe free-tropospheric trends since 1995 are predominantly positive. Using a different statistical approach, i.e. a nonlinear regression fit of a quadratic polynomial to normalized, deseasonalized monthly mean ozonesonde (merged data from Uccle, Hohenpeissenberg, and Payerne) and MOZAIC/IAGOS data (Frankfurt) between 3 to 4 km altitude, Parrish et al. (2020) indicated that those ozone concentrations 1425 increased through the 1990s, reached a maximum in the years 2001 (merged ozonesonde) and 2007 (IAGOS) and have since then decreased.

To explain the tropospheric ozone concentration trends, Griffiths et al. (2020) used a chemistry-climate model employing a stratosphere-troposphere chemistry scheme, and found that for the period 1994-2010, despite a levelling off in emissions, increased stratosphere-to-troposphere transport of recovering stratospheric ozone drives a small increase in the tropospheric 1430 ozone burden. Taking advantage of the high vertical resolution of the ozone profiles and the high frequency of launches at Uccle, we focus on the time variability of specific cases of deep intrusions of stratospheric air into the troposphere, i.e. tropopause folds. Akritidis et al. (2019) stressed the role of tropopause folding in stratosphere-to-troposphere transport (STT) processes under a changing climate, suggesting that tropopause folds will be associated with both the degree of and interannual variability in ozone STT. These Tropopause folds occur because of the ageostrophic circulation at the jet 1435 entrance and coincide with the frontal zone beneath the jet. The automatic algorithm applied in this work detects tropopause folds in the Uccle ozone sounding profiles as ozone rich (two criteria), stable (one criterion) and dry (one criterion) air mass layers located in an upper level front in the vicinity of an upper tropospheric jet stream (two criteria), and is described in Van Haver et al. (1996). This identification by means of those six criteria is also illustrated in by means of an example of an ozone sounding containing a tropopause fold in Fig. S86.

1440 Tropopause folds are rather rare events at Uccle: out of the 6526 soundings analysed for the 50 year period (1969-2018), only 290 soundings (or 4.4%) showed evidence of a tropopause folding. However, similar occurrence rates (between 3 and 10%) have been found over Europe at French ozonesonde sites (Beekmann et al., 1997) and with other techniques (Rao et al., 2008, and Antonescu et al., 2013). On a monthly scale, most folding events occur in March, June, July and August (occurrence > 5%), whereas in January, April, May and December, the amount is lower (Fig. 46). What is most important 1445 here within the context of the tropospheric ozone trends is the dramatic increase of the amount of tropopause folding events over time with $0.14 \pm 0.02\% \text{ yr}^{-1} \text{ per year}$ (see also Fig. 46). Van Haver et al. (1996) detected a smaller and insignificant trend of $0.07 \pm 0.11\% \text{ per year}^{-1}$ at Uccle for the 1969-1994 period. On one hand, the large increase over the entire time period might be explained due to some technical aspects. First, the higher vertical resolution of the sounding data in the more recent digital era (since 1990) might have an impact on the larger detected number of tropopause folds (thinner layers might 1450 be detected), although the amount of events has continuously increased since then, at a slightly smaller annual rate of $0.12 \pm 0.05\%$. Secondly, a visual inspection of all profiles fulfilling at least five of the tropopause fold detection criteria, led to a higher number of (manually) identified events (around 50 more), and (relatively) especially in the beginning of the time

1455 series. This is explained by the fact that the low humidity criterion was often not met in the automatic detection, because
 1460 there were no humidity data or the humidity sensor was iced (following the icing recognition algorithm of Leiterer et al.,
 2005). More recent types of radiosonde humidity sensors (in use since 2007 at Uccle) prevent ice contamination by heating
 them during flight. However, this manual (and hence more subjective) mode of the algorithm still gives a $0.09 \pm 0.02\%$
 annual increase of the tropopause fold events since 1969. Therefore, we believe that the significant increase, although
 possibly overestimated by the automatic procedure, is nevertheless a robust feature of the analysis. On the other
 hand, Additionally, a higher rate of tropopause folding events is expected due to climate change (Tarasick et al., 2019, and
 1460 references therein): climate change is projected to increase planetary wave activity and so cause inducing an
 accelerated Brewer-Dobson circulation. This projected acceleration, along with stratospheric ozone recovery, will lead to
 increased transport of ozone from the stratosphere into the troposphere and hence more an expected higher rate of
 tropopause folding events. (Tarasick et al., 2019, and references therein). Akritidis et al. (2019) elaborated that the degree of increase in
 the downward transport of stratospheric ozone is partially driven by the long-term changes in tropopause fold activity.



1465 **Figure 46:** Left: Relative frequency of detected tropopause folding events per year in the ozone soundings at Uccle. Right: Relative frequency of tropopause folding events per month.

1470 To conclude, we found very consistent positive vertical ~~linear~~-tropospheric ozone trends between Uccle, De Bilt, and Frankfurt (IAGOS) since 1995 (~~and even since 2000~~), which are consistent with other studies, both observational and from a modelling approach, but different processing and statistical methodologies can result in different European trend patterns for the last two decades.

4.4 Surface ozone trends

1475 In this section, we elaborate more on the trends in the time series of surface ozone and several ozone precursors measured in Uccle. As a matter of fact, the ozonesonde launch site at the urban background site Uccle also hosts surface measurements of ozone, NO, and NO₂, performed by the Brussels Environment Agency. CO measurements are available from a nearby urban

1480 traffic location at Elsene (< 5 km). From the surface measurements, we consider the (half-hourly averaged) values at 11h30
UT closest to the ozonesonde launch time. The monthly mean time series of those surface measurements are shown in Fig. 7
(monthly anomalies in Fig. S7), together with the lowest 1 km mean ozone measurements derived from the ozonesondes.
1485 The agreement between the surface ozone measurements from both devices is, in terms of monthly means, excellent, apart
from a more or less constant offset. Both time series reveal a statistically significant (according to Spearman's test, see e.g.
Lanzante, 1996) increase in surface ozone concentrations since 1986 (the onset of the surface ozone measurements at the
Uccle site), with a trend value 25% higher for the surface ozone measurements compared to the sonde lowest 1 km
measurements (0.47 vs. $0.38 \mu\text{g m}^{-3} \text{yr}^{-1}$ in absolute terms). Uccle is a suburban site, so, its increase in mean surface ozone
1490 concentrations is in line with the findings from Yan et al. (2018) over European suburban and urban stations during 1995–
2012[†], with trends between 0.20 – $0.59 \mu\text{g m}^{-3} \text{yr}^{-1}$. For the 1995–2018 time period, the ozonesonde trend ($0.41 \mu\text{g m}^{-3} \text{yr}^{-1}$, see
also green curve in Fig. 5 for relative trend) is more elevated than the surface ozone trend ($0.28 \mu\text{g m}^{-3} \text{yr}^{-1}$ or 6.4 ± 2.9
%/decade), and both are statistically significant. This former ozonesonde trend estimate equals the value for the entire
ozonesonde time series 1969–2018 ($0.39 \pm 0.07 \mu\text{g m}^{-3} \text{yr}^{-1}$), as was the case for the entire tropospheric ozone trends (see
again Fig. 5).

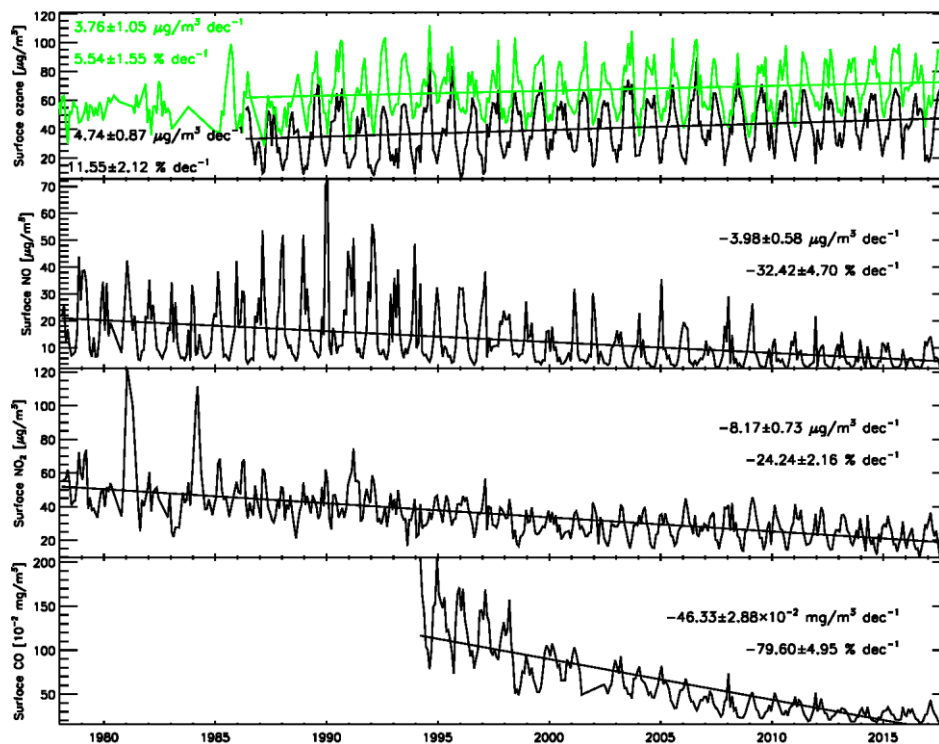
We now compare the surface ozone trends with the trends of the ozone precursor concentrations measured at or near the
Uccle site (NO , NO_2 , and CO , see Fig. 7, and monthly anomalies in Fig. S7). Apparently, there seems to be a mismatch
between the increase in ozone concentrations and the strong decreases of all available measured ozone precursor
concentrations, also noted in other studies (e.g. Tørseth et al., 2012; Lefohn et al., 2018). However, different aspects should
1495 be taken into consideration here. First, the photochemical production of tropospheric ozone also involves reactions implying
volatile organic compounds (VOCs) and hydroxyl radical oxidation of methane and non-methane hydrocarbons, in the
presence of nitrogen oxides (Monks et al., 2015). In NO_x -limited conditions (i.e. rural locations or downwind of urban
plumes and major point sources and at times of high photochemical activity on hot sunny summer days), increases in NO_x
emissions lead to ozone increases while increases in VOC emissions may have limited impacts. In VOC or radical limited
1500 conditions (in areas with large NO_x emissions such as urban core areas and power plant plumes, and under conditions of
lower photochemical activity like night time hours, cloudy days, wintertime days), increases in NO_x emissions may lead to
localized ozone decreases, while increases in VOC emissions result in ozone increases (Lefohn et al., 2018 and references
therein). Unfortunately, VOC measurements are not available at the Uccle surface site. Secondly, although tropospheric
ozone is mainly produced from the photolysis of NO_2 , NO destroys ozone especially during night time, implying that
1505 reductions in NO_x emissions might adversely result in more ozone, especially in highly polluted areas such as cities (Yan et
al., 2018). Chang et al. (2017) also noted that in Europe, the NO_2 column amount tends to be negatively correlated with
ozone in urban sites. Moreover, they mention that in the warm season NO_x emissions tend to produce ozone, while in the
cold season fresh emissions tend to destroy ozone in urban areas, which is also observed on a European scale (Tørseth et al.,

[†]For comparison, over the same period, the Uccle surface ozone trend is $0.37 \pm 0.20 \mu\text{g m}^{-3} \text{yr}^{-1}$, but only $0.07 \pm 0.23 \mu\text{g m}^{-3} \text{yr}^{-1}$ for the ozonesonde measurements.

1510 2012). For Uccle, however, we do not find substantial differences between summertime and wintertime ozone trends in both
 datasets. Furthermore, the ozone trends also depend heavily on the chosen ozone metric (Lefohn et al., 2018). Here, we used
 the monthly means of the 11h30 UTC values, because the ozonesondes are launched around this time, with a very limited
 frequency for surface ozone measurements (hence the need for monthly means). However, the large NO_x emission
 reductions that have occurred in the past several decades in the European Union (EU) have led to a compression of the ozone
 1515 distribution, where the high levels shift downward (reduced ozone peak concentrations) and the low levels shift upward
 (increase in the ozone baseline level), as noted and explained by e.g. Tørseth et al. (2012), Lefohn et al. (2018). These trends
 are actually observed for sites in Brussels (Paoletti et al., 2014) and for the Uccle site (see Fig. S8), although there seems to
 be a levelling off in those opposite trends for low and high ozone concentrations since 2000 compared to the decade before.
 Finally, the trends in 11h30 UT surface ozone measurements can be impacted by changes in meteorology and weather
 regimes, or long range transport patterns due to e.g. climate change.

1520 To conclude, explaining the increasing mean surface ozone amounts in combination with the decreasing ozone precursor
 emissions at Uccle is less straightforward than the (opposing) trends in high and low level ozone concentrations. This is due
 to the interplay of many factors such as meteorology, the non-linear dependence of the ozone concentrations on the
 emissions of VOC and NO_x , the dual role of NO_x as ozone source or sink depending on the season, and the amount of NO_x
 emissions.

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1530 ~~Figure 7: Monthly mean time series of Uccle surface ozone (upper panel, black) and mean ozone in the lowest 1 km above Uccle from the ozonesonde launches (upper panel, green) and ozone precursor measurements at Uccle (NO, NO₂) and Elsene (CO, 5 km from Uccle). Linear trends are shown, together with the absolute and relative trend estimates (calculated from monthly anomalies), and their 2σ uncertainties. The monthly anomaly time series of these measurements can be found in Fig. S7.~~

5 Validation of satellite ozone retrievals with Uccle ozonesonde data

1535 Ozonesondes are virtually all-weather, i.e., unaffected by clouds and precipitation, in contrast to most spectroscopic techniques, and they provide high vertical resolution ozone profiles from the ground to about 30 km. Therefore, satellite algorithms are based on ozonesonde climatologies and in turn satellites are validated by the sondes. Since the start of the ozone measuring satellite era, ozone profiles from soundings at Uccle have been used for validation of satellite ozone retrievals, e.g. the Stratospheric Aerosol and Gas Experiment (SAGE) II satellite profiles (Attmannspacher et al., 1989, De Muer et al., 1990). In this section, we give some recent examples of the application of the Uccle ozone profile data for operational satellite validation (Sect. 5.1), and for the scientific evaluation of both stratospheric (Sect. 5.2) and tropospheric (Sect. 5.3) ozone profile retrievals by satellite instruments. In these latter two sections, we also illustrate that a consistent and
1540 homogenous ozonesonde dataset like the Uccle one is crucial to determine the long-term stability of (merged) satellite ozone retrievals.

5.1 Operational validation within EUMETSAT AC-SAF

As partner of the EUMETSAT Atmospheric Composition Satellite Application Facilities (AC SAF), RMI is responsible for the validation of different ozone products (ozone profiles and (tropical) tropospheric ozone columns) ~~and aerosol products,~~
1545 ~~see~~ (Hassinen et al., 2016, Valks et al., 2014, van Peet et al., 2014) from Global Ozone Monitoring Experiment GOME-2 and Infrared Atmospheric Sounder Interferometer (IASI) instruments on board the MetOp A/B/C satellite platforms. Those different instruments give ~~us~~ the opportunity to obtain a unique dataset, retrieved with an identical technique, from the beginning of the MetOp-A/GOME-2 instrument in 2007, until the end of the third, MetOp-C/GOME-2, foreseen in 2022. GOME-2 ozone profiles are given as partial ozone columns, expressed in Dobson Units, on 40 varying pressure levels
1550 between the surface level and 0.001 hPa and are calculated by the Ozone Profile Retrieval Algorithm (OPERA, van Peet et al., 2014). The a-priori information used for the retrieval is obtained from McPeters and Labow (2012).

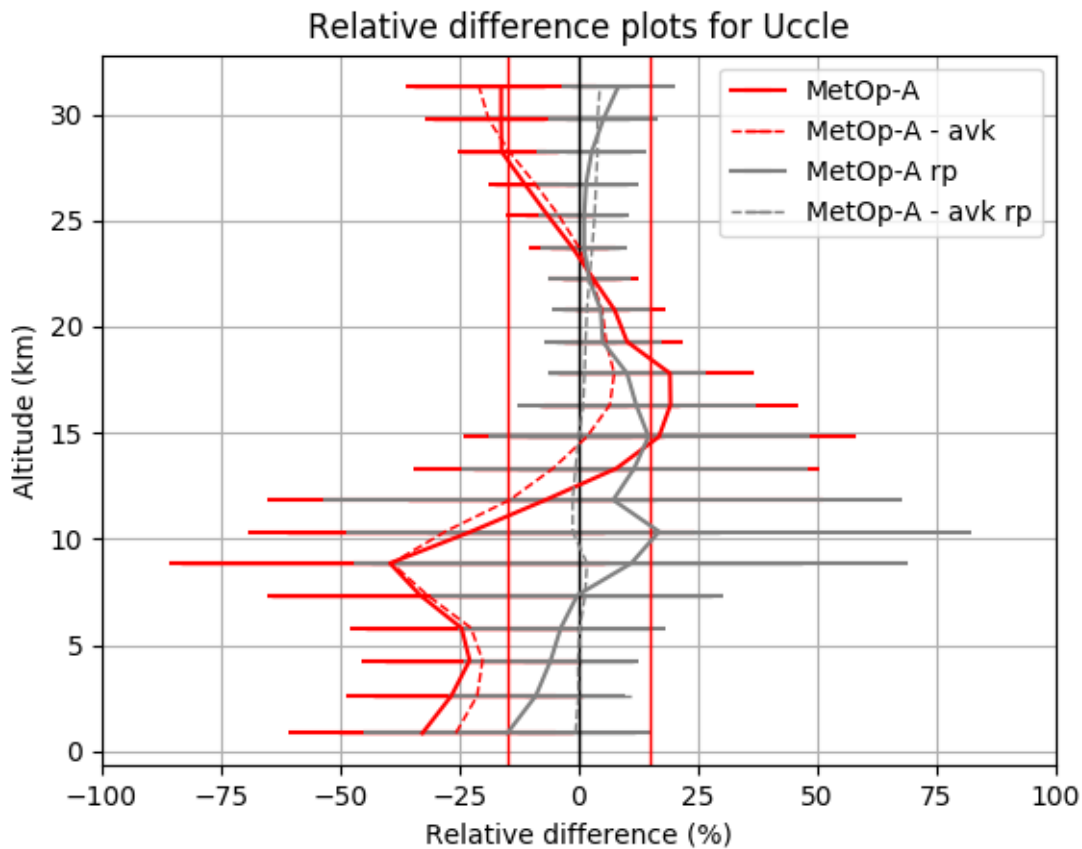
For the validation of GOME-2 ozone profiles within the AC-SAF, ozonesonde measurements are extensively used. However, for a meaningful comparison, the ozonesonde profiles need to be integrated first between the GOME-2 pressure levels. When comparing a single ozonesonde profile with different GOME-2 profiles, the actual reference ozone values are
1555 not identical due to the fact that the GOME-2 vertical levels vary from one measurement to another. GOME-2 has a nominal spatial resolution of 80 km x 40 km, but for the shortest UV wavelengths the integration time takes eight times longer because of the lower number of photons arriving on the detector pixels. Secondly, as the ozonesondes and the satellite do not have the same vertical resolution, it is necessary to take into account the averaging kernels (AVK), to “smooth” the ozone soundings towards the resolution of the satellite (Rodgers, 2000).

1560 | In Fig. ~~ure~~ 58 the relative differences between the MetOp-A operational ozone profile product and the Uccle ozonesonde profiles are shown for the year 2018 (red colour). The following co-location criteria were applied: a geographic distance of less than 100 km between the GOME-2 pixel centre and the sounding station location, and a time difference of less than 10 hours between the pixel sensing and the sounding launch time. The figure highlights two different aspects of the operational validation. First, it can be noted that applying the averaging kernels to the sounding profiles improves the comparison with the GOME-2 ozone product significantly, i.e. by 15%, in particular in the lower stratosphere (compare the full lines with dashed lines in Fig. 58). The lower stratosphere is the region with the highest ozone variability, so smoothing the high resolution ozonesonde profiles to the GOME-2 vertical resolution will have the largest effect here by removing details of the differences. Moreover Secondly, as the GOME-2 ozone profile product is based on UV measurements, it is sensitive to degradation of the UV sensor (van Peet et al., 2014, Munro et al., 2016). For example, the measured values of the GOME-2A irradiance in the UV (below 300nm) has reduced by roughly 80% in 2016 (since its launch in 2007). Since the vertical ozone profile retrieval algorithm depends on an absolute calibrated reflectance (sun normalised radiance) there is a need to correct for this temporal change of the (joint) radiance and irradiance. This method depends on the assumption that, taken as an average across the globe, the atmospheric constituents (mainly ozone) will be close to the multi year climatological value from McPeters and Labow (2012). The climatological ozone profile is then scaled with the Assimilated Total Ozone columns to get the overall ozone absorption correct (Tuinder et al., 2019). ~~This~~ therefore, a degradation correction has been developed for the MetOp-A data, already launched in 2007, and applied to the data for the relative differences with the Uccle data in Fig. 58 (in grey). From this figure, it should be clear that this degradation correction improves significantly the agreement with the Uccle ozonesonde data compared to the operational product (in red), resulting in relative differences between GOME-2 ozone profiles and the Uccle data within the target error range of 15% (marked by the vertical red lines).

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1580 | The improvement after degradation correction is a promising result, showing the challenge for UV-VIS sounders to obtain a stable ozone profile product on different sensors (GOME-2A/2B/2C) for different periods using the same type of optical instrument. More feedback on the status of ~~this the~~ operational EUMETSAT product can be obtained in the validation reports, available on the AC SAF website (<https://acsaf.org>, e.g. Delcloo and Kreher, 2013).



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Figure 58: Relative mean differences and standard deviations between ozone profiles retrieved from MetOp-A/GOME-2 and Uccle ozone profiles for the time period January to December 2018. The red graph represents the mean differences when using the operational MetOp-A/GOME-2 product, the grey graph when the UV sensor degradation correction has been applied in the MetOp-A/GOME-2 ozone retrieval. Relative mean differences denoted by dashed lines are obtained after applying the averaging kernel to the Uccle sounding data. Finally, the thin red vertical lines mark the $\pm 15\%$ target error range of the MetOp-A/GOME-2 ozone profile product.

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5.2 Validation of AURA-MLS stratospheric ozone profiles

The Microwave Limb Sounder (MLS, Froidevaux et al., 2008) is one of the four instruments on the Earth Observing System (EOS) Aura Satellite. MLS has been measuring vertical profiles of atmospheric trace gases, including ozone, along with temperature, geopotential height, relative humidity, cloud ice water content and cloud ice water path, since its launch in 2004. Global measurements (from 82° S to 82° N), in a near-polar, sun-synchronous orbit, at two fixed solar times, noon-night, at around 01:30 a.m./p.m. are achieved, with the number of profiles over e.g. ozonesonde sites varying between 0 and 6 daily. MLS products have been validated to be very accurate and stable (Jiang et al., 2007, Froidevaux et al., 2008), and have been used in many studies involving ozonesonde measurements (e.g. Witte et al., 2017, Stauffer et al., 2020). Here, we

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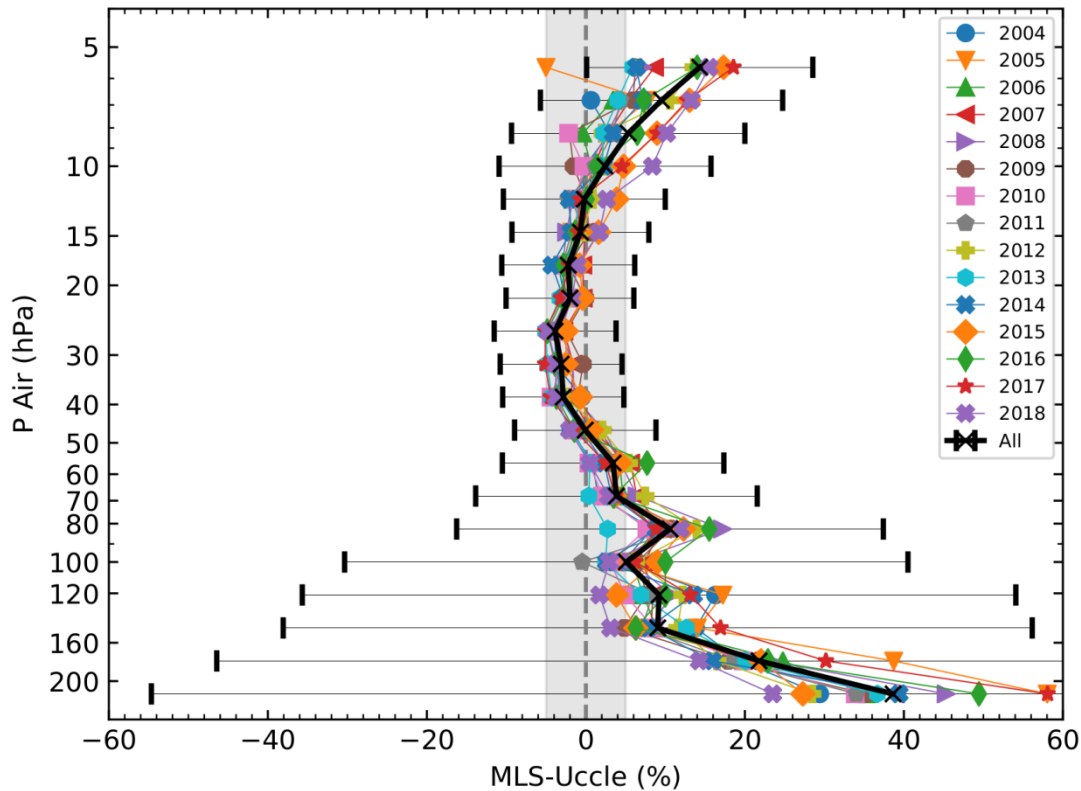
1600 have implemented the ~~latest~~ MLS v4.2 data, screened according to the v4.2 Level 2 MLS Data Quality document (Livesey et al., 2020), and compared the satellite overpass measurements, ~~without averaging kernel~~, with coincident ozonesonde profiles at Uccle. Because there are multiple profiles crossing over Uccle at a fixed time, the profile closest in distance is used for the validation. Both the noon and night overpasses have been used, as we did not find significant differences between those. As a result, ~3000 profiles were included into the validation. ~~To account for the difference in resolution, Uccle ozonesonde data are linearly interpolated to the MLS vertical resolution (between 3–6 km, depending on the altitude). Thanks to the relatively dense and regular MLS vertical resolution of around 2.5 km in the 10–200 hPa pressure range, it is feasible to interpolate the Uccle ozonesonde data to the MLS pressure levels on a fine pressure grid of 2.5 km. Applying the time invariant MLS averaging kernel on the latitude of Uccle on the ozonesonde data did not have a large effect on the smoothing of the vertical ozonesonde profile, as compared to applying the identify matrix to the ozonesonde vertical profile (< 1%). This contrasts strongly with the GOME-2 and TES retrievals (see Sect. 5.3), where the spatio-temporal varying averaging kernels affect the vertical ozone profiles substantially, and as such should be used on the sonde data for pairwise comparison.~~ The mean annual relative differences between MLS and Uccle ozonesondes are shown in Fig. 69. Different conclusions can be drawn from this figure. First, MLS and the Uccle ozonesondes compare very well, within $\pm 5\%$ between 10 and 70 hPa (grey shading in Fig. 69). At pressures smaller than 10 hPa, ozonesonde measurements are ~~known to be less accurate systematically underestimating ozone due to the evaporation or freezing of the sensing solutions~~ (see also ~~Sect. 2.2~~ the composite ECC-MLS Fig. 3 in Stauffer et al., 2020), and they have a larger uncertainty due to increased pump efficiency uncertainty at low pressures.; ~~On the other hand, while~~ at pressures larger than 70 hPa, the MLS ozone retrieval is more challenging because of the longer atmospheric path and the lower ozone volume mixing ratios increasing the relative differences. Another important finding from this figure is that the mean annual relative differences are very consistent over the different years, which means that both the MLS instrument and the Uccle ozonesonde time series are very stable with respect to each other. In addition to this ~~(see Fig. S9)~~, we also want to mention that the relative differences between MLS and Uccle ozonesondes are very similar for the different seasons, see Fig. S9.

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1625 **Figure 69:** Relative ozone profile differences between MLS and Uccle ozonesondes. The different colours correspond to the different yearly averages, illustrating the large consistency among those. ~~The~~ the black line represents the overall mean relative difference, with the error bars the one standard deviations due to the individual differences. Note that individual differences are relatively large at some pressure levels, but they are cancelled out in the yearly mean. The error bar is only shown for the overall and corresponds to one standard deviation.

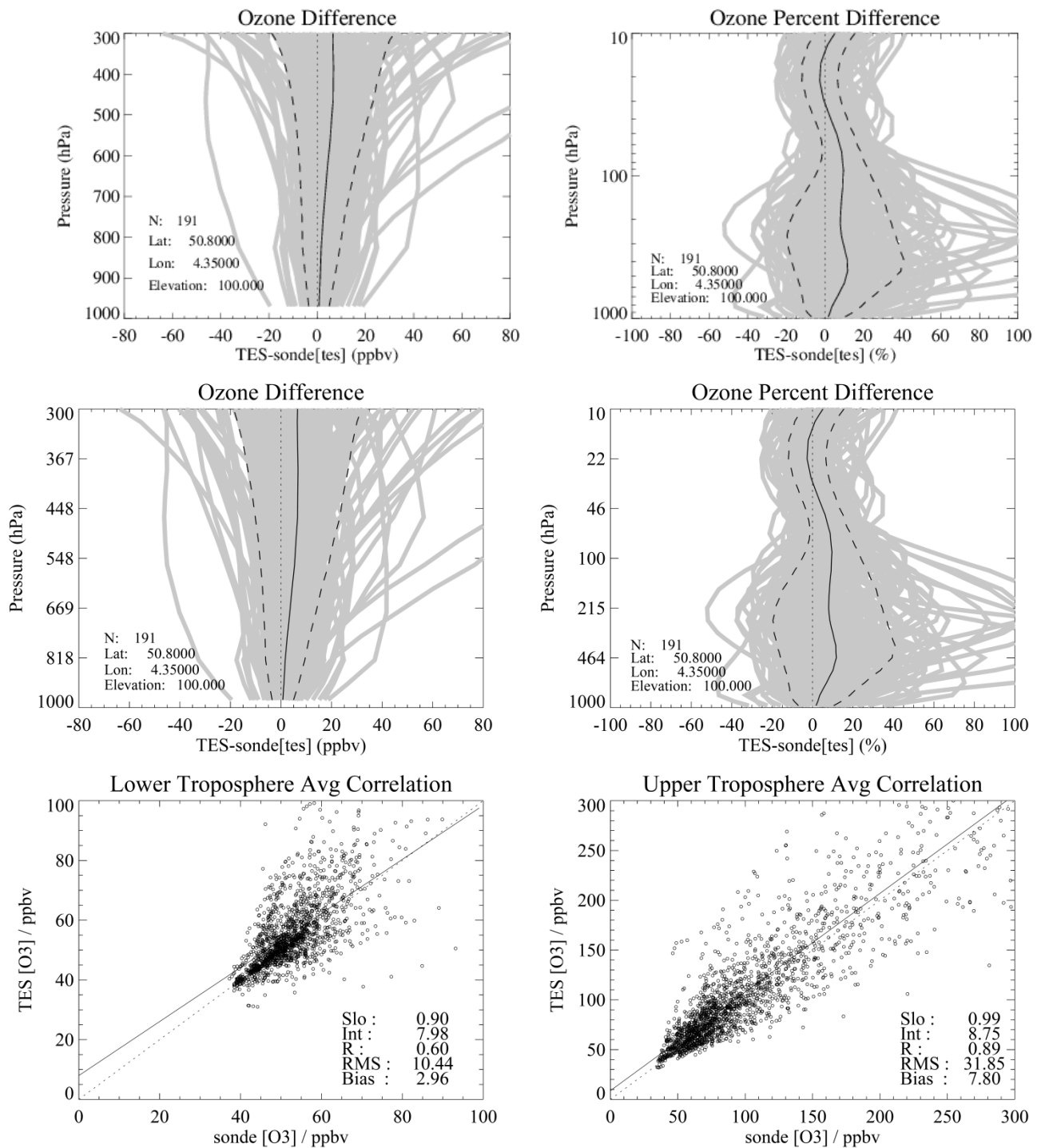
5.3 Validation of AURA-TES tropospheric ozone profiles

1630 Here we compare the tropospheric vertical ozone profiles of the Uccle sondes coinciding with the observations from the Tropospheric Emission Spectrometer (TES) sensor on-board the Aura satellite for the period late 2004 to early 2018, when the instrument was decommissioned. TES is an infrared Fourier transform spectrometer (Beer et al., 2001; Beer, 2006) ~~following a near polar, sun-synchronous orbit with equator crossing times of 13:40 local mean solar time for the ascending part of the orbit. TES is predominantly nadir viewing and that~~ measures radiance spectra of Earth's atmosphere, predominantly nadir viewing, at wavelengths between 3.3 and 15.4 μm . The nadir vertical profiles are spaced 1.6° apart
 1635 along the orbit track and have a footprint of approximately $5 \times 8 \text{ km}^2$ (Beer et al., 2001; Beer, 2006).

The vertical sensitivity of the TES-retrieved ozone is the largest for the troposphere, with a vertical resolution for ozone profiles of 6-7 km, corresponding to 1-2 degrees of freedom in the troposphere (Jourdain et al., 2007). Prior to applying TES ozone data, they are subject to screening, using the TES ozone master quality flag that accounts for clouds and a too large difference between observed and simulated radiances (Osterman et al., 2008).

As in Nasser et al. (2008) and Verstraeten et al. (2013), we apply temporal and spatial coincidence criteria of ± 9 h and ± 300 km respectively between the sonde and TES observations. These criteria can provide enough profiles for a statistically meaningful comparison while it is sufficiently strict to warrant a high probability that both instruments sample similar air masses. A mapping matrix is used to interpolate the sonde data to the 67-level pressure grid (from 1212 to 0.1 hPa) used in the TES retrievals. Then, the TES ~~observation operator~~ averaging kernel was applied to the 67-level pressure grid of the Uccle sonde data to ensure a consistent comparison between TES and ozonesonde data excluding the influence of the a priori ozone profile needed to regulate the TES retrieval (Verstraeten et al., 2013).

By applying all these constraints (coinciding criteria and the TES ozone master flag), 191 suitable coincidences or data pairs for the full time range from 2004 to 2018 were collected. The upper panels in Fig. 7.40 presents TES-sonde tropospheric ozone profile differences for the Uccle sondes. The left upper panel shows the absolute ozone vertical profile differences (TES-sonde) in the troposphere (1000-300 hPa). The right upper panel shows the relative differences ((TES-sonde) \times 100/sonde) for the full vertical ozone profile (1000- 1 hPa).



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Figure 740: Absolute TES–sonde tropospheric ozone vertical differences (left **upper panel**) and relative differences (right **upper panel**) for the whole profile of Uccle. Individual difference profiles are shown in grey; the mean difference and one standard deviation profiles are in black. N is the number of valid profiles after flagging TES data and using the maximum 300 km and 9h

1660 coinciding criteria. Lower left panel illustrates the correlation between TES and sonde O₃ for the lower troposphere (1000 to 500 hPa) including the slope (slo), intercept (Int) and correlation (R) of the linear regression, the root mean square error (RMS) and the bias. Similar for the lower right panel for the upper troposphere (500 hPa to tropopause).

1665 Figure 7+9 indicates that TES is generally positively biased within the troposphere by up to ~10 ppbv, corresponding to relative differences up to ~15 %. The TES bias slightly varies as a function of pressure. TES appears to be almost unbiased with respect to the sondes in the lower troposphere, but this actually reflects the non-sensitivity of TES to ozone in the lower atmosphere for situations with lower brightness temperature as encountered at higher latitudes. Since the TES signal in the troposphere has typically 1–2 degrees of freedom, analysing the TES bias for two vertical regimes - the lower troposphere (LT, 1000 to 500 hPa) and the upper troposphere (UT, 500 hPa to tropopause)- might be meaningful (Nassar et al., 2008). From a linear regression of all TES vs. sonde ozone data pairs for Uccle in the lower troposphere (Fig. 7, lower left panel), we find a slope of 0.90 with an intercept of 7.98 (R = 0.60) with a bias of +2.96 ppbv. For the upper troposphere (lower right panel in Fig. 7) the bias is a bit higher (7.80 ppbv), the correlation (R) is 0.89 and the slope and intercept are 0.99 and 8.75 respectively. These values are in line with reported ones for data pairs collected for the whole northern mid-latitudes (Verstraeten et al., 2013).

1670 The temporal stability of the TES sensor for tropospheric ozone can be assessed by applying the Theil Sen trend statistics (Theil 1950a, 1950b, 1950c; Sen, 1968) on the time series of the TES-sonde data pairs for each pressure level in the troposphere (surface to 300 hPa). Analysis shows that all p-values are larger than 0.05 indicating that all slopes of the linear regression are not statistically different from zero in the troposphere. All R² values are smaller than 0.01. Thus, there is no reason to assume any temporal trend for data pairs in the troposphere. This is in line with the same analysis for the 464 hPa level by Verstraeten et al. (2013).

6 Conclusions and outlook

1680 Having started operationally in 1969 to use ozone as a tracer to study the general air circulation in the troposphere and the lower stratosphere, the high-frequency (three times a /week) mid-latitude Uccle ozone sounding time series now extends over more than 50 years, covering ~~around~~ over 7000 profiles. Over this entire period, attention has always been paid to the consistency of the time series, resulting in only one major change: the switch from BM to En-Sci ECC sondes in 1997. This change was however well documented with dual launches and pump efficiency laboratory measurements of both pump types, so that a unique correction method for both sonde types, a PRESSure and Temperature dependent total Ozone normalization (PRESTO, Van Malderen et al., 2016), has been developed (De Backer et al., 1998a,b) to guarantee the data homogeneity. Another distinct feature of the Uccle ozonesonde dataset is the correction for urban SO₂ interference with the chemical reactions in the ozone cells in the first half of the period.

1690 Although satellites provide global routine measurements of ozone profiles with increasing accuracy and spatial resolution, ozonesondes are the only technique that can provide, since 50 years, accurate (around 5-10%), vertically resolved observations from the surface up to the lower stratosphere, unaffected by clouds or precipitation. Furthermore, they can

1695 resolve strong gradients in the UTLS (upper troposphere/lower stratosphere), while precisely locating the thermal tropopause (Thompson et al., 2011). In this paper, we illustrated the importance of the Uccle ozonesonde dataset in two specific application areas: for the assessment of the long-term vertical ozone trends and for the validation of satellite retrievals of ozone profiles. The strength of the ozonesonde measurements (and the Uccle time series in particular) lies exactly in combining those two aspects of ozone research, together with its applicability in process studies. The major conclusions are summarized here.

1700 Making use of the LOTUS multiple linear regression model including the QBO, the solar radio flux, ENSO, and AOD as explanatory variables, we found that the stratospheric ozone concentrations at Uccle declined at a significant rate of around 2% dec^{-1} since 1969. This overall decline can mainly be attributed to the increasing ODS emissions, with a rather consistent decline rate around -4% dec^{-1} for the period 1969-1996. Since 2000, a recovery between +1-3% dec^{-1} of the stratospheric ozone levels above Uccle is observed, although not significant and not for the upper stratospheric levels measured by ozonesondes. A significant decline in lower stratospheric ozone amounts since 1998, as reported by Ball et al. (2018, 2019), is hence not present in the Uccle and nearby De Bilt time series. For the considered periods, we found an overall agreement between the sign of the stratospheric temperature trends and those ozone concentration trends, i.e. a cooling of the stratosphere in 1969-2018 and 1969-1996 and an insignificant warming for all but the lower stratospheric layers since 2000, underlining the possible mutual interaction between stratospheric ozone concentration and temperature changes.

1710 ~~In Appendix A, we showed that the total column ozone loss at Uccle between 1971-1996 (at a rate of -2.51.6% dec^{-1}) is almost entirely compensated by the gain has nearly fully recovered by the (+1.92% dec^{-1}) gain since 2000 between 1997-2019.~~ In the light of the discussion on the stratospheric ozone trends in the previous paragraph, this would mean that the tropospheric ozone amounts at Uccle should increase since the mid-90s. We indeed confirmed a very consistent increase of the ozone concentrations at 2 to 3 % dec^{-1} throughout the entire free troposphere, a number which is in almost perfect agreement with the trends derived from the IAGOS ascent/descent profiles at Frankfurt, and 1% dec^{-1} lower than the De Bilt tropospheric ozone trends. The Uccle 1995-2019 trend is even 0.5 to 1% dec^{-1} higher than the 1969-2019 trend. 1715 Despite the levelling off in tropospheric ozone precursor emissions, the tropospheric ozone amounts in Uccle are still increasing. Based on chemistry-climate model calculations, Griffiths et al. (2020) found that an increase in the tropospheric ozone burden might be driven by increased stratosphere-to-troposphere transport of recovering stratospheric ozone. It should also be noted that the amount of tropopause folding events in the Uccle time series increased significantly over time, which might be an indicator for increased transport of ozone from the stratosphere into the troposphere. However, in line with the free-tropospheric ozone, also the surface ozone concentrations at Uccle continue to increase since the beginning of those measurements in the 1980s, despite trends behave similarly: the decreasing on-site concentrations of precursor trace gases CO, NO, and NO₂ have significantly decreased, but the surface ozone concentrations continue to increase since the beginning of those measurements in the 1980s and 1990s (see Appendix B). To explain this, we should keep in mind that Uccle is a

~~(sub)urban site, and in such an environment of elevated NO_x concentrations, the photochemical production of ozone might lead to rising concentrations, even with declining NO_x emissions.~~

For the operational validation of the GOME-2 and IASI ozone profiles within the EUMETSAT AC-SAF, the role of ozonesonde profiles is crucial. We showed how the Uccle dataset can be used to evaluate the performance of a degradation correction for the GOME-2 UV sensors. The Uccle ozonesondes are also used to assess the accuracy and stability of satellite ozone retrievals. Here, we showed that the AURA-MLS overpass ozone profiles agree very well with the ozonesonde profiles, within $\pm 5\%$ between 10 and 70 hPa. Another instrument on the same AURA satellite platform, TES, has its largest vertical sensitivity for ozone in the troposphere, and is generally positively biased with respect to the Uccle ozonesondes in the troposphere by up to ~ 10 ppbv, corresponding to relative differences up to $\sim 15\%$. Using the Uccle ozonesonde data series as reference, we also found that the temporal stability of both satellite retrievals is excellent. Vice versa, satellite total ozone retrievals and MLS have enabled the detection of a post-2013 drop-off in total ozone at a third of global ozonesonde stations (Stauffer et al., 2020), ~~a number now reduced to about 20% (12 of 60 global stations, Stauffer et al., 2021, private communication).~~ Our analysis with MLS here confirmed their finding that Uccle is not affected by any total column drop-off of more than 3% in its time series.

A higher flexibility of ozonesonde launch times toward satellite overpass times is an emerging issue that needs to be considered against the preference for a fixed launch time for e.g. the assessment of tropospheric ozone trends. Moreover, for over a decade, weather prediction centres have been incorporating chemistry into operational forecasts, assimilating satellite ozone retrievals, and ozonesondes are used for external evaluation of those model forecasts (e.g. for tropospheric ozone: Flemming et al., 2015), analyses (e.g. for stratospheric ozone: Lefever et al., 2015) and reanalyses (e.g. Inness et al., 2019). Those services require a near real-time delivery of the ozonesonde measurements, with an operational quality assessment/quality control tool, ~~as the total column ozone drop off in a third of the ozonesonde stations (Stauffer et al., 2020) made obvious.~~ These are the challenges for operational applications of ozonesondes. For the assessment of the long-term variability of ozone concentrations at different atmospheric altitudes and the interaction between climate change and ozone (also studied in coupled chemistry-climate and chemistry-transport models, see e.g. Morgenstern et al., 2017), the availability of a long-term homogeneous dataset is crucial. Homogenization efforts of ozonesonde networks and/or datasets (Tarasick et al., 2016; Van Malderen et al., 2016; Thompson et al., 2017; Witte et al., 2017, 2018, 2019; Sterling et al., 2018) should therefore be continued and extended. With these developments in mind, we aim at continuing the pioneering role that the Uccle time series had in some of the research areas during its half a century lifetime.

Appendix A: The Uccle total ozone trends

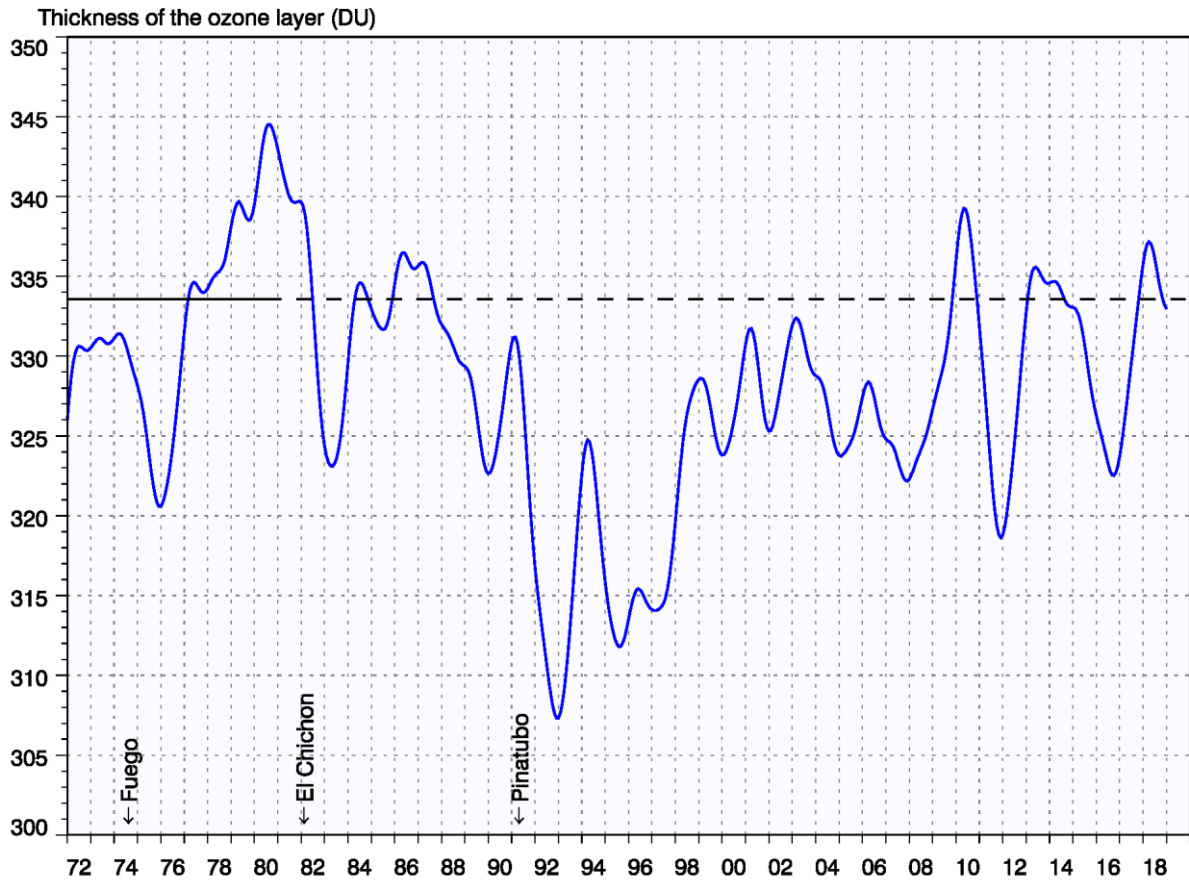
The total column ozone amounts at Uccle, available since 1971, are retrieved with a Dobson UV-spectrophotometer (no. 40, 1971-1989), a single Brewer UV spectrophotometer (no. 16, 1990-current, but used in the time series until the end of 2001).

1760 and a double Brewer UV spectrophotometer (no. 178, 2002-current). The calibration history of the Dobson instrument is documented in De Muer and De Backer (1992) and the transition to the Brewer instrument is described in De Backer and De Muer (1991). Both Brewer instruments were recalibrated against the traveling standard Brewer instrument no. 17 in 1994 (no. 16 only), 2003, 2006, 2008, and against the travelling reference Brewer no. 158 since 2010 every second year. The stability of the instruments is also continuously checked against the co-located instruments (with the Dobson no. 40 from 1991 until May 2009, between both Brewers since 2001). Internal lamp tests are performed on a regular basis to check whether a Brewer instrument is drifting. When instrumental drift is detected, it is corrected for.

1765 The time series of total ozone measurements is shown in Fig. A1, but has been smoothed by applying a low-pass Gaussian filter with a width at half height of 12 months, to filter out variations with frequencies higher than one year. With this representation, the impact of the major (strato)volcanic eruptions of Fuego (Guatemala, Oct 1974), El Chichon (Mexico, Mar/Apr 1982), and Pinatubo (the Philippines, Jun 1991) is shown in the significant dips in Uccle total ozone. Indeed, the episodes of enhanced stratospheric aerosol-related ozone loss after those major volcanic eruptions are confirmed by model results (see e.g. Tie and Brasseur, 1995, Solomon 1999, Aquila et al., 2013 for a description of the mechanism behind) and can clearly be identified in the time series. Also the other inter-annual variability in Fig. A1 is very similar to the Northern

1770 Hemisphere (NH) annual mean total ozone time series of five bias corrected merged datasets in the 35–60° N latitude band in Weber et al. (2018; their Fig. 2). In 2010, the Uccle ozone levels were unusually high, as over the entire NH extratropics. An unusually pronounced and persistent negative phase of the Arctic Oscillation and North Atlantic Oscillation in 2010, with the co-occurrence of northern winter 2009/2010 with the easterly wind-shear phase of the QBO have been identified as major contributors (Steinbrecht et al., 2011) of this excess ozone. The 2011 ozone low anomaly cannot be fully explained by including this Arctic Oscillation and other dynamical proxies (e.g. for the Brewer-Dobson circulation) in the used multiple linear regression model in Weber et al. (2018), but might be linked to the strong Arctic ozone loss in 2011 (Manney et al., 2011). The below-average annual mean Uccle and NH total ozone in 2016 is partly ascribed to the severe Arctic ozone depletion in the same year and related to the anomalous quasi biennial oscillation (QBO) induced meridional circulation changes (see references in Weber et al., 2018).

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Figure A1: Evolution of the total ozone column at Uccle as observed with Dobson D40 (1972-1989), Brewer no. 16 (1990-2001), and Brewer no. 178 (2002-present). The horizontal black full line marks the 1972-1980 total ozone average (period before catalytic ozone loss), extended until the end of the time series by the dashed horizontal line. The periods of major volcanic eruptions affecting the ozone layer are indicated on the time axis as well.

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To study the long-term temporal variability of the total ozone amounts at Uccle, we make use of the LOTUS MLR regression model that we also applied to estimate the stratospheric vertical ozone trends in Sect. 4.1. The model fit and the different contributors are shown in Fig. A2. The interannual variability is reasonably captured by this model, although this MLR is not able to model the large excursions in some years, e.g. 2011-2012, without the use of some additional terms accounting for the Arctic Oscillation or the Brewer-Dobson circulation (Weber et al., 2018). As can be noted from the

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observation-model residuals, the long-term temporal variability is well described by the two independent linear trends. Before 1997, ozone declined at Uccle at a rate of $-1.6 \pm 0.5 \text{ \% dec}^{-1}$ due to the anthropogenic production of ozone depleting substances (ODS), transported into the stratosphere, with peak concentrations in 1997. This decline rate is comparable to the NH mid-latitude value of -2 to -3 \% dec^{-1} (Weber et al., 2018; WMO, 2018), especially considering that the Uccle total ozone time series starts earlier than the used satellite total ozone time series in those assessments (from 1979). Subsequently, from

1795 2000 onwards, the total ozone increased again at a rate of $+1.9\pm 0.8\%$ dec^{-1} at Uccle. This ozone recovery estimate is
significantly larger than the NH mid-latitude trend of $+0.2$ to $+0.5\%$ dec^{-1} (Weber et al., 2018; WMO, 2018) and even larger
than the expected NH trends from Equivalent Effective Stratospheric Chlorine (EESC) changes, which are about $+1\%$ dec^{-1}
(WMO, 2018). At Uccle, the total ozone amount seems to have nearly fully recovered yet, as could also be noted by looking
at the monthly anomaly time series in Fig. A2. Because the Dobson and Brewer spectrometers are calibrated regularly (see
1800 above), we have no doubts on the homogeneity of the time series. In general, according to Weber et al. (2018), the ozone
increase after 2000 is not only due to the (slow) decrease in ODSs in the stratosphere, but also because of atmospheric
dynamics, notably ozone transport via the strengthening Brewer–Dobson circulation. At Uccle, the strongest ozone increase
since the beginning of this century took place in late winter – early spring (Feb–Apr), at a rate of $3\text{--}4\%$ dec^{-1} , while the ozone
transport by the Brewer-Dobson circulation from its tropical source region poleward and downward into the lower
1805 stratosphere is strongest during wintertime (e.g. Butchart, 2014; Langematz, 2019).
While total ozone seems to have nearly fully recovered at Uccle, the stratospheric ozone amounts have not (see Fig. 2 and
Fig. S5 for the monthly anomaly time series of the ozone concentrations in a layer 10 km above the tropopause height). The
stratospheric ozone concentrations decreased between 1969-1996 with a rather consistent rate around -4% dec^{-1} (between 5
to 20 km above the tropopause), hence larger than the total ozone decline rate. Since 2000, a recovery between $+1$ to $+3\%$
1810 dec^{-1} of the stratospheric ozone levels above Uccle is observed, although not significant and not for the upper stratospheric
levels measured by ozonesondes. This value is comparable to the total ozone recovery rate at Uccle. To reconcile the
stratospheric ozone trends from the ozonesondes with the total ozone trends at Uccle², it should also be noted that,
throughout the entire free troposphere (contributing for about 10% to the total ozone amount), a very consistent increase of
the ozone concentrations at $+2$ to $+3\%$ dec^{-1} is measured since both 1969 and 1995 (see Fig. 3).

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² Note that the ozone measurements with the ozonesondes at Uccle are normalized (dependent on the pressure and temperature) to the total ozone measurements from the co-located spectrophotometers.

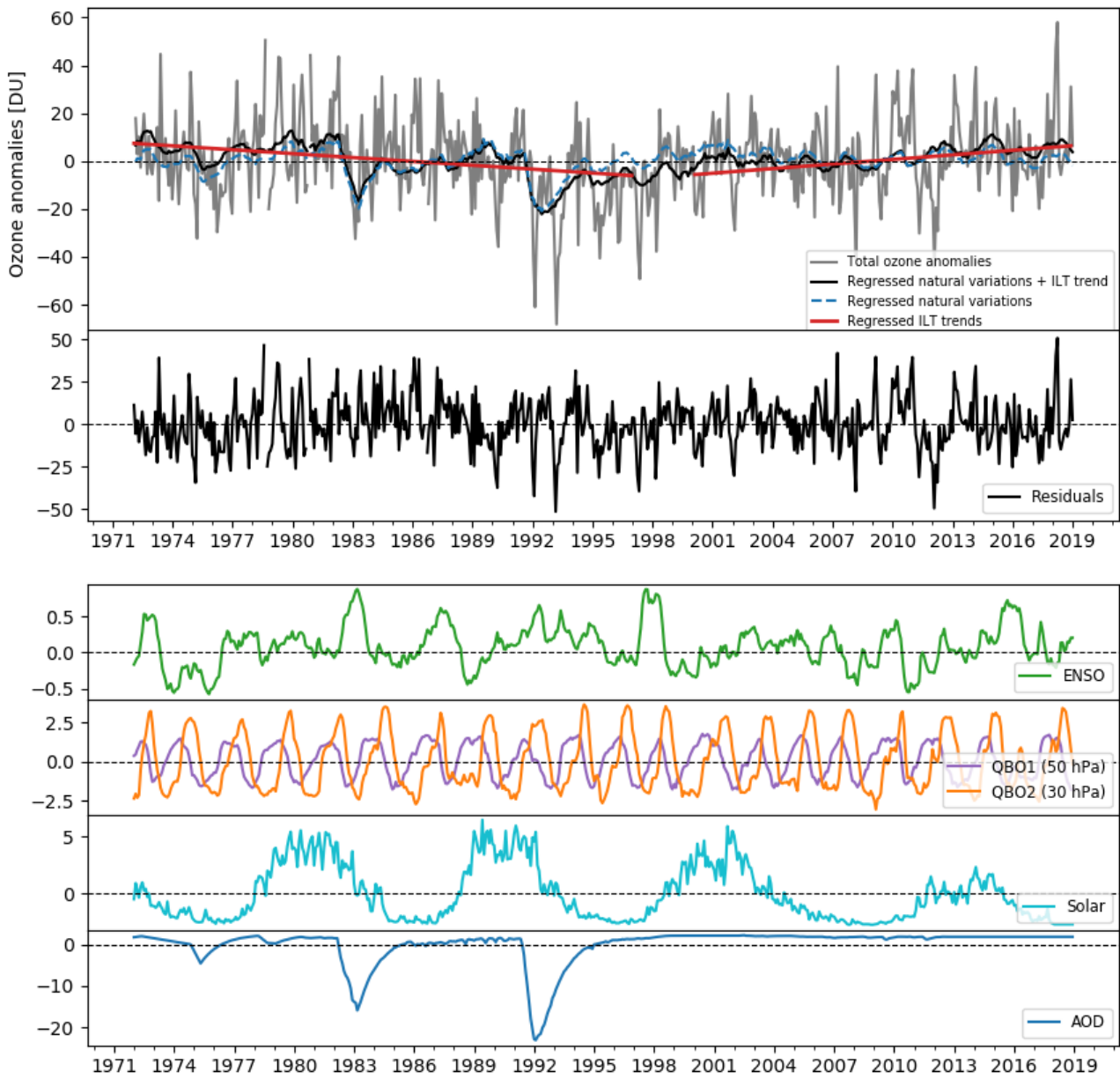


Figure A2: Terms in the LOTUS multiple linear regression model for the Uccle total ozone amounts. The top panel shows the observed total ozone monthly anomalies in grey. The black line is the result of the full LOTUS regression model including the independent linear trends (ILTs, thick red lines). The pre-1997 trend equals $-1.6 \pm 0.5\% \text{ dec}^{-1}$, the post-2000 trend $+1.9 \pm 0.8\% \text{ dec}^{-1}$. The dashed blue line shows the sum of the terms of the LOTUS model without the ILTs included. The middle panel shows the residuals in the observed ozone concentrations with the full LOTUS model subtracted. The bottom panel shows the contributions of (from top to bottom) the ENSO, QBO, solar cycle, and aerosols to the reconstructed time series (blue dashed line) in the top panel.

Appendix B: Surface ozone trends at Uccle

1825 In this appendix, we elaborate more on how representative and complementary the surface ozone trend derived from the
ozonesonde data at Uccle is, compared to the one from a surface monitoring station at the same site. The ground network of
(air quality) stations provides surface ozone measurements at higher temporal and horizontal resolution, and with higher
accuracy than ozonesonde measurements, but these latter provide vertical ozone profiles in the lower troposphere as well,
and sometimes even over a longer time span. As a matter of fact, the ozonesonde launch site at the urban background site
1830 Uccle also hosts surface measurements of ozone since 1986, performed by the Brussels Environment Agency. From the
surface measurements, we consider the (half-hourly averaged) values at 11h30 UT, closest to the ozonesonde launch time.
The monthly mean time series of those surface measurements are shown in Fig. B1, together with the lowest 1 km mean
ozone measurements derived from the ozonesondes. The agreement between the surface ozone measurements from both
devices is, in terms of monthly means, excellent, apart from a more or less constant offset. This offset might be explained by
1835 the difference in air masses for which the ozone concentrations are measured (surface vs. surface to 1 km above the ground),
and by some Uccle pre-launch procedure of testing the ozonesonde-interface-radiosonde configuration by exposing the
ozonesonde shortly (< 30s) to (stratospheric) ozone concentrations between 15 to 30 minute prior to launch. Because of the
slow time constant of 20-25 minutes in the chemical reactions in the cell, this pre-launch ozone exposure might still
contribute to the measured cell current immediately after launch, resulting in a positive bias in the boundary layer ozone
1840 measurements with the ozonesondes.
Both time series reveal a statistically significant (according to Spearman's test, see e.g. Lanzante, 1996) increase in surface
ozone concentrations since 1986 (see Fig. B2), with a trend value 25% higher for the surface ozone measurements compared
to the sonde lowest 1 km measurements (0.47 vs. $0.38 \mu\text{g m}^{-3} \text{yr}^{-1}$ in absolute terms). Uccle is a suburban site, so, its increase
in mean surface ozone concentrations is in line with the findings from Yan et al. (2018) over European suburban and urban
1845 stations during 1995–2012³, with trends between 0.20 – $0.59 \mu\text{g m}^{-3} \text{yr}^{-1}$. For the 1995-2018 time period, the ozonesonde trend
($0.41 \mu\text{g m}^{-3} \text{yr}^{-1}$, see also green curve in Fig. 3 for relative trend) is more elevated than the surface ozone trend ($0.28 \mu\text{g m}^{-3}$
 yr^{-1} or $6.4 \pm 2.9 \%$ dec^{-1}), and both are statistically significant. This former ozonesonde trend estimate equals the value for the
entire ozonesonde time series 1969-2018 ($0.39 \pm 0.07 \mu\text{g m}^{-3} \text{yr}^{-1}$), as was the case for the entire tropospheric ozone trends
(see again Fig. 3).

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³ For comparison, over the same period, the Uccle surface ozone trend is $0.37 \pm 0.20 \mu\text{g m}^{-3} \text{yr}^{-1}$, but only $0.07 \pm 0.23 \mu\text{g m}^{-3} \text{yr}^{-1}$ for the ozonesonde measurements.

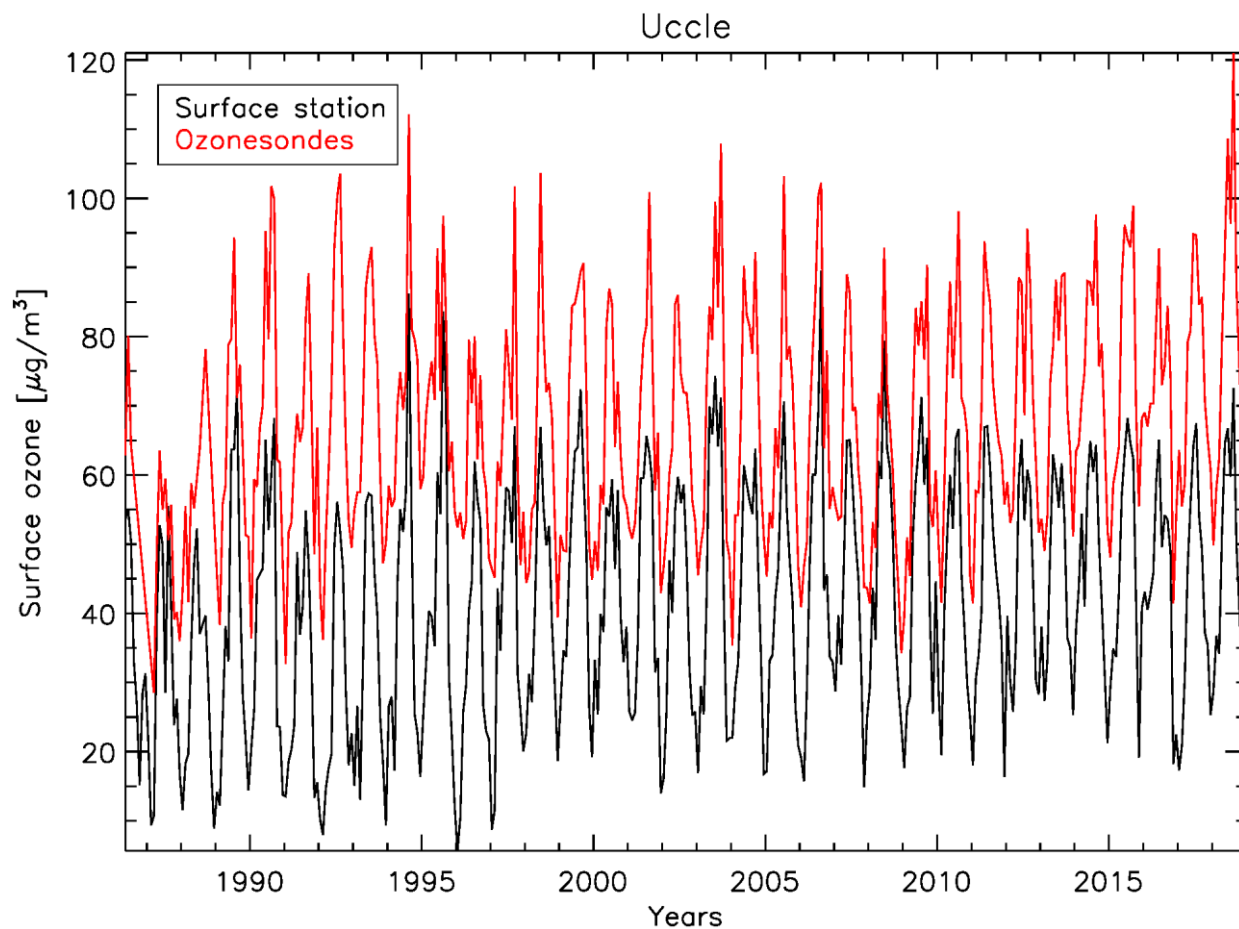


Figure B1: Monthly mean time series of Uccle surface ozone (black) from the air quality monitoring station and mean ozone in the lowest 1 km above Uccle from the ozonesonde launches (red).

In general, trends in surface ozone measurements are ascribed to changes in local and regional anthropogenic precursor emissions, in natural ozone precursors and/or their sources, in meteorology and weather regimes, or long-range transport patterns due to e.g. climate change (Monks et al., 2015, Lefohn et al., 2018). As ozone precursor concentrations are measured at (NO and NO₂) or near the Uccle site (CO measurements are available from a nearby urban traffic location at Elsene, < 5 km), we concentrate here on a possible link between the changes of those precursor mean concentrations (see Fig. B2) to the positive surface ozone trend. Apparently, there seems to be a mismatch between the increase in ozone concentrations and the strong decreases of all available measured ozone precursor concentrations, also reported in other studies (e.g. Tørseth et al., 2012; Lefohn et al., 2018). However, it should be noted first that the photochemical production of tropospheric ozone also involves reactions implying volatile organic compounds (VOCs) and hydroxyl radical oxidation of methane and non-methane hydrocarbons, in the presence of nitrogen oxides (Monks et al., 2015). Unfortunately, those components are not measured at the Uccle site. Moreover, the observed NO_x decreases in Fig. B2 at the Uccle site can have a

1865 reverse impact on the surface ozone trends, depending on the NO_x (and VOC) concentrations. In NO_x limited conditions (i.e. rural locations, but also at times of high photochemical activity on hot sunny summer days), a long-term reduction in NO_x emissions lead to a surface ozone decrease. In polluted or urban areas with large NO_x emissions (VOC or radical-limited conditions), or under conditions of lower photochemical activity like night-time hours, cloudy days, in wintertime, decreasing NO_x concentrations can increase ozone, also because ozone titration by NO is reduced (Lefohn et al., 2018 and
1870 references therein). Furthermore, the ozone trends also depend heavily on the chosen ozone metric (Lefohn et al., 2018). Here, we used the monthly means of the 11h30 UTC values, because the ozonesondes are launched around this time, which is a very limited frequency for surface ozone measurements. Making use of full frequency (at least hourly) of surface ozone measurements, e.g. Tørseth et al. (2012) and Lefohn et al. (2018) reported that the large NO_x emission reductions that have occurred in the past several decades in the European Union (EU) have led to a compression of the ozone distribution, where
1875 the high levels shift downward (reduced ozone peak concentrations) and the low levels shift upward. These trends are actually observed for sites in Brussels (Paoletti et al., 2014) and for the Uccle site (see Fig. S10), although there seems to be a levelling off in those opposite trends for low and high ozone concentrations since 2000 compared to the decade before (see again Fig. S10).
To conclude, explaining the increasing mean surface ozone amounts in combination with the decreasing ozone precursor emissions at Uccle is less straightforward than the (opposing) trends in high and low level ozone concentrations due to the compression of the surface ozone distribution. The interpretation of the increasing mean surface ozone concentrations is hampered by the interplay of many factors such as meteorology and transport, the non-linear dependence of the ozone concentrations on the emissions of VOC and NO_x, the dual role of NO_x as ozone source or sink depending on the season, and the amount of NO_x emissions.

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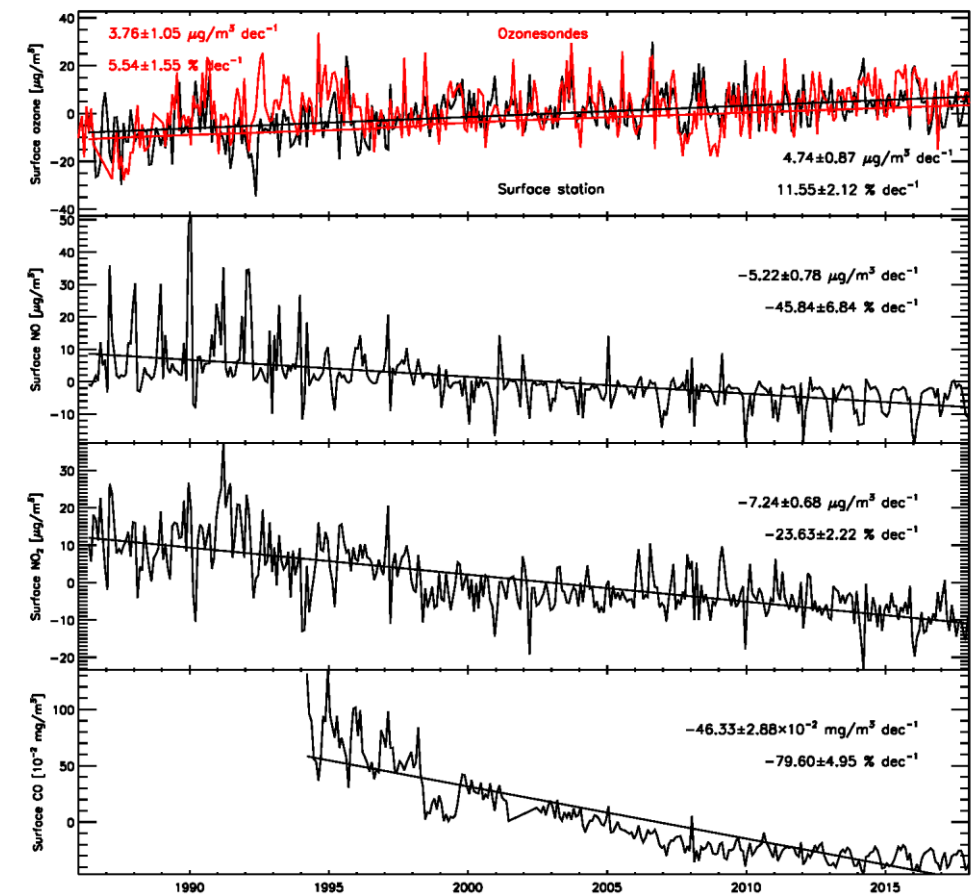


Figure B2: Monthly anomaly time series of Uccle surface ozone (upper panel, black) and mean ozone in the lowest 1 km above Uccle from the ozonesonde launches (upper panel, red) and ozone precursor measurements at Uccle (NO, NO₂) and Elsene (CO, 5 km from Uccle). Linear trends are shown, together with the absolute and relative trend estimates, and their 2σ uncertainties.

1890 Code/Data availability

The ozonesonde and total column ozone data used in this paper is publicly available through the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) and the Network for the Detection of Atmospheric Composition Change (NDACC). The MOZAIC/CARIBIC/IAGOS data are available at <http://www.iagos.fr> and the surface ozone and ozone precursor data at Uccle can be found at <http://www.irceline.be>, the website of IRCEL-CELINE (Belgian Interregional Environment Agency). The AURA MLS v4.2 Uccle overpass data were obtained at <http://avdc.gsfc.nasa.gov/pub>, the TES data at <https://search.earthdata.nasa.gov/>. The source code of the LOTUS regression model is publicly available at https://arg.usask.ca/docs/LOTUS_regression.

Author Contribution

RVM prepared the manuscript, with contributions from all authors. DDM wrote and took the lead of Sect. 2, HDB wrote Sect. 2.2.3 and made Fig. ~~3~~ [in Sect. 4.1A1](#). DDM and HDB developed the ozonesonde data processing method and tools. DP performed the analysis for Sect. 4. ~~12~~ and 5.2, and wrote Sect. 5.2. WWV wrote and did the analysis for Sect. 5.3 and helped in preparing Sect. 4. ~~2~~, [Sect. 5.2](#), and [Appendix B3](#). VDB performed part of the analysis in Sect. 4. ~~23~~ and wrote that part. AD wrote and did the analysis for Sect. 5.1. MA provided the De Bilt ozonesonde dataset and gave feedback. FF provided the surface ozone and ozone precursor data at Uccle, prepared Fig. [S108](#), and helped in preparing Sect. 4. ~~23~~ and [Appendix B4.4](#). VT gave guidance on the use of the IAGOS data at Frankfurt airport. All authors provided comments on the manuscript.

Competing Interests

The authors declare that they have no conflict of interest.

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