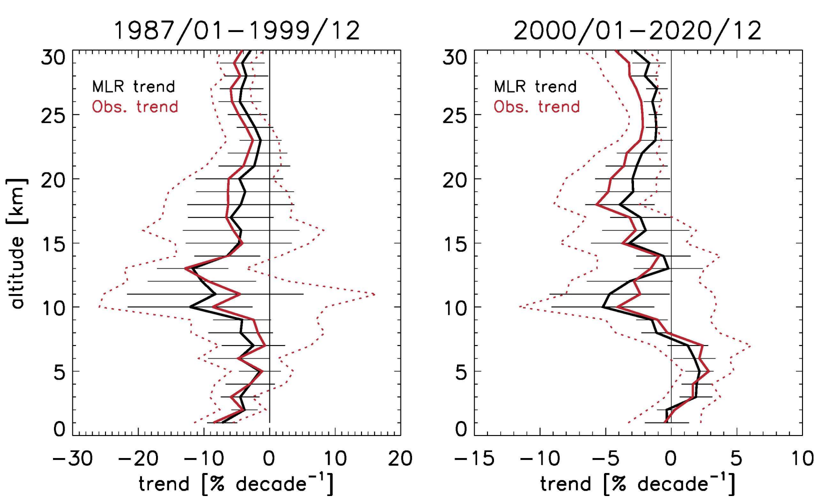
Authors' response to reviewers' comments by Guang Zeng et al.

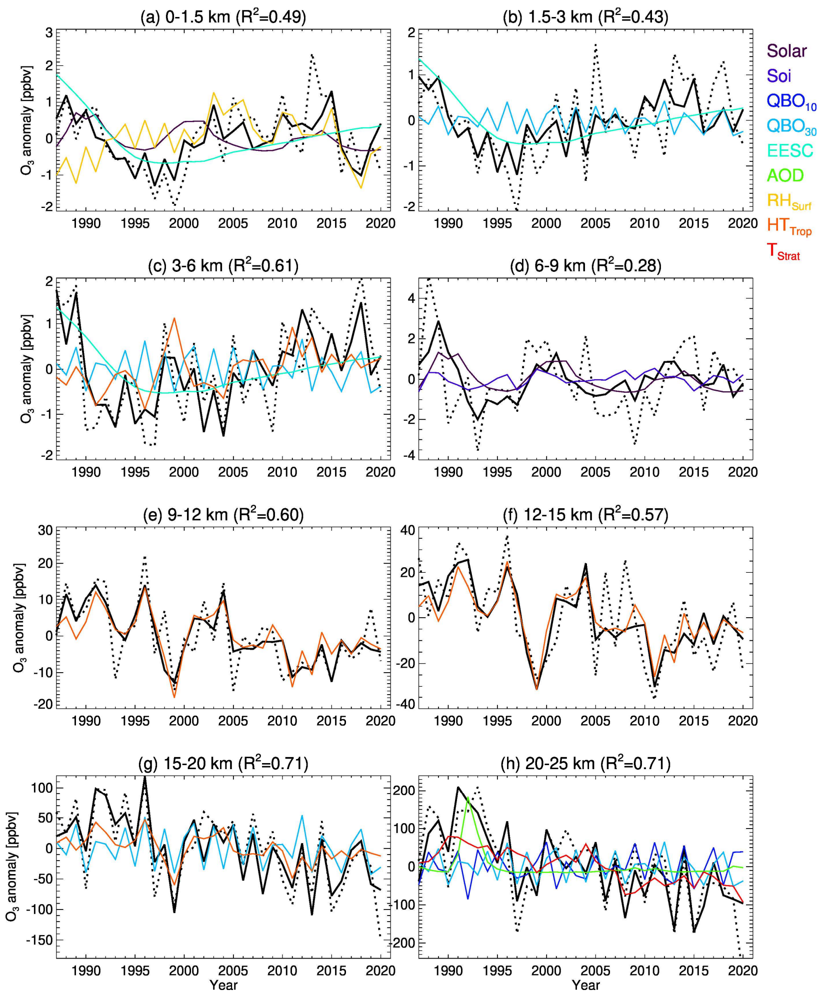
We thank both reviewers for their encouraging comments and very useful suggestions, which have helped improve the manuscript. We respond to each comment and suggestion that was raised by the reviewers and revised the manuscript accordingly. Our response is highlighted in blue. We note in red where the changes occur in the revised manuscript.

Major changes are summarised as below.

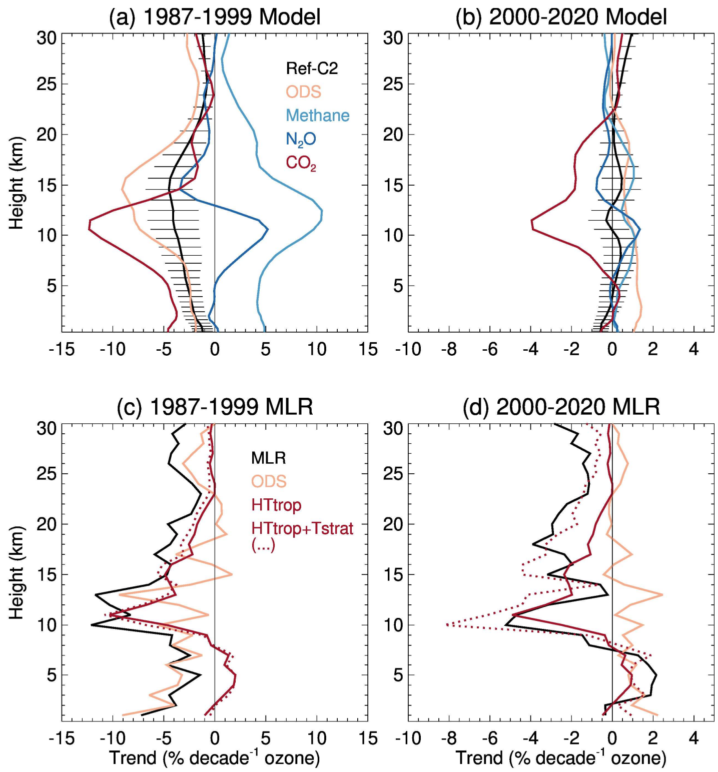
1) In response to the reviewers' concerns, we have modified the MLR model by removing the linear term, using non-detrended tropopause height for regressor, and added stratospheric temperature (averaged between 22 and 30 km) for regression. The stratospheric temperature accounts for the CO2 impact above the UTLS region where the impact of tropopause height change is minimal. As a result, the related texts and discussions have been changed accordingly.
2) We use annual mean ozone anomalies for all trend calculations and for MLR regression to minimise autocorrelation in the data. The trend values are similar with those calculated using the monthly data but with some change in the uncertainty range.
3) We added vertically resolved linear trend in regressed ozone and compare it with the linear trend in observed data (New Figure 4). The trend values are similar, but the uncertainty range is smaller in the MLR trend. There is a systematic underestimation of the MRL trend above 18km indicating that the MLR regressors do not fully capture the observed trend there, although the difference is quite small.
4) We have used all available ensemble numbers of the NIWA-UKCA simulations, in contrast to that only one ensemble number of each simulation was used in the original manuscript. This results in a small change in the modelled trend, but there are no qualitative changes.
5) We have added the contribution of EESC, tropopause height, and the combined tropopause height and stratospheric temperature (to more completely account for the CO2 effect) to the trend in regressed ozone to compare with the model attribution (new Figure 7).
6) We have combined Figures 7 and 9 (new Figure 5), and removed Figures 5, 6 and 8.
7) The subsection headings have changed slightly to better reflect the contents.



New Figure 4. Vertically resolved linear trends in regressed ozone (black) and in homogenised observed ozone (red) and their uncertainties (±2σ) at Lauder over two periods, i.e., 1987-1999 and 2000-2020. Data used for linear trend calculations in both cases are annual mean anomalies.



New Figure 5. Regressed ozone anomalies (black curves) and Observed ozone anomalies (black dotted curves, homogenised data) at Lauder (1987-2021) for eight vertically averaged layers. Contributions from Leading regressors for each layer are displayed in coloured curves (colour keys in the right of the plot.



New Figure 7. Upper panel: Vertically resolved trends in modelled annual mean ozone anomalies averaged in the area of 160E-180E and 40S-50S (representing the location of Lauder) for the periods of 1987-1999 (a) and 2000-2020 (b) from the NIWA-UKCA CCMI RefC2 simulation with the 2σ uncertainty range (black), and the trend changes due to changes in the ozone depleting substances (ODSs), methane, nitrous oxide (N_2O), and CO_2 over the same period. Lower panel: Vertically resolved ozone trends (at 1 km resolution) in the predicted ozone by the multiple linear regression (black) and the contribution from EESC (orange) , the tropopause height (red), and the combination of the tropopause height and the stratospheric temperature changes (dotted red), for the periods of 1987-1999 (c) and 2000-2020 (d).

Response to reviewers' comments

Our responses are highlighted in blue.

Review #1

The paper describes an updated / homogenized ozone sonde dataset from the Lauder station in New Zealand. It shows results from a trend analysis of the updated sonde dataset, as well as trend results for the region around New Zealand from simulations by the NIWA-UKCA chemistry climate model. The Southern Hemisphere is a region with quite sparse ground-based observations, so Lauder is a very important station and the presented material is, in principle, well suited for publication in ACP.

Overall the manuscript is generally well written, the Figures are clear, and the use of English is generally good.

Nevertheless, I have a number of questions and suggestions that should be addressed before the paper can be accepted for ACP.

We appreciate the reviewer's encouraging comments and address individual comments as below.

In section 2.2 the authors introduce their multiple linear regression model, which includes a single linear trend and EESC as proxies for long-term variations / trends. However, in section 3.2 / Figs. 3 and 4, they show linear trends for two periods (1987 to 1999 and 2000 to 2020). It seems that these trends were calculated with just one linear trend proxy in a simple linear regression, without additional proxies. Is that correct? If so, that needs to be stated very clearly, and the sequence of the (sub)-sections should maybe be reordered. In the current text, this has confused me, and will probably confuse most readers. Alternatively, two linear trends (one over each period), or a trend and change of trend (hockey stick), could be used in the multiple linear regression to be consistent throughout the paper.

The ozone trends shown in Figures 3 and 4 are indeed linear trends calculated for pre-2000 and post-2000 period separately. We have clarified this in the revised version. We have also combined Figure 3 and 4 into new Figure 3. (Sect 3.2)

We have now added the linear trend calculated for the ML regressed homogenised ozone for comparison with that calculated directly from the observed (new Figure 4). They are broadly consistent but the uncertainty for the observed linear trends is slightly larger. (Line 236-246, Sect. 3.3)

In constructing the MLR model, we use EESC as the regressor which resembles a hockey stick. We then calculated two different linear trends for the two separate periods, to compare with linear trends calculated directly based on the observed data. (Line 139-142, Sect. 2.2)

The same kind of question applies to the trends for the CCMI simulations. Were these obtained with just a simple linear trend, over the two different periods, or with the full multiple linear regression model?

Indeed, the trends for the CCMI modelled ozone are just simple linear trends for the two separate periods, calculated from the diagnosed annual mean anomalies for each vertical level, the same as in Zeng et al. (2022). We have clarified that in the revised manuscript. We do not use MLR on the modelled data. Instead, we use the sensitivity simulations to attribute the causes to changes in ozone trends.

In line 134, the authors state that "Observations as well as basis functions are smoothed using a 12-month boxcar filter". This is not a usual approach and could affect the derived uncertainties quite significantly, because essentially the number of independent data points is reduced by a factor of 10. How is this accounted for in the uncertainties? How are the uncertainties derived in the first place? How do the authors account / correct for autocorrelation in the residuals (ε (t) in their Eq. 1)? How would the results and uncertainties look without doing this 12-month boxcar? I would assume that much lower values for R^2 would be found compared to the quite high values in Table B1. My suggestion is to not use this 12-month boxcar, and go with the standard approach using monthly means without smoothing. In any case, these questions need more analysis and more discussion in the manuscript.

Indeed, the regression using smoothed data has introduced autocorrelation. We now use observed annual mean anomalies in constructing the regression model. In this case, the autocorrelation of the data has been largely removed (as shown by the Durbin-Watson test), but the uncertainties have increased. The results are largely unchanged. (Line 132-134, Sect. 2.2)

We have decided not to use the monthly mean data as the simple MLR we applied here cannot capture the very noisy monthly signal. We aim to identify the drivers for large-scale interannual variations and trends. We deliberately use the regressors that are known to impact the trends and variability of ozone, e.g., the tropopause height changes, that are not included in the LOTUS regression model. Capturing seasonal variations would require a more complicated MLR with more predictors, like the LOTUS regression model. We hope this approach is satisfactory and believe is more suited to quantify the processes that drive the ozone variation and trend. The linear trend calculated for the MLR predicted ozone is very similar to the linear trend calculated for the observed ozone data (new Figure 4), but with a smaller uncertainty range.

Figures 3 and 4: I suggest to combine both figures and plot the trends before and after homogenization in the same plot, in different colors. As the figure stands now, it is very difficult to see how the homogenization changed the trends (very little after 2000?).

We have combined the two figures as suggested (new Figure 3).

Figure 11: This is a very good and interesting Figure. However, I sorely miss the observed trends here. Please include those in the two panels. How do the vertical profiles of regressed EESC and GHG / overall linear trends from the multiple linear regression of the observed data look like in the two different time periods? How does that compare to the corresponding simulated trends (orange lines, red lines in the Figure)? How do the overall trends compare (to the black lines in the Figure).

We have added those regressed trends in the figure (new Figure 7) and the related discussion in the revised version. It shows that the modelled trends underestimate the regressed observed negative trends in general, especially in the UTLS region where the dynamical change is large and difficult to model. We have added some discussion on the difference between modelled and observed trends. (Line 342-352 and line 378-382, Sect. 3.4)

Line 27: Here you write that SH stratospheric ozone trends are dominated (controlled largely might be a better expression) by Antarctic ozone depletion (which is large and significant and controlled by ODS). Yet in your regression you find hardly any significant impact of ODS / EESC (e.g. in Fig. 8). This is quite a big and important discrepancy. Yet in the later parts of the manuscript, in the conclusions and in the abstract, this discrepancy is hardly mentioned at all, let alone resolved. You do mention negative stratospheric ozone trends, but those seem to be from simple linear regression, not multiple linear regression. So what's going wrong / different with the multiple linear regression? Is the right approach used? I think this needs to be cleared / understood.

We have modified the statement at the beginning of the introduction to “Since the late 1970s, due to the release of man-made ozone depleting substances (ODSs), Southern-Hemisphere stratospheric O3 changes are mainly characterised by Antarctic ozone depletion leading to negative trends in stratospheric ozone``. (Line 35)

In this study, we have identified that, although the most important ozone driver before 2000 in the Southern Hemisphere is ozone depleting substances which cause Antarctic ozone depletion, ODSs do not appear to be the only major factor that drives negative ozone trends over Lauder. CO2 plays an important role in driving the regional ozone trends in the lower stratosphere over Lauder.

We have now added the attribution of regressed ozone trend changes due to EESC and the tropopause height (and the combined tropopause height and middle stratospheric temperature) for comparison in new Figure 7. It shows that ODS contribution to the regressed negative ozone trend is comparable to the CO2 impact in the lower stratosphere over the period 1987-1999. The attribution of the ODS and the CO2-driven impacts in the MLR are broadly consistent with the model results. We have emphasised this finding in the revised version. (sect. 3.4)

Line 67ff: Please indicate if these differences are resolved now. It seems that the Lauder sondes trends don't change very much by the homogenization (Fig. 3, post 2000 trends are almost the same, pre 2000 trends have become more negative). So I would assume that your paper does not change the Godin et al. results, except for the Lauder FTIR data trends which have changed and now fit with the sonde trends. I find this question important, and I would like to see answers, both already here, and also later in the paper, e.g. in the conclusions.

We agree that our trend calculations are within the uncertainty range of Godin-Beekmann et al. and we have clarified this in the revised version where appropriate. (Line 72-79, line 212-215)

Line 130: What happens when tropopause height is not detrended? I think this should be tried and discussed. If non-detrended tropopause height picks up a GHG induced climate-change related ozone trend, that might be the correct way to do the trend analysis. One could argue that the mechanism underlying short term changes of tropopause height and ozone changes also acts on the long time-scale, because climate-change statistically favors high tropopause conditions. So the acting processes could be the same. There may not be the need for a different process acting on the long time scales. E.g. in the annual cycle you also have a close correlation (on a longer time scale) between high tropopause height and low ozone in the lower stratosphere. While this is somewhat different from the short time-scale processes due to high and low pressure systems, both time scales give similar correlation of high tropopause with low ozone.

We have modified the MRL approach and now use the non-detrended tropopause height. We also removed the linear term in MLR, but added stratospheric temperature which explains to a certain degree the negative trend above 20km. (Sect. 2.2)

Line 146: Please explain what forcings are included in RefC2. I assume all forcings.

The RefC2 simulations include all forcings (ODS, GHGs, ozone and aerosol precursors). This has been added in the text. (Line 165-167)

Lines 147, 150: I think this should be "corresponding fixed forcing simulation" not "corresponding single forcing simulation".

Yes, that's correct. We have modified the text. (Line 169, 171)

Around Lines 186, 197: I think these different trends need to be explained / need a bit more dicussion. If Godin et al. get different trends from the same data, there must be an explanation. What happens if you try Godin et al.s regression? Could the difference come from excluding a few extreme soundings? Generally, differences from previous findings need more explanation. They should not just be mentioned and then ignored. I have also done multiple linear regression (with hockey-stick trends) on HEGIFTOM data from Lauder and find trends very similar to your trends in Fig. 4. This is reassuring. Maybe the Godin at al. Lauder trends were too negative? Anyways, I think this needs a bit more discussion, and maybe should be mentioned in conclusions and abstract as well. Of course it needs to be worded appropriately.

We agree with the reviewer that we need to understand the cause of the discrepancy. A rationale for this study is that we verify the trend calculated by Godin-Beekmann et al. by calculating the linear trend on the observed time series, and the trend in the MLR predicted ozone. As mentioned in the manuscript, excluding some outliers would not change the trend much but would only reduce the uncertainty slightly. We then calculate the linear trend of the regressed observed ozone and find that the difference between them is quite small, especially below 18km where the largest difference occurs between our calculation and Godin-Beekmann's. However, our trends are within the uncertainty range of Godin-Beekmann et al. We have clarified this in the revised version. The observed linear trend and the MRL linear trend in our study also show some small differences in the stratosphere. The reason might be that there is some process missing in the regression, for instance, the recent Australian bush fires that affect the stratospheric ozone but are not considered in the regression. Therefore, it is important to verify the trend calculation with different approaches. (Line 217-219, line 236-246)

Line 237: As mentioned above, this really begs the question what happens with non detrended tropopause height in the regression.

We have now modified the regression model to use non detrended tropopause height and revised the related discussion. (sect. 2.2)

Line 242: I am not sure about this linear trend term. It seems like a very unspecific overall collector of various things, picking up a confusing mix of ODS-related, GHG-related and other changes. I think there should be better proxies, e.g. hockey stick, two linear trends, ... For me, the insignificant ozone changes picked up by the EESC term are a warning sign. If, according to the regression, ODS changes had no impact on stratospheric ozone at Lauder, I very much wonder if we can trust this regression.

We have now removed the linear term in the MLR model and used non-detrended tropopause height as regressor. We have also added the stratospheric temperature as a regressor to additionally account for the impact of CO_2 change in the stratosphere not covered by the impact of tropopause height changes.

The small regression coefficient representing the EESC impact is for the whole period in regression, which could obscure its more significant impact for the pre-1999 period. As shown in new Figure 7, the impact of ODS on stratospheric ozone trend at Lauder is not negligible in the pre-1999 period, especially in the lower stratosphere in the MLR attribution (Figure 7 lower panel); this is consistent with the model attribution (Figure 7 upper panel). We have explained this in the revised version. (Sect. 2.2, Sect3.4)

Around line 258: This is a weird argument. We see EESC effects in the troposphere, but we don't see them in the stratosphere, where they should be coming from? Can that be resolved? Might different proxies in the regression help (as suggested above)?

As stated above, the impact of ODS on ozone at Lauder is more important in the pre-1999 period, shown in both the MLR and model attributions (new Figure 7). The small regression coefficient due to EESC in the stratosphere is perhaps obscured by the large impact of CO2-driven dynamical changes throughout the whole observational period, not just for the pre-1999. The post-2000 period sees the impact of EESC dropping considerably therefore the small regression coefficient. However, we do calculate the trend in two separate periods to account for the effect of the EESC's transition from increasing to declining. We hope that this explanation is satisfactory. We have added more discussions in various places in the revised version.

Consequently, the EESC impacts tropospheric ozone at Lauder through transport. Model simulations (Figures B2 (d) and B3 (d)) show that tropospheric ozone at SH mid-latitudes can be influenced by polar ozone changes in the lower stratosphere. Our model simulations also capture the impact of ODS on tropospheric ozone trend in both the pre-1999 and post-2000 periods (new Figure 7).

(e.g., line 346-352)

Around line 320 (and in several other places). Are these simple single linear trends? Or are they from the multiple linear regression? This keeps me confused throughout the manuscript. I would much prefer to just have one type of regression, or a much better explanation of what was done, and why there might have been two approaches.

They are simple linear trends. We have modified the text to make it clear which of the two approaches (LR and MLR) we have used. We have added the MLR trend in the revised version for comparison – they are quite similar. This means that the MLR captures the observed trend well and we have confidence in that it can be used to explain what drive the observed trend.

After line 339: As also mentioned above for Fig. 11, I think there needs to be more discussion about how the observed and simulated trends fit together, or do not fit together.

We have added discussion on the new Figure 7. Although the model underestimates the observed (regressed) trend especially in the lower stratosphere, the roles of ODS and CO2 on ozone are consistent between the model simulation and the regression. (Sect. 3.4)

Review #2

In the manuscript "Analysis of a newly homogenised ozonesonde dataset from Lauder, New Zealand", Guang Zeng et al. derive long-term MLR trend estimations from the ozone sonde dataset of Lauder. The dataset, recently homogenised in the frame of HEGIFTOM, shows negative pre-1999 trends in better agreement with the previous literature. Post-2000 trends are shown to be significantly negative in the stratosphere and positive in the troposphere, and are in very good agreement with trends derived from a co-located FTIR instrument. The analysis by MLR imputes the negative post-2000 stratospheric trend to anthropogenic forcing led by CO2 related to positive trends in tropopause height and tropospheric temperature and negative trend in stratospheric temperature. CCM simulations from NIWA-UKCA attribute the negative pre-1999 trends not only to the ODS increase but also to a GHG increase with opposing impacts of CH4 and CO2. For the post-2000 period, the CCM analysis assess the role of the dynamical changes driven by CO2 on the negative lower stratospheric trends.

The manuscript fits well within the scope of ACP and is of high scientific quality. It is generally well written despite some very long sentences which do not read well. The results are well presented. The homogenization of the ozone sonde dataset succeeded in improving the pre-1999 trend agreement with other observation techniques and with the literature.

The detailed analysis of the MLR results is a very good contribution to the understanding of the underlying issues of stratospheric and tropospheric pre- and post-2000 trends. The results derived from CCM analysis and derived from MLR on observational dataset enhance the role of CO2-driven dynamical changes in the lower stratospheric trend which represents a significant step towards understanding trends in this atmospheric region.

I list below general and specific comments. General comments are questions and remarks which need clarifications before the paper can be accepted for ACP. Specific comments are minor revisions which may help to improve the readability of the manuscript.

We thank the reviewer for his/her encouraging comments and very useful suggestions, addressed below.

General Comments

Line 64-74:

The same ozone sonde dataset is used in Godin-Beckmann et al. 2022 and in the present study. The authors say the trend values of Godin-Beckmann et al. to be "exceedingly large" (line 69). However, the present study trends are within the Godin-Beckmann et al. uncertainties and respectively, except for the 25km value. The authors should comment on these differences.

We agree with the reviewer's assessment that the trends calculated in this study are broadly within the range of uncertainty by Godin-Beekmann et al. We have clarified this in the revised version. The different trend calculations (e.g., the linear trend, MLR trend, and LOTUS trend) may have led to some differences. Differences in the regressor selection could lead to some differences in the regressed ozone values. The purpose here is to calculate the simple linear trend based on observed data, although the uncertainty in observed trend is often larger than the regressed trend, as demonstrated in our revised version in which the observed linear trend and the MLR trend are compared. (Line 212-215)

Why are the uncertainties of both studies so different? Is the residuals autocorrelation taken into account in the present study? What is the impact of using EESC as an additional explanatory variable on the trend values of the present study?

Indeed, there is autocorrelation in our monthly data used for linear trend and MLR model. In the revised version we use annually averaged ozone anomalies for the trend calculation and the MLR model construction, which largely removed the autocorrelation. As a result, the uncertainty of the trend becomes slightly larger. Our trend uncertainties sit within or close to that by Godin-Beekmann et al. (Sect. 2.2, Sect. 3.2)

The EESC explains the tropospheric more than the stratospheric ozone trend. This might be due to that its impact on stratospheric ozone is obscured by the larger impact of dynamical changes (due to CO_2) on stratospheric ozone. We have discussed this in the revised version. (Sect. 3.4)

Line 74:

“which has been updated from the dataset used in Godin-Beckmann et al. (2022)”. Could you please comment already here (instead of Line 194) on the FTIR dataset update as the trends reported in Godin-Beckmann et al. are very different from the present study?

We have revised the text to add comments on updated FTIR retrieval here. (Line 84-87)

Line 77:

“into the near future”: I cannot not see any mention of post-2022 results in the manuscript. What do you mean?

This sentence has been removed in the revised version.

Line 89:

Table1 indicates only 3 dual flights to evaluate the effects of the sensing solution change. Is the transfer function/correction factor derived from these 3 dual flights or is a general transfer function used?

A general transfer function was used.

Line 122:

« Surface humidity are measured » : please replace by: « Surface humidity is measured »

Is this RH?

"Surface humidity are measured by the radiosonde that has a humidity sensor.": please be more specific.

Yes, it is "relative humidity". We have added this information and made the correction. (Line 142)

Line 126:

Why is QBO10 used for the whole altitude range? Would QBO50 have been more appropriate in the troposphere?

We use the same regressors for all levels. We have tested using the QBO50 index but it makes no difference versus using the QBO30 index.

Line 130:

"Fig 6." is the first mention of a figure in the manuscript, in that case, this should be Fig 1.

I suggest to remove this figure or to move it to Appendix B.

We have removed this figure from the revised version.

Line 140:

"coarse resolution » : do you mean spatial or temporal resolution? Please be more specific.

It refers to spatial resolution. "spatial" has been added before "resolution". (Line 160)

Line 177:

"marked differences": the pre-1999 trends values are different but similar within their uncertainties. The marked difference lies in the significance of the trends. This should be made clear. Furthermore, the differences would be clearer if both trend profiles (uncorrected and homogenised) were represented on a single figure.

Indeed, that's the case. We have now combined the original Figures 3 and 4 into new Figure 3 and replace "marked" with "systematic". (Line 203)

Line 184:

“Figure 3 and 4”: I suggest to merge the information into one single figure with 2 panels (pre-1999 and post-2000) to make comparisons easier.

We have combined these two figures into one and modified the text accordingly. (Sect. 3.2)

Line 187:

same comment as for Lines 64-74. Please explain why the trends values are different in both studies. The role of the additional proxies used is non negligeable. For instance, the significative contributions of the EESC or HTtropo proxies (Table B1) are influencing the remaining linear trend value when compared to a MLR not considering these proxies.

Originally the MLR model contained terms that had not been detrended. This implies that the explicit trend term differs from what would result from a straight linear fit calculation. However, the MLR model has now been changed so the linear trend term no longer exits, and the HT_trop term is not de-trended. (Sect. 2.2)

Line 195:

“updated version”: See comment for Line 74.

We have revised the manuscript as suggested. (Line 84-87, line 220-224)

Line 197:

The agreement between ozone sonde and FTIR trends is good however trends of the present study stay negative and significant above 15 km while WMO 2022 shows positive but non significant trends. A reason or a tentative explanation should be given.

Indeed, the combined satellite data show slightly negative trends in the lowermost stratosphere averaged over 35S-60S but with a very large uncertainty. Both CCMI-1 and CMIP6 models show slightly positive trend but none are significant. Lauder’s ozonesonde and FTIR data both show significant negative trends. Globally, the role of CO_2 in driving the negative ozone trend in the lowermost stratosphere is robust, especially in the tropics to the mid-latitudes (shown in the model attributions on the global scale: Figures B2 and B3). There are also large dynamical variations in this region which may obscure and reduce the significance of the trend. (Line 360-365)

Line 200-207:

At that point, I cannot find a reason for these considerations on the seasonal variation.

I suggest to remove this section unless it is used in the explanation of the trends estimation.

We have removed this section and the figure in the revised version.

Line 213:

As said in comment of Line 130, Figure 6 could be removed. If kept and moved in Appendix B, please replace “de-trened” by “de-trended” and “12-boxcar” by “12-month boxcar”.

We have removed this figure.

Line 210-217 and Figure 7 8,9 and table B1:

Separate and redundant information is spread over 3 figures and 1 table, I find this difficult to handle. I would suggest the following simplification:

Information on Fig 7 is given in Fig 9 except for the R2 value and the regressed timeseries. You could remove Fig 7 and keep only Fig 9 with the observed timeseries in dashed, the regressed timeseries in black and with the R2 information in the respective subplot titles.

We have combined Figures 7 and 9 into the new figure 5 and moved Table B1 to the main text (Table 3).

Figure 8 and table B1:

The information is redundant and Figure 8 could be replaced by a highlighting of the major contributor(s) in table B1. Table B1 should then be part of the manuscript and not of the Appendix B.

Table B1 is now new Table 3 with the major contributions being highlighted in bold. We have removed Figure 8.

Line 223:

“The downward trend in the stratospheric ozone is clearly explained by the significant negative linear trend that represents all quasi-linear, monotonic drivers of change”

Please add which drivers and that you will discuss these below.

Why can these drivers not be used as proxies but have to be treated apart?

We have now modified the MLR and removed the linear term. As suggested, we now use non-detrended tropopause height as a regressor. We have also added stratospheric temperature (averaged between 22-30km) as a regressor to explain the negative ozone trend in the stratosphere (the impact is most significant at above 20km). We have revised the text to accommodate the modification of the MLR analysis. (Sect. 2.2, Sect. 3.3)

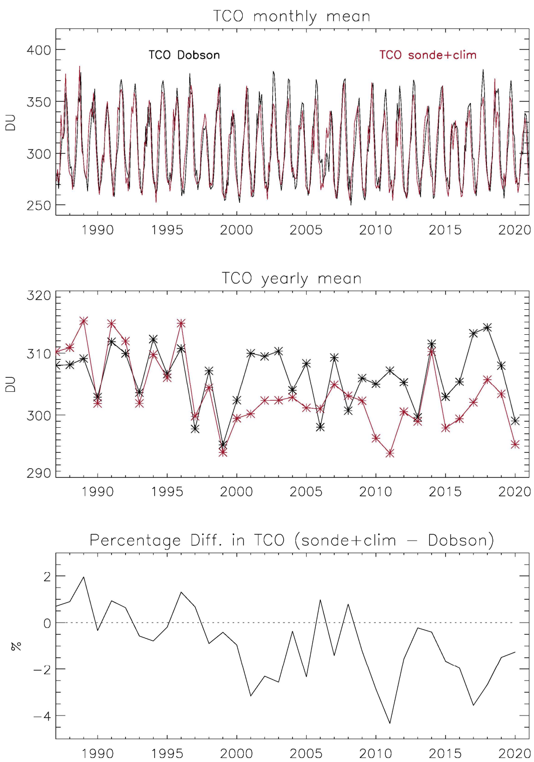
Why is the contribution of EESC negligeable here?

The contribution of EESC is most significant in the polar lower stratospheric region. In the SH mid-latitude, as we show here, the CO2-driven dynamical changes play a more significant role than the ODSs. Also the effects of ODSs and GHGs are coupled, and it is not easy to separate them in a regression that does not identify causes. From our model results, the role of ODSs is significant in the pre-1999 period (new Figure 7). We have added discussions on this. (e.g., Sect 3.4)

Can you exclude this negative trend to be due (evt partly) to a drift or step in the timeseries which has not been considered or corrected by the homogenisation?

We cannot 100% exclude that there might be drift or step in the time series. However, the study by Bjorkland et al. (2023) (https://doi.org/10.5194/egusphere-2023-2668) shows that there is no significant drift in the stratosphere between ozonesonde and FTIR ozone partial columns at Lauder.

To test, we calculated the total ozone column from integrating the partial columns below 50hPa (which is usually below the balloon's burst point) and an ozone climatology above 50 hPa. We have compared this integrated column with Dobson TCO at Lauder (see the figure below). Although there are some differences between the sonde and Dobson TCO, the data after 2000 does not show visible trend difference between these two datasets, apart from some offsets. The difference between the datasets before 2000 is relatively minor compared to the large change in TCO over this period. We need further investigation on this but is outside the scope of this study.



Line 231:

How is this trend estimated?

Meng et al. trend values are estimated "on the natural variability–removed time series" (volcanic eruption, ENSO and QBO). Are you estimating the trends with a similar model as Meng et al. or with your eq.1? Please comment.

The trend in tropopause height is calculated as simple linear trend of the annual mean anomaly of the tropopause height. We have noted this in the text. (Line 278-279)

Line 236-7:

"This indicates that the negative contribution to the ozone trend in the lower stratosphere (between ~9 to 15 km) can largely be projected on the significant increase in tropopause height»

Figure 9 shows that de-trended HTtropo is a significant proxy for explaining the ozone variation between 9 and 15 km height. Instead of making considerations about the correlation between de-trend and non de-trended HTtropo and ozone, would it be possible to use non-detrended HTtropo as a proxy? If not, why?

We have modified regression model and now use the non-detrended tropopause height as the regressor and have modified the text accordingly. (Sect. 2.2 and Sect. 3.3)

Line 267:

"The regression function we construct here is more suitable to explain the stratospheric ozone changes." Does it mean that the trend values estimated below 6 km are not reliable?

The trend in tropospheric ozone is well captured by the regression model (Figure 4) but the interannual variation is less well captured in the free troposphere (Figure 5). We have clarified this. (Line 307-308)

Line 269-270:

How are the modelled ozone trends estimated? What do you mean by "separately"? If you apply a ILT multi-linear regression on simulated ozone values, please describe the MLR used in that case. If the model directly outputs the trends, please clarify.

The model outputs ozone values in units of mixing ratio. We mean that we calculate trends in the modelled data separately for the two periods (i.e., pre-1999 and post-2000). We have not applied the MLR method to modelled ozone, rather, the modelled ozone trend was calculated as simple linear trend for the two period. We have clarified this in the text. (Line 317-318)

Line 272:

Please specify here that/if the attribution of the changes in modelled O3 is done as in Zeng et al 2022 and Morgenstern et al. 2018.

Morgenstern et al. 2018 and Zeng et al. 2022 computed the ozone response to changes in each forcing and performed linear regression at each grid point. Then the linear regression coefficient was used as the measure of ozone response to that forcing. In addition, Zeng et al., 2022 also calculated the trend changes due to each forcing in attributing regional ozone changes. We added "the same approach as in Zeng et al. (2022)." After "individual forcings". (Line 314)

Line 275:

" are broadly in agreement with the Lauder observations (Figure 4)." I suggest to add the trend values estimated on the ozone sonde dataset in Figure 11.

We have now added the regressed trend and the contributions from ODSs, HT_Trop, and the combined HT_trop and T_strat to the new Figure 7.

Line 318:

"with a maximum of −9% decade−1 around 13 km": Figure 4 shows a maximum of -12%/dec at 13km.

“with significant trends at the 95% confidence above 12 and below 5 km. » Please adjust to values shown on Figure 4.

We have adjusted these numbers. (Line 393)

Specific Comments

Line 5-7:

The sentence doesn’t read well. It’s too long and contains 3 brackets. Please rephrase.

We have substantially modified the abstract to reflect changes made in the revised version.

Line 10-13:

Please, make 2 sentences from this one.

Revised

Line 19:

“…but clearly shows …” : Please replace by “… , it clearly shows …”

Changed

Line 20:

“have had an increasingly important role”: please replace by “have played an increasingly important role”

Revised

Line 20:

“in this region”: do you mean in the lower stratosphere or in New Zealand? Please specify.

We refer to the Lauder region - revised. (Line 28)

Line 23:

“and the radiation budget” : please replace by “and in the radiation budget”

Adopted

Line 39:

“over the period 2000-2020, but such observed trends are”. Please replace by “over the period 2000-2020. Such observed trends are”

Adopted (Line 47)

Line 42-45:

please make 2 sentences.

Revised

Line 48:

“attempts of attribution using the models”: Please replace by “any attempt at attribution using models”

“well-positioned » please replace by “well-suited”

Adopted (Lines 57 and 58)

Line 58:

Please remove the) after NDACC

Done

Line 60:

“Any heterogeneities the data have”: please replace by. “Any heterogeneities in the dataset »

Adopted (line 69)

Line 64:

Make a separate sentence with “although we only take the data from January 1987 to December 2020 for analysis here.”

Revised (Line 73)

Line 79:

« In the next section », please replace by "In section 2"

Changed to "Sect. 2"

Line79-80:

"section", "Sect.": please make it uniform in the manuscript.

We have adopted "Sect."

Line 98:

"For example » : please replace by "For instance"

Done

"is needed for the change of sensing solution because there was a 2-year period when the EnSci ECCs started to be used, but with the 1% solution, rather than the 0.5% solution which has become the recommendation for the EnSci ECCs." is difficult to read. You could replace that sentence with:

"a transfer function is applied to the data after the change in sensing solution type from 1% to 0.5% KI following the O3S-DQA recommendation"

Btw, is the transfer function applied to the data before or after the change of sensing solution?

A general transfer function is used to correct the profiles that used the 1% KI solution instead of the recommended 0.5% for the EnSci ECCs flights during the period of 1994-1996. We have revised the text to make this clearer. (Line 111-115)

Line 100:

"re-process » : please replace by "re-processing"

Adopted (Line 115)

Line 105-110:

this paragraph does not read well and need to be rephrased.

For instance: "Both homogenised and the uncorrected datasets have been post-processed for trend calculations and the regression analysis (in the case of homogenised data)."

Shoud be: "Both homogenised and uncorrected datasets have been post-processed for trend calculations and for regression analysis."

What do you mean by "(in the case of homogenised data)"?

We use the homogenised data for MLR analysis.

We have revised this paragraph. It now reads "*In this study, we include a total of 1958 flights between August and June 2021, which the data have been homogenised. Both homogenised and uncorrected dataset have been post-processed for linear trend calculations. The homogenised datasets are used in the MLR analysis. Linear piecewise regression was applied to interpolate the original ozone profiles from the surface to 30~km at a 1~km vertical resolution. We then exclude some extreme ozone values, identified as the values that are outside the 3 standard deviation range, to create monthly means by averaging the data available for that month at each re-gridded vertical level*.'' (Line 120-126)

Line 118:

Please add 3 "the": The tropopause height (HTTrop), the surface relative humidity (RHsurf), the aerosol optical depth (AOD),

Done (Lines 137 & 138)

Line 123:

Please replace "WMO (1957)" by "(WMO, 1957)"

Done (Line 144)

Line 124:

"using the co-measured temperature data of each ozonesonde flight": please replace by "from the temperature measured by the radiosonde during each ozonesonde flight"

Adopted (Line 144)

Line 129:

"normalized to vanishing means and unit standard deviation": do you mean "standardized"?

Yes, that's correct. Now we have added "standardized". (Line 149)

Line 152-154:

not clear, I can see redundant information in the same sentence. Please rephrase.

Do you simply mean that the impact of CO2 equals the impact of combined GHGs minus the impact of CH4 and NO2?

Yes, that's correct. We have rephrased this sentence as "*However, no simulation was performed to directly assess the impact of CO2 within CCMI-1; instead, it will be assessed by subtracting the impacts of methane and N$_2$O from the impact of the combined GHGs*." (Line 173-175)

Line 160:

I would use "as measured" instead of "uncorrected" (first mention is on Line 93).

Thanks very much for the suggestion. After some consideration, we have decided to stick with the wording "uncorrected" in contrast to "homogenised".

Line 162:

"in the vertical to 1 km grid using piecewise linear regression for each profile.": please replace by: "to a 1 km vertical grid using piecewise linear regression."

Adopted (Line 183)

Line 167:

"The effect of changes to the concentration of the KI solution on the conversion efficiency"

Please replace by:

"The effect of the changes of the KI solution concentration on the conversion efficiency"

Adopted (Line 190-191)

Line 169:

Please move "The correction procedure and the impact of each correction are described in more detail in Appendix A." before the reference to Figure A1(3).

Done (Line 184-185)

Line172:

"outliers where ozone is outside the 3 standard deviation": please replace by: "outliers defined for ozone being outside the 3 standard deviation"

Adopted (Line 195)

Line 202:

“Figure (5)”: please replace by “Figure 5"

This paragraph has been removed.

Line 205:

“Some slight differences between seasons are below 5 km”: please add “visible” after “are”

This paragraph has been removed.

Line 219-220:

“, with R2 ranging from 0.27 to 0.49 in the troposphere and 0.50 to 0.73 in the stratosphere, implying that the stratospheric ozone variations and trends are better explained by the MLR model than tropospheric features.”, I would say:

“. With R2 values ranging from 0.27 to 0.49 in the troposphere and 0.50 to 0.73 in the stratosphere, the stratospheric ozone variations and trends are better explained by the MLR model than tropospheric features”

Changes made as suggested. There are slight changes in R2 values as the result of modified MLR. (Line 254-257)

Line 228:

Please remove “the” before “cooling”

Done (Line 275)

Line 229:

Please remove “the” before “reanalysis”

Done (line 276)

Line 234:

"at the 9-12 km layer": please replace by: "in the 9-12 km layer"

Changed, but now the text has been removed.

Line 235:

"at the 12-15 km layer": please replace by: "in the 12-15 km layer"

Same as above.

Line 238:

Replace "at a correlation" by "with a correlation"

Text removed. This paragraph has been modified to accommodate the revised MLR analysis.

Line 245:

Replace the 2nd "together with" by "and"

Text has been revised. (Line 262-264)

Line 256:

"This trend transition follows the evolution of EESC, which after a peak in 1997 has been declining since then (Figure 9), and indicates the stratospheric impact on the tropospheric ozone through stratosphere-to-troposphere transport reflecting the effect of stratospheric ozone depletion and recovery." Too much information for one single sentence. What is "reflecting the effect of stratospheric ozone depletion and recovery " the EESC evolution or the stratospheric impact or the transport…? Please rephrase.

We have rephrased this sentence as "*This trend transition in tropospheric ozone coincides with the evolution of EESCs which increases since the late 1980s before declining after 1997; this indicates that the impact of stratospheric ozone changes due to changes in ODSs could impact tropospheric ozone through transport.*" (Line 295-298)

Line 258:

Please assign a letter or a number to the panels and refer to them as Figure 9 (a) and so on.

We have updated this figure (new Figure 7) as suggested.

Line 260 and 265:

Please replace “for example” by “for instance“

Done (Line 302)

Line 271:

Please remove “single” between “individual” and “forcings”.

Done (Line 314)

Line 312:

Please replace “height-resolved” by “vertically resolved”

Done (Line 387)

Line 320:

“In both these altitude regions the trends are substantially stronger than trends in the uncorrected data which are largely insignificant ». Trends “in” altitude regions are compared to trends “in” uncorrected data. Please rephrase.

We have removed this sentence in the revised version.

Line 326:

Please replace “altitudes” by “pressure levels”

Done (Line 408)

Line 333:

Please add “the” before “tropospheric height”

Done (Line 416)

Line 340:

Please add “on” before “the zonal mean ozone profiles”

Done (Line 427)

Line 343:

"and increases in CO_2 which lead": please replace by "and to increase in CO_2 which leads"

Adopted (Line 432)

Line 356:

Please remove "The Stratospheric Aerosol Optical Depth data was obtained from the NASA Langley Research Center Atmospheric Science Data Center (https://asdc.larc.nasa.gov/)" from the Acknowledgements but add "(https://asdc.larc.nasa.gov/) in Table 2.

Done

Line 291:

Please replace "attained" by "reached"

Done (Line 354)

Line 302:

Please replace "are negative" by "is negative" and "which maximises" by "and maximises"

Done (Lines 370, 371)

Figure 2:

please replace "3 box-car" by "3-month boxcar"

Adopted

Figure 5:

"ozone sonde" or "ozonesonde" : please make it uniform throughout the paper

This figure has been removed.

Figure 10:

Please label the panels with a, b and c. and refer to this in the text.

Done

Figure B2:

Place label the panels with a, b and c. and refer to this in the text.

Done

Lines 407,409,492

Please check Bodecker et al., Boyd et al. and Seidel et al. for doi

Doi Added

Lines 539-545:

WMO 2011,2014,2018 and 2022: citations are not complete

Corrected

Analysis of a newly homogenised ozonesonde dataset from Lauder, New Zealand

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Abstract. This study presents an updated and homogenised ozone time series covering 34 years (1987-2020) of ozonesonde measurements at Lauder, New Zealand, and ~~derived~~attributes vertically resolved ozone trends using a multiple linear regression (MLR) analysis and a chemistry-climate model (CCM). Homogenisation of the time series leads to a marked difference in ozone values before 1997, ~~Over the period of 1987-1999,~~ in which the ozone trends ~~in the homogenised ozone data~~ are predominantly negative from the surface to ∼30 km, ranging from ~ -2 to -13% decade^{-1}, maximising at around 12-13 km, in contrast to the uncorrected time series which shows no clear trends for this period. ~~These negative trends are statistically significant at 95% confidence level below 4 km and above 23 km.~~ For the post-2000 period, ozone at Lauder shows negative trends in the stratosphere~~(but the trends are only statistically significant above 17 km)~~, maximising just below 20 km ($\sim -5\%$ decade^{-1}), despite stratospheric chlorine and bromine from ozone-depleting substances (ODSs) both declining ~~in this period~~since 1997. ~~In the troposphere~~However, the ozone trends change from negative for 1987-1999 to positive in the post-2000 period in the free troposphere. The post-2000 ozone trends calculated from the ozonesonde measurements compare well with those derived from the co-located low-vertical resolution Fourier-transform infrared spectroscopy (FTIR) ozone time series. ~~A multiple-linear regression~~The MLR analysis ~~indicates~~identifies that the increasing tropopause height, associated with CO_2-driven dynamical changes, is the leading factor driving the continuous negative trend in lower stratospheric ozone at Lauder over the whole observational period, whilst the ozone-depleting substances (ODSs) only contribute to the negative ozone trend in the lower stratosphere over the pre-1999 period. Meanwhile, stratospheric temperature changes contribute significantly to the negative ozone trend above 20 km over the post-2000 period. ~~anthropogenic forcing plays a significant role in driving the significant negative trend in the stratospheric ozone at Lauder, in which the effect of greenhouse gas (GHG)-driven dynamical and chemical changes is reflected in the significant positive trends in tropopause height and tropospheric temperature, and significant negative trends of stratospheric temperature observed at Lauder. The interannual variation in lower stratospheric ozone is largely explained by the variation in tropopause height at Lauder, which is highly anti-correlated with stratospheric temperature and correlated with tropospheric temperature.~~ Furthermore, ~~the impact of ODSs and GHGs on ozone over Lauder is assessed in a chemistry-climate model using a series of single forcing simulations.~~the chemistry-climate model (CCM) simulations that separate the effect of individual forcings ~~The model simulations~~ show that the predominantly negative modelled trend in ozone for the 1987-1999 period is driven not only by ODSs, but also by increases in GHGs, with large but opposing impacts from methane (positive) and CO_2 (negative), respectively. Over the 2000-2020 period, the model

results show that the CO_2 increase is the dominant driver for the negative trend in the lower stratosphere, in agreement with the MLR analysis. Although the model underestimates the observed negative ozone trend in the lower stratosphere for both periods, ~~but~~it clearly shows that CO_2-driven dynamical changes ~~have had~~have played an increasingly important role in driving the lower stratospheric ozone trends ~~in this region.~~in the vicinity of Lauder.

1 Introduction

Ozone (O_3) plays a central role in atmospheric chemistry and in the radiation budget. The stratospheric ozone layer protects life on Earth by preventing harmful ultra-violet radiation from reaching the surface. Stratospheric ozone is also a natural source of tropospheric ozone via cross-tropopause transport; it accounts for around 30% of tropospheric ozone production (Lelieveld and Dentener, 2000). Since the late 1970s, due to the release of man-made ozone depleting substances (ODSs), Southern-Hemisphere stratospheric O_3 changes are ~~dominated~~mainly characterised by Antarctic ozone depletion leading to negative trends in stratospheric ozone (e.g., World Meteorological Organization (WMO), 2014, 2018). Due to the successful implementation of the Montreal Protocol (MP) in 1987 and its subsequent amendments, concentrations of ODSs have been declining. The most recent assessment (World Meteorological Organization (WMO), 2022) confirms that upper-stratospheric ozone is recovering, in agreement with model simulations (Godin-Beekmann et al., 2022; Zeng et al., 2022).

However, while the ODSs are declining, the future evolution of ozone depends critically on changes in greenhouse gases (GHGs). For example, decreases in stratospheric temperature caused by increasing CO_2 and other GHGs will accelerate stratospheric ozone recovery (Randeniya et al., 2002; Rosenfield et al., 2002). In the tropical lower stratosphere, climate change increases tropical upwelling, leading to less time for O_3 production and hence decreasing O_3 in this region (Eyring et al., 2010). As a result, both observations and models indicate a small but uncertain decrease of ozone in the tropical lower stratosphere which is consistent with the Brewer-Dobson circulation (BDC) change driven by increases in greenhouse gases (World Meteorological Organization (WMO), 2022). In both mid-latitude regions, the combined satellite stratospheric ozone trends are generally negative albeit non-significant over the period 2000-2020. ~~but such~~Such observed trends are not reproduced by either CCMI-1 or AerChemMIP model simulations which show generally non-significant positive trends in these regions (Godin-Beekmann et al., 2022; Zeng et al., 2022; World Meteorological Organization (WMO), 2022). The ozone distribution is typically affected by large dynamical variability in the lowermost stratosphere, limiting any attribution to anthropogenic factors. Furthermore, future changes of stratospheric O_3 could also significantly impact tropospheric O_3 and potentially air quality through stratosphere-troposphere exchange (STE) (e.g., Zeng et al., 2010; Hegglin and Shepherd, 2009). ~~in~~In the Southern Hemisphere, ~~where~~ the stratospheric ozone influx plays a larger role in the tropospheric ozone budget, relative to in-situ ozone formation, than in the more polluted Northern Hemisphere.

High vertical resolution ozone measurements are key to understanding the impact of various anthropogenic forcings on ozone changes, especially in the upper troposphere and the lower stratosphere (UTLS) where the large dynamical variability may obscure any ~~attempts of~~attempt at attribution using the models. The high vertical resolution ozonesonde measurements are ~~well-positioned~~well-suited to detect changes in ozone from the surface to around 35 km. An extensive ozonesonde measurement network exists throughout the Northern Hemisphere, but, it is sparse in the Southern Hemisphere (SH). Lauder, New Zealand (45ºS, 170ºE, 370 m above sea level), a clean rural site that is representative of the SH mid-latitude background atmosphere, is a primary member of the Network for the Detection of Atmospheric Composition Change (NDACC). The Lauder ozonesonde measurements started in 1986 and continue to provide weekly high-resolution vertically resolved ozone data from the surface to around 35 km; this is of particular relevance to detecting long-term changes in both the stratospheric and tropospheric ozone in the SH clean background air (Oltmans et al., 2006, 2013; Zeng et al., 2017).

Recently, the Lauder ozonesonde data have been subjected to a homogenisation process under the guidance of the Ozonesonde Data Quality Assessment (O3S-DQA) activity (Smit and the O3S-DQA panel, 2012), which is part of the SPARC/IO3C/IGACO-O3/NDACC~~)~~ (SI2N) initiative (Harris et al., 2011, 2012). Homogenisation is designed to produce consistent datasets with reduced uncertainties and offsets in long-term ozone vertical profiles that arise from instrumental and operating procedure changes over the observational periods. Any heterogeneities ~~the data have~~in the dataset can adversely affect trend calculations. Many other ozonesonde measurement sites have gone through this homogenisation process (Tarasick et al., 2016; Van Malderen et al., 2016; Thompson et al., 2017; Sterling et al., 2018; Witte et al., 2017, 2018, 2019; Ancellet et al., 2022), and we have applied the same procedure to homogenise the Lauder ozonesonde timeseries between August 1986 (when the observation started) and June 2021. ~~although~~Here we ~~only~~ take the data from January 1987 to December 2020 for analysis. The post-2000 homogenised Lauder ozone dataset was included by Godin-Beekmann et al. (2022) in their evaluation of near-global (60^oS–60^oN) stratospheric ozone profile trends from satellite and multiple ground-based instruments, along with datasets from several other ozone measurements, using an updated version of the Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS) regression model (LOTUS, 2019). Godin-Beekmann et al. (2022) show that the negative ozone trends in the lower stratosphere from the Lauder ozonesonde timeseries were ~~exceedingly~~significantly larger in absolute terms ~~in comparison with~~than the trends calculated from the satellite data ~~and from other instruments~~ at the same site.

In this paper, we present the homogenised Lauder ozonesonde record covering the whole observational period of 1987-2020, and evaluate vertically resolved ozone trends from the surface to 30 km for both the pre-1999 and the post-2000 periods, and contrast these with the data series without homogenisation. We calculate simple linear trends for the two periods separately and compare the post-2000 lower stratospheric ozone trend with that calculated by Godin-Beekmann et al. (2022) based on the LOTUS regression model. We also compare the homogenised post-2000 ozonesonde data to that derived from Fourier-transform infrared spectroscopy (FTIR) ozone measurements, from which low-resolution vertical profiles are derived (e.g., Vigouroux et al., 2015). The FTIR ozone profile has since been updated from the dataset used ~~in~~by Godin-Beekmann et al. (2022), based on the updated retrieval strategy presented by García et al. (2022) and Björklund et al. (2023). We aim to identify ~~the dominating~~dominant forcings that drive ozone variations and trends at Lauder using a multiple linear regression (MLR) model analysis. We will assess the roles of ODSs and GHGs (including methane, N_2O, and CO_2) in driving ozone changes over ~~the~~

last few decades and into the near futurethe pre-1999 and post-2000 periods around Lauder using simulations from a chemistry-climate model. in relation to changes in ozone trends at Lauder, representative of the background O_3 changes in the Southern Hemisphere mid-latitudes. In the next sectionSect. 2, we describe the homogenised ozone time series, construct the MLR model, and describe the CCM simulations. We then present the results and discussions in Sect. 3. Conclusions are drawn in sectionSect. 4.

2 Data and regression model

2.1 Homogenised ozonesonde records

Weekly electrochemical cell (ECC) ozonesondes have been launched in tandem with radiosondes at Lauder since August 1986, measuring profiles of ozone, temperature, pressure, humidity, and wind speeds and directions from the surface up to about 35 km (Boyd et al., 1998; Bodeker et al., 1998). The ECC used for ozone sounding at Lauder are the Science Pump Corporation (SPC) series 4A/5A/6A (before 1996) and the Environmental Science (EnSci) Z series (after 1996), although there are some overlap period when both types were used. These ECC series were operated with a 1.0% buffered potassium iodide (KI) cathode solution until July 1996 and a 0.5% KI solution from August 1996 until present. These changes are relevant to the homogenisation process and are detailed in Table 1.

The homogenisation procedure, described in the Assessment of Standard Operating Procedures for Ozonesondes (ASOPOS 2.0) documentation (Smit et al., 2021) and in the Ozonesonde Data Quality Assessment (O3S-DQA) activity (Smit and the O3S-DQA panel, 2012), was applied to the Lauder ozonesonde timeseries, available at NDACC. These NDACC data, named "uncorrected data" hereafter, have been obtained by converting the raw currents measured with an ozonesonde to ozone partial pressures by subtracting a measured background current, using a conversion efficiency of 1.0, the measured pump temperature and pump flow rate measured prior to launch in the lab, and correcting for the pump efficiency decrease with increasing altitudes. The O3S-DQA homogenization, however, add corrections to the pump temperature, the pump flow rate (due to the moistening effect), and the background current (avoiding too high values) on top, and uses a set of transfer functions applied to the conversion efficiency to remove biases due to changes in the instrument or operating procedures. For exampleinstance, as the 0.5% KI solution has become the recommendation for the EnSci ECCs, a transfer function is needed for the change of sensing solution because there was a 2-year period when the EnSci ECCs started to be used, but with the 1% solution, rather than theis applied to the profiles between 1994 and 1996 where the 1% KI solution instead of the 0.5% KI solution was used for the EnSci ECCs at Lauder. which has become the recommendation for the EnSci ECCs. The re-processing of the Lauder data according to the O3S-DQA guidelines were carried out by the HEGIFTOM working group (Harmonization and Evaluation of Ground Based Instruments for Free Tropospheric Ozone Measurements, https://hegiftom.meteo.be) within the TOAR-II (Tropospheric Ozone Assessment Report phase II, https://igacproject.org/activities/TOAR/TOAR-II) initiative. Further details regarding the corrections are summarised in Appendix A.

In this study, we include a total of 1958 ozonesonde flights between August 1986 and June 2021, which the data have been homogenised. Both homogenised and the measured uncorrected datasets have been post-processed for linear trend calculations. The homogenised data are used in the MLR analysis(in the case of homogenised data). Linear piecewise regression was applied to interpolate ~~the recorded ozonesonde data~~the original ozone profiles ~~vertically~~ from the surface to 30 km at a 1 km vertical resolution ~~grid~~. We then exclude some ~~points with~~ extreme ozone values, identified as the values that are outside the 3 standard deviation range, ~~for the whole time series at each vertical level)~~ to create monthly means by averaging the data available for that month at each re-gridded vertical level. ~~We calculate the ozone trends in two periods, i.e., the 1987-1999 and the 2000-2020 periods for grouped vertical layers from the surface to 30 km.~~

2.2 ~~Regression~~Multiple linear regression model

We construct a multiple linear-regression (MLR) model to identify the dominant factors that are associated with O_3 variations and trends. The regression model approximates the annual mean ozone anomalies for each level as well as for eight grouped layers where annual mean ozone anomalies are averaged ~~The homogenised O_3 mixing ratios are averaged over eight layers~~ (0–1.5, 1.5–3, 3–6, 6–9, 9–12, 12–15, 15–20, and 20–25 km). ~~We then construct regression models for each layer. In total,~~ The auto-correlation that usually exists in the monthly-varying data has been largely removed by averaging them into annually-varying data. The purpose of this study is to analyse the interannual variations and trends in annual mean ozone. The regression models include nine terms representing the Solar Index (SI) which captures solar variability and is defined by the solar radio flux at 10.7 cm, the Multivariate El Niño Southern Oscillation Index (MEI), the Quasi-Biennial Oscillation at 30 hPa and 10 hPa, respectively (QBO_{30} and QBO_{10}), the tropopause height (HT_{Trop}), the stratospheric temperature (T_{Strat}), the surface relative humidity (RH_{surf}), the aerosol optical depth (AOD), and the equivalent effective stratospheric chlorine (EESC). The two QBO indices are orthogonalized w.r.t. each other. The EESC is defined as a relative measure of the potential for stratospheric ozone depletion that combines the contributions of chlorine and bromine from surface observations from ODSs (Newman et al., 2007), and is calculated based on the ozone-depleting substances from the Coupled Model Project 6 (CMIP6) historical (until 2014) and Shared Socio-economic Pathway (SSP245) (for 2015-2021) scenarios (Meinshausen et al., 2017). Surface relative humidity ~~are~~is measured by the radiosonde that has a humidity sensor. We define the tropopause based on the WMO lapse rate definition (WMO, 1957), which is calculated ~~using the co-measured temperature data of each ozonesonde flight~~from the temperature measured by the radiosonde during each ozonesonde fligh. The regressed O_3 anomaly is expressed as

$$
\begin{aligned}
Ozone(t) = {} & a_1 \cdot SI(t) + a_2 \cdot SOI(t) + a_3 \cdot QBO_{10}(t) + a_4 \cdot QBO_{30}(t) + a_5 \cdot EESC(t) \\
& + a_6 \cdot AOD(t) + a_7 \cdot HT_{Trop}(t) + a_8 \cdot T_{Strat}(t) + a_9 \cdot RH_{surf}(t) + \epsilon(t).
\end{aligned} \tag{1}
$$

Here $Ozone(t)$ is the ~~monthly~~annual mean ozone anomalies ~~minus its mean annual cycle~~, ϵ is the regression residual, minimized in the RMS, ~~a_1 is the linear trend (or L-trend) and~~ a_{1-9} are the regression coefficients for the corresponding regressors, all normalized to vanishing means and unit standard deviation (i.e., standardised). All forcings used in regression are summarised in Table 2.~~, and their time series are displayed in Fig 6~~. ~~The tropopause height anomaly is de-trended because it is well-known to be influenced by increasing GHGs (whose influence is already encapsulated in the linear trend term $a_1 t$). The surface relative humidity is not de-trended as it does not have a significant linear trend over the observation period. All other regressors represent external forcings which are not coupled to the GHGs and none of them have significant trends therefore they are not de-trended.~~ ~~The two QBO indices are orthogonalized w.r.t. each other.~~ ~~Observations as well as basis functions are smoothed using a 12-months boxcar filter.~~

2.3 Chemistry-climate model simulations

We use the NIWA-UKCA model simulations from the Chemistry-Climate Model Initiative project (CCMI-1: Eyring et al. (2013), Morgenstern et al. (2017)) to assess the impact of the major anthropogenic forcings, including greenhouse gases (GHGs) and ozone depleting substances (ODSs), on ozone changes at Lauder. The Lauder ozonesonde measurements cover both the ozone depletion and the recovery periods of 1987-2020. Global chemistry-climate models (CCMs), including NIWA-UKCA (Morgenstern et al., 2009; Zeng et al., 2015, 2017), generally have coarse spatial resolutions. Therefore it is not often ideal to use the simulations from the CCMs for reproducing the observed trends at a specific location. Instead, the CCMs can be used to attribute the trends to various forcings on a wider spatial and temporal scale. Here, we calculate the ozone trends using the NIWA-UKCA simulations to gauge the impacts of GHGs and ODSs on ozone changes at Lauder over the observational period in a limited area covering Lauder (averaged over 160-180°E and 40-50°S). We also show the simulation results on a global scale in context. The CCMI-1 simulations from NIWA-UKCA used here consist of the all forcing (including time-varying GHGs, ODSs, and ozone and aerosal precursors) coupled atmosphere-ocean reference experiment "RefC2", covering the simulation period of 1960-2100 (we keep the same experiments naming convention as defined by Eyring et al. (2013)) and its corresponding fixed single forcing sensitivity ~~simulations~~experiments (sen-C2-fODS, sen-C2-fGHGs, sen-C2-fCH_4), and sen-C2-fN_2O) in which ODSs, the combined GHGs, methane (CH_4), and N_2O are individually fixed at their 1960's levels, respectively. The impact of each single forcing on ozone is derived from the differences in ozone between ~~the reference simulation~~the RefC2 ensemble mean (5 members) and the ensemble averages of the corresponding fixed single-forcing simulations (1 to 3 ensemble numbers for each experiment). We ~~can~~ directly assess the impacts of changes in ODS, combined GHGs, methane, and N_2O on ozone trends, based on available simulations. However, no simulation was performed to directly assess the impact of CO_2 within CCMI-1; instead, it will be ~~derived from the available fixed methane, N_2O, and combined GHGs experiments to subtract~~ assessed by subtracting the impacts of methane and N_2O from the impact of the combined GHGs (Morgenstern et al., 2018). ~~Unlike the ODSs, which peaked in the late 1990s, GHGs (including CO_2, methane, and N_2O) are mostly monotonically increasing. The impacts of GHGs changes on future ozone evolution are expected to be dominant while the ODSs are declining.~~ We ~~therefore also~~again separately examine the changes in modelled ozone trends over the periods of 1987-1999 and 2000-2020. The detailed description of the model and experiments can be found in Morgenstern et al. (2018) and in Zeng et al. (2017) and the references therein.

3 Results

3.1 Homogenised versus uncorrected ozonesonde time series

The homogenised and the uncorrected datasets are directly compared without any temporal interpolation, but are both interpolated ~~in the vertical to 1 km grid using piecewise linear regression for each profile~~to a 1 km vertical grid using piecewise linear regression. Figure 1 shows the percentage difference between vertical ozone profiles from the two datasets for all flights. The correction procedure and the impact of each correction are described in more detail in Appendix A. Overall, corrections lead to mostly increased ozone values in the homogenised time series, reaching 6 to over 10% before 1995 due to the pump temperature correction (Figure A1(3)). The pump flow rate correction results in a uniformly positive effect of less than 2% in general (Figure A1(4)). There are scattered increases in ozone in the homogenised time series compared to the uncorrected data, especially between 2012 and 2015 when a modified background current correction is applied (Figure A1(2)). ~~The effect of changes to the concentration of the KI solution on the conversion efficiency~~The effect of the changes of the KI solution concentration on the conversion efficiency (Figure A1(1)) is mainly negative between 1994 and 1996 but positive in the beginning of the time series (1986), when a smaller cathode sensing solution amount has been used (2.5 ml instead of 3 ml). ~~The correction procedure and the impact of each correction are described in more detail in Appendix A.~~

The differences between the homogenised and the uncorrected monthly mean ozone time series are calculated excluding ~~outliers where ozone is outside the 3 standard deviation~~outliers defined for ozone being outside the 3 standard deviation interval of all data points for that level (Figure 2). This step removes less than 1% of data points from the monthly mean ozone calculations. Most of these outliers are around 10 km where the ozone is subjected to large dynamical variations. ~~We carry out trend calculations based on the monthly mean ozone values Sect. 3.2~~

3.2 Vertically resolved trends in observed ozone at Lauder

Figure 3 displays simple linear trends from the surface to 30 km for both homogenised and uncorrected datasets over the pre-1999 and post-2000 period, respectively. Also displayed is the observed FTIR ozone trend for the post-2000 period for comparison. All trend calculations use annual mean ozone anomalies to minimise the auto-correlation in data, as opposed to using the monthly mean data. ~~Consequently,~~ It shows that there are ~~marked~~systematic differences in ozone trends in the homogenised data compared to those in the uncorrected data over the 1987-1999 period (Figure 3(a)). During this period, the vertically resolved trends in the uncorrected data set are slightly negative throughout most of the domain above 10 km and positive below 10 km, although ~~below 25 km~~ these trends are generally insignificant at the 95% confidence level. In contrast, the trends in the homogenised data are negative throughout the domain (~ -2 to -13% decade^{-1}), with most of the trends below 5 km and above $\sim$24 km being statistically significant at the 95% confidence level. This result is more consistent with the impact of increasing ozone depleting substances (ODSs) over this period.

In the post-2000 period, the calculated trends are very similar between homogenised and uncorrected ozone profiles (Figures 3). Both show significant positive trends of up to $\sim$+2% decade^{-1} in the free troposphere and a significant negative trend of ~ -2 to -6% decade^{-1} above $\sim$16 km in the stratosphere, which peaks around 18 km. We note that the lower stratospheric ozone negative trend of ~ -3 to -6% decade^{-1} between 15 and 20 km looks visibly smaller in magnitude than the trend presented in Godin-Beekmann et al. (2022) where it exceeds 6-7% decade^{-1} in the same region, but are within the uncertainty ranges displayed by Godin-Beekmann et al. (2022). Distinct negative trends of $\sim$2-4% decade^{-1} also exist in the upper troposphere and the lower stratosphere between 8 and 16 km albeit with large statistical uncertainty, highlighting the large dynamical variability typical for this region. We find that the vertically resolved ozone trends calculated by excluding the outliers ~~from creating the monthly mean ozone values~~ are very similar to the trends calculated by including all data points. ~~in the monthly mean.~~ The only difference is that by excluding the outliers we have reduced the trend uncertainties around the 10 km region (not shown). The vertically resolved trends in observed ozone for the post-2000 period are in excellent agreement with the trends drived from the FTIR ozone data (Figure 3(b)). ~~We compare the homogenised post-2000 ozonesonde data to low-resolution ozone profiles derived from Fourier-transform infrared spectroscopy (FTIR) ozone measurements, from which low-resolution vertical profiles are derived (e.g., Vigouroux et al., 2015).~~ Note that the updated FTIR ozone data presented here, obtained using an updated retrieval strategy (García et al., 2022; Björklund et al., 2023), are markedly different from the FTIR data shown ~~in~~by Godin-Beekmann et al. (2022). The negative trends in the lower stratosphere in both the sonde and the FTIR ozone data shown here are noticeably larger in magnitude than the trends in the satellite data shown ~~in~~by Godin-Beekmann et al. (2022) which have typically insignificant trends of smaller than -2% decade^{-1}.

~~We also examine how seasonal variations in vertical distributions of ozone might have contributed to the overall trends over the observation period. The seasonal ozone anomalies representing austral summer (DJF), autumn (MAM), winter (JJA), and spring (SON) from the homogenised ozonesonde data are shown in Figure 8~~. ~~It shows that the ozone evolution among all seasons is broadly consistent at the selected vertical levels from the surface to 25 km. The surface ozone in all seasons show decreases until the late 1990s before peaking before ∼2010. The seasonal variation is also weak above 5 km showing downward trends in all seasons. Some slight differences between seasons are below 5 km, where the DJF trend is weaker before the mid-1990s. Also the JJA ozone seems quite flat around 5 km whereas the DJF ozone has a sharp drop after 2015; it warrants a further investigation with a longer time series into the future.~~

3.3 Variations and trends explained by ~~regression~~the MLR analysis

~~In order to identify the drivers of ozone variability and trends, we construct a~~ The regression model (eq. 1). ~~The deseasonalised monthly~~was constructed using annual mean ozone anomalies of the homogenised ozonesonde data. The regression is performed for each level from the surface to 30 km at a 1 km resolution, and the linear trend of the predicted ozone at each level was then calculated. Figure 4 shows that the vertically resolved ozone trends from the MLR predicted ozone are quite similar to the simple linear trends in the homogenised ozonesonde data (also shown in Figure 3), but the uncertainty in the MLR predicted ozone trends are generally smaller than those in the observed trend. The trends in the MLR predicted ozone are also systematically smaller in magnitude in the stratosphere above ∼18 km for both periods but the difference is slightly larger for the post-2000 period, within the uncertainty ranges of the observed trends. It indicates that the regressors used in the MLR model do not fully capture the observed trend there. Despite the slight differences in magnitude, trends calculated here and by Godin-Beekmann et al. (2022) point to that the significant negative trends in the lower stratosphere exist in Lauder ozonesonde data, and these negative trends are underestimated by the satellite products.

We then group the vertical ozone profile into eight layers from the surface to 25 km, for identifying the drivers of ozone variability and trends for each vertical layer. The same MLR is performed individually for each layer. ~~The independent regressors used in the regression are shown in Figure 5. and~~ The observed and regressed ozone anomalies, together with the leading contributions from individual regressors, are shown in Figure 5. The ozone variance explained by the regression is given by the multiple regression coefficient of determination, R^2 (Figure 5 and Table 3). The standardised individual regression coefficients for each regressor can be used to measure their contributions to the total variance explained at that level (Table 3). ~~The leading contributions from individual terms to overall regressed ozone variations are demonstrated in Figure 9.~~

The regression model matches the observed anomalies well, in particular in the stratosphere, the upper troposphere, and near the surface (Figure 5 and Table 3). With R^2 values ranging from ~~0.27 to 0.49~~0.28 to 0.61 in the troposphere and ~~0.50 to 0.73~~0.57 to 0.71 in the stratosphere. ~~implying that~~ the stratospheric ozone variations and trends are better ~~explained~~captured by the MLR model than tropospheric features. Indeed, interannual variations in ozone anomalies in the upper troposphere and the lower stratosphere (9-15 km) are especially well explained by variations in tropopause height (Figure 5 and Table 3). The downward trend in the stratospheric ozone between 9 and 20 km is clearly ~~explained~~driven by the significant negative linear trend ~~that represents all quasi-linear~~in tropopause height (Figure 6(c)). ~~whereas the decreasing stratospheric temperature explains some of the negative trend above 20 km.~~ The QBO at 30 hPa also explains a large part of the ozone variability for the layer 15-20 km, together with tropopause height (Figure 5 and Table 3). Above 20 km, the QBO at both 30hPa and 10 hPa, the AOD, and the stratospheric temperature (T_{Strat}) ~~together with the tropopause height anomaly explain the ozone variation there,~~ that has a significantly negative trend (Figure 6(b))~~linear~~. We note that the correlation between AOD and the Lauder stratospheric ozone is positive, e.g., after the Mt Pinatubo eruption in 1991, despite the potential ozone depletion in the years following a volcanic eruption (~~Figures 8 and~~Figure 5(h)). This lack of ozone depletion was attributed to the perturbation of the stratospheric dynamics by the Mt Punatubo eruption that obscured the chemical effect in the southern extra-tropics (Aquila et al., 2013). We also note that the prolonged decline in ozone above 15 km at the end of the time series (around 2020) can not be explained by the MLR model (Figure 5(g) and (h)); this might be the result of the Australian bush fires in January 2020 which depleted stratospheric ozone (Salawitch and McBride, 2022). The lack of this process in MLR might also explain the weaker negative trend in the regressed ozone than that in the observed ozone above 18 km (Figure 4(b)). ~~monotonic drivers of change (Figures 8 and 9). Note that the tropopause height anomalies have been de-trended for use in regression here, therefore do not explain the negative trends in ozone. No significant trends exist in other regressors either.~~

It is well established that CO_2 increases influence temperature, humidity, and circulation, which in turn affect ozone chemistry and transport (Brasseur and Hitchman, 1988; Butchart et al., 2006; Fleming et al., 2011). Warming in the troposphere and ~~the~~ cooling in the stratosphere due to the increase in CO_2 drives the increase in tropopause height over the last several decades, based on radiosonde observations, ~~the~~ reanalysis data, and modelling (Highwood et al., 2000; Seidel et al., 2001; Seidel and Randel, 2006; Santer et al., 2003b, a; Meng et al., 2021). The tropopause height derived from the Lauder sonde data shows a significant positive trend of 117±82 m decade^{-1} (at 95% confidence) over the observational period, calculated as the simple liner trend in the annual mean anomaly (Figure 6(c)), which is larger than the trend of ∼50-60 m decade^{-1} in the northern hemisphere (20°N-80°N) over 2001-2020 based on radiosonde data ~~in a recent study~~ (Meng et al., 2021). ~~If the tropopause anomaly is not de-trended, the correlation coefficients between the tropopause anomaly and the ozone anomaly are highly anti-correlated with a correlation coefficients of −0.94 in the 9-12 km layer and −0.95 in the 12-15 km layer. However, with a de-trended tropopause height, the correlation coefficients are −0.71 and −0.73 respectively. This indicates that the negative contribution to the ozone trend in the lower stratosphere (between ∼9 to 15 km) can largely be projected on the significant increase in tropopause height.~~

~~The observed tropopause height anomalies at Lauder are also closely correlated with tropospheric temperature (with a correlation coefficient of 0.74) and anti-correlated with stratospheric temperature (-0.76), with significant positive and negative trends respectively (Figure 6), which are mainly driven by the CO_2 increase (Mitchell et al., 1995; Santer et al., 1996). Here, the use of tropopause height as a regressor accounts for the overall dynamical changes, while~~

excluding the effect of inter-dependence of the changes in tropopause height and temperature. Therefore the negative linear trend term accounts for the overall linear effects including the changes in both the stratospheric and tropospheric temperatures.

However, in the middle and upper troposphere (6-9 km), the regression function explains the least ozone variations compared to those at levels above and below (Figure 5(d)). Here, although the solar influence is the strongest in relative terms, influences from all other regressors, except the QBO at 10 hPa, contribute non-negligibly to explaining the ozone variations at this level.

In the lower and free troposphere (below 6 km), the sharp decreases in ozone during the early period of the record and the large negative anomalies in 1997/1998 are well reproduced by the regression, as well as the subsequent increases in ozone there. But the large year-to-year variability is less well captured in the free troposphere. This trend transition in tropospheric ozone coincides with the evolution of EESCs which increases since the late 1980s before declining after 1997; this indicates that the impact of stratospheric ozone changes due to changes in ODSs could impact tropospheric ozone through transport. Indeed, the interannual variation in the free tropospheric ozone is shown to be influenced by the QBO at 30 hPa (Figure 5(b) and (c)). This trend transition follows the evolution of EESC, which after a peak in 1997 has been declining since then (Figure 7), and indicates the stratospheric impact on the tropospheric ozone through stratosphere-to-troposphere transport reflecting the effect of stratospheric ozone depletion and recovery. In the troposphere, increases in ODSs are expected to drive an increase in ozone after the late 1990s, whilst the response to the CO_2 increase in the troposphere is more complex. For exampleinstance, the associated increasing humidity would lead to more chemical destruction of ozone in the troposphere, and the increase in temperature may result in more ozone production through NO_x-CH_4 (and volatile organic compounds) chemistry (e.g., Stevenson et al., 2006; Zeng et al., 2008). Here, relative humidity and surface ozone are anti-correlated (Figure 5(a)). Relative humidity has a large negative impact on surface ozone (Table 3). Moreover, we have not considered changes in ozone precursor concentrations and other meteorological parameters in the regression that could substantially impact tropospheric ozone. Therefore, more explanatory variables can be included in a MLR model that is specifically focused on tropospheric ozone; this is subjected to a future study.Indeed, for instance, the continuing downward trend in surface ozone after ∼2003 cannot be explained by the reduction in ODSs. The regression function we construct here is more suitable to explain the stratospheric ozone changes.

3.4 Attribution of modelled Lauder ozone changes to ODSs and GHGs

We examine the modelled vertically resolved ozone trends in the vicinity of Lauder (160-180°E and 40-50°S) from the NIWA-UKCA model over the ozone depletion (1987-1999) and recovery (2000-2020) periods separately, as well as the effects of changes in ozone trends due to individual single forcings, the same approach as in Zeng et al. (2022). Meanwhile, in order to help understand Lauder ozone changes in a global context, we also show the modelled zonal mean ozone trends covering all latitude bands, and the changes that are attributable to ODSs and GHGs in Appendix B (Figures B2 and B3 in Appendix B). The modelled and attributable trends are linear trends in diagnosed annual mean ozone anomalies calculated from model simulations.

3.4.1 Pre-1999 period

Over the ozone depletion (pre-1999) period, the modelled ozone trends at Lauder (Figure 7(a)) are significantly negative (at the 95% confidence level) throughout the height range covered by the sondes, and the magnitude maximizes at $\sim -5\%$/decade at around ∼14 km. The modelled trend over this period are ~~broadly~~qualitatively in agreement with the Lauder observations, although it generally underestimates the observed trends in magnitude, especially in the lower stratosphere (∼10-13 km) where the observed negative ozone trend is much larger at $\sim -12\%$ (Figures 4(a) and 7(c)).

The modelled Lauder ozone trends over this period are attributable to increases in ODS, methane, N_2O, and CO_2 (Figure 7(a); the uncertainties of these contributions are displayed separately in Figure B1). The increase in ODSs contributes significantly to the negative ozone trend in the lower stratosphere (∼9-18 km), which is the result of ozone depletion at SH mid-latitudes (Figure B2(c)). ~~The N_2O increase also contributes moderately to the negative ozone trends between ∼13 km over Lauder but the effect is not statistically significant at 95% confidence (Figure B1). In contrast, the N_2O increase leads to ozone increase in the upper troposphere (5-13 km) as a result of the self-healing effect which was explained by Morgenstern et al. (2018) using the same set of model simulations as used here.~~ The increase in methane during this period (1987-1999) has a considerable positive impact on ozone trend over Lauder ~~below 25 km~~ which maximises at around 12 km (Figure 7(b)) and is statistically significant at the 95% confidence level below 15 km (Figure B1). The ozone increase caused by the growth of methane is partly due to its reaction with chlorine which leads to reduced ozone depletion especially in the stratospheric polar region, and partly through chemical ozone production in the troposphere (Figure B2(d)). The N_2O increase also contributes moderately to the negative ozone trends above ∼13 km over Lauder but the effect is not statistically significant at 95% confidence (Figure B1). In contrast, the N_2O increase leads to ozone increase in the upper troposphere (5-13 km) as a result of the self-healing effect which was explained by Morgenstern et al. (2018). ~~using the same set of model simulations as used here.~~ The increasing CO_2 (derived from the all-GHG forcing and the separate methane and N_2O forcing experiments) has a relatively large negative contribution to ozone over Lauder below 20 km which maximises at a lower altitude of around 10-12 km, ~~but only~~and is statistically significant at the 95% confidence level below 13 km (Figures 7(f) and B1). It shows that the impacts of dynamical changes and the ozone depletion on stratospheric ozone occur at different altitudes.

We examine contributions of ODSs and CO_2-driven dynamical changes to ozone changes from the MLR model, and compare those to the modelled attribution. The CO_2-driven tropopause increase (Figure 7(c) exhibits a large contribution to the MLR ozone trend between ∼9 and 22 km, maximising at ∼10-12 km, which is consistent with the model attribution (Figure 7)(a)). The impact of stratospheric cooling (reflected in T_{Strat}) shows a small negative impact on the MLR ozone trend over this period. The contribution of ODS to the regressed negative ozone is most pronounced above 23 km and below ∼17 km, again consistent with the modelled attribution. However, the impact of ODSs on stratospheric ozone trend shown here is not reflected in the small and insignificant regression coefficient due to EESC in the stratosphere (Table 3); most likely, the impact of EESC is obscured by the more prominent impact of CO_2-driven dynamical changes throughout the whole observational period, not just for the pre-1999 period. The post-2000 period would see the impact of EESC dropping considerably, explaining the small regression coefficient. We also see that the tropospheric ozone trend in the MLR model is mostly attributable to ODS changes (Figure 7(c), likely the result of stratospheric polar ozone changes through transport (Figure B2(c).

3.4.2 Post-2000 period

The stratospheric equivalent chlorine ~~attained~~reached its maximum in the late 1990s and has been declining since. Consequently, over the period of 2000-2020, the model shows a small but largely significant positive ozone trend of up to 1% decade^{-1} above $\sim$23 km in the stratosphere (Figure 7(b)). There is no significant trend in modelled ozone below 23 km, except for a small negative trend near the surface. ~~In the lower stratosphere, however, a small negative ozone trend of less than 2% in magnitude occurs between 15 and 25 km, and the trend is statistically significant between 17 and 22 km. This simulated negative trend is about half in magnitude compared to the observed trend at Lauder which covers a larger vertical domain from 8 km to 30 km (Figure 4). In the troposphere below 8 km, the modelled and the observed trends are both positive which are up to $\sim$2% decade^{-1} in magnitude.~~ Clearly, the model cannot reproduce the significant negative trend in the lower stratosphere exhibited by observed ozone and underestimates observed trends at all levels in this period. (Figure 4(b) and 7(d)). In a recent assessment, combined satellite datasets indicate a negative trend over the period of 2000-2020 in the SH mid-latitude (35-60°S) of the lower stratosphere, but multi-model results generally show non-significant positive trends (Godin-Beekmann et al., 2022; World Meteorological Organization (WMO), 2022), which is typically associated with a large dynamical variability in this region.

Over this period, the effects of ODSs, methane, and N_2O on modelled ozone ~~above 15 km~~ are generally small (Figure 7(b)) and statistically insignificant at the 95% confidence level (Figure B1). ~~but they become slightly larger and sometimes significantly positive.~~ The impact of ODSs is consistently positive from the surface to about 23 km, as the result of ODSs declining. The impacts from methane is a small positive contribution to the modelled ozone trends, whilst the N_2O mainly contributes negatively above $\sim$13 km and positively below (Figure 7(b)). In contrast, the impact of CO_2 on ozone at Lauder ~~are~~is negative between $\sim$5$-$22 km ~~which~~and maximises at $\sim$12 km with a contribution of $-$4% decade^{-1} (Figures 7(b)). Although the impact of the CO_2 increase is much larger than those from other forcings during this period, the trend is not statistically significant (Figure B1), possibly due to the typically large dynamical variation in the UTLS region. However, on a global scale, the impact of ODSs and CO_2 on ozone trends in the UTLS region can be significant at the SH mid-latitudes (Figure B3). ~~With declining ODSs, CO_2 plays a dominant role in driving ozone trends in the future.~~ The modelled results here are consistent with previous findings on the response of global ozone changes to ODSs and GHGs using either the CCMI-1 (Morgenstern et al., 2018) or Aerosol and Chemistry Model Intercomparison Project (AerChemMIP) simulations (Zeng et al., 2022).

The attribution of MLR ozone trends shows that the impact of CO_2, reflected in the change in tropopause height, is an important driver for the observed negative ozone trends at Lauder in the UTLS region after 2000 while the ODSs have been declining (Figure 7(d)). It also shows that continuous stratospheric cooling, driven by the CO_2 increase, plays a increasingly important role in contributing to the negative stratospheric ozone trend above 15 km observed at Lauder since 2000 (Figure 7(d)). The role of ODSs during this period is consistent with the modelled attribution which is largely positive but small. ~~The result here is consistent with previous findings on the response of global ozone changes to ODSs and GHGs using either the CCMI-1 (Morgenstern et al., 2018) or Aerosol and Chemistry Model Intercomparison Project (AerChemMIP) simulations (Zeng et al., 2022).~~

4 Conclusions

We have updated the Lauder ozonesonde timeseries by homogenising the dataset with a series of well-defined correction steps accounting for changes in hardware and operating procedure. We have analyzed this homogenised dataset for height vertically resolved linear ozone trends over the 1987-1999 and 2000-2020 periods, characterised by increasing and decreasing trends of total chlorine and bromine, respectively. There are significant differences between the homogenised and the uncorrected data for the pre-1999 period due to these corrections, in which the uncorrected data are low-biased compared to the homogenised data in general. This leads to significantly stronger negative stratospheric ozone trends in the homogenised data compared to the uncorrected data over the 1987-1999 period. The homogenised data typically show negative ozone trends of ~ -6 to -2% decade^{-1} from the surface to 30 km with a maximum of $\sim -13\%$ decade^{-1} around 13 km, substantially stronger than trends in uncorrected data which are largely insignificant. with significant trends at the 95% confidence above 12 and below 5 km. Trends in both these altitude regions are substantially stronger than trends in uncorrected data which are largely insignificant. For the post-2000 period, the homogenisation does not alter ozone trends significantly; both datasets show significant negative trends in the stratosphere up to $\sim -6\%$ decade^{-1} and small positive trends of up to +2-3% decade^{-1} in the troposphere. The post-2000 trends in ozonesonde data are in excellent agreement with trends in co-located FTIR ozone profiles.

In addition, we calculated linear trends in the MLR predicted ozone for the two periods, which show a very good agreement with the observed linear trends, except for the region above 18 km where the MLR trend is slightly smaller in magnitude in the post-2000 period. The large negative trend in the lower stratosphere is consistent with the trend calculated by Godin-Beekmann et al. (2022) based on data from the LOTUS regression model, although the negative trend by Godin-Beekmann et al. (2022) is insignificantly larger in magnitude. The uncertainty ranges found here comfortably fit within or close to the uncertainty ranges stated by Godin-Beekmann et al. (2022). Differences in the best-estimate trends could be down to the difference between the regression models used.

By using a multiple linear regression analysis we have identified the dominant factors driving the Lauder vertically resolved ozone trends and variations. The regression model consists of independent regressors including solar flux, the state of ENSO, the QBO at two different altitudespressure levels, stratospheric equivalent chlorine, and the aerosol optical depth representing volcanic influences. A linear term accounts for monotonically changing anthropogenic forcings (led by CO_2). Additionally we have included the detrended tropopause height anomaly, representing the dynamical variability that drives the interannual variability in ozone, the stratospheric temperature anomalies that are averaged over 22-30 km to account for the impact of stratospheric cooling induced by the CO_2 increase, and surface relative humidity that reflects the effect of humidity on near surface ozone, as regressors. We find a persistent negative stratospheric ozone trend at Lauder represented by the significant negative trends in the linear termthe tropopause height and the stratospheric temperature of the regression function. The variation in tropopause height, which anti-correlates with stratospheric but correlates with tropospheric temperature, largely explains the interannual variations in upper tropospheric and lower stratospheric ozone. Significant trends in the tropopause height (positive), the stratospheric temperature (negative), and the tropospheric temperature (positive) measured at Lauder are consistent with the well-established impact of stratospheric circulation changes driven by CO_2 increases (e.g., Mitchell et al., 1995; Butchart et al., 2006). The QBO and AOD indices explain much of the stratospheric ozone variations above 20 km and the stratospheric temperature partially explains the significant negative trend at and above this altitude. In the troposphere, the interannual variations and trends in ozone are less well explained by the regression function in comparison with those in the stratosphere. However, Surface relative humidity explains a substantial amount of surface ozone variability. The impact of ODSs on tropospheric ozone at Lauder is demonstrated by ~~correlate with~~ the downward and upwards trends in tropospheric ozone before the late 1990s and after 2000 coinciding with the increase and decrease in ODSs, ~~were increasing and decreasing,~~ respectively. ~~Surface relative humidity explains a substantial amount of surface ozone variability.~~

We have also used a series of chemistry-climate model single forcing simulations to gauge the impact of changes in GHGs, including methane, N_2O, and indirectly CO_2, and ODSs on ozone profiles at Lauder, as well as on the zonal mean ozone profiles covering all latitude bands in a global context. For 1987-1999, simulations show significant negative ozone trends throughout the vertical domain (up to 30 km), broadly in agreement with observed ozone trends at Lauder during this period, except for in the lower stratosphere where the modelled ozone trend is substantially smaller in magnitude than the observed negative trend. Fixed Single forcing simulations attribute the negative ozone trend to ODS-driven ozone depletion in the SH mid-latitudes and ~~increases in CO_2 which lead~~to the increase in CO_2 which leads to changes in stratospheric circulation and temperature that impact ozone. However this negative impact on ozone is offset by the positive impact of methane. N_2O plays a smaller role with both negative impacts on ozone above ~ 13 km and positive ones below that level. Note that, although the MLR coefficient representing the impact of ODSs on stratospheric ozone is small and insignificant for the whole analysis period, the impact of ODSs on stratospheric ozone is apparent from both the modelled and MLR trend attributions over the 1987-1999 period. The impact of ODSs on tropospheric ozone appears to be affected by the polar stratospheric ozone through transport.

Over the period of 2000-2020, although the model ~~underestimates~~can not capture the observed significant negative ozone trend in the upper troposphere and lower stratosphere over Lauder, it points to a significant negative impact of the CO_2 increase on ozone in this region, offset by much smaller positive impacts from the reduction in ODSs and increases in methane and N_2O. This modelled negative impact from CO_2 on ozone through dynamical changes is reflected in the observed tropopause height increase at Lauder, and this impact will grow if CO_2 is continuously increasing in the future. Therefore, long-term vertically resolved monitoring of ozone is of particular importance to understanding the impact of climate change on the ozone distribution and vice versa.

Data availability. The "uncorrected" Lauder ozonesonde data can be accessed at the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) archive (https://woudc.org/data/explore.php) and at the Network for the Detection of Atmospheric Composition Change (NDACC) archive (https://www-air.larc.nasa.gov/missions/ndacc/data.html). The homogenised Lauder ozonesonde data can be obtained from the TOAR-II HEGIFTOM Focus Working Group (https://hegiftom.meteo.be/datasets). ~~The Stratospheric Aerosol Optical Depth data was obtained from the NASA Langley Research Center Atmospheric Science Data Center (https://asdc.larc.nasa.gov/).~~

Appendix A: Homogenisation of Lauder ozonesonde time series

The corrections that are applied to the Lauder Ozonesonde time series are detailed below. All the corrections are applied on the raw ozone currents. When these cell current were not archived in the early period, they need to be reconstructed from the ozone partial pressure data in the NDACC archive with the available metadata (e.g. pump flow rate, pump temperature, background current, pump efficiency correction table used). Then, correction functions are applied according to those recommended in Smit and the O3S-DQA panel (2012). The effect from each correction is shown in Figure A1.

A1 Conversion efficiency

The stoichiometry correction was applied for the 1986 data where 2.5 ml instead of 3 ml of cathode solution was used. The EnSci sondes with a 1.0% buffer solution strength over the period of 1994-1996, instead of a 0.5% strength, were also corrected.

A2 Background current

A consistent background current correction was applied to the Lauder data. If the background current values fall above the mean value + 2 standard deviation (σ), these values are replaced by the mean value. The mean and corresponding standard deviations are calculated and applied separately in two periods (i.e., before and after 1996), as the background current values are systematically larger for the period before 1996 and smaller for the period after 1996.

A3 Pump temperature measurement

Truest pump temperature correction is applied according to Eq. 13 of the O3S-DQA Guidelines (Smit and the O3S-DQA panel, 2012). SPC-4A sondes (until 1989), SPC-5A (from 1989 to 1994), and EnSci sondes (from 1994) were launched in the configuration where the pump temperature measurement was made inside the pump. However, the SPC-4A and SPC-5A pump temperature measurements need additional corrections (see Smit and the O3S-DQA panel, 2012).

A4 Pump flow rate (moistening effect)

Eq. 15 of the O3S-DQA Guidelines was applied to correct the moistening effect of the pump flow rate. There are missing metadata including temperature and humidity of the laboratory before February 2014. The climatological means calculated for each month are then used for these missing metadata.

A5 Pump flow efficiency

Eq.22 of the O3S-DQA Guidelines (Smit and the O3S-DQA panel, 2012) was applied using the Pump flow correction factors (CPF) as a function of air pressure (Table 6 of this guideline). These are also applied on the "uncorrected data", as a part of the conversion from the ozone currents to ozone partial pressures. The small change in these correction factors around 1994 is due to the fact that different correction factors need to be applied for SPC and En-Sci ozonesonde pumps.

Appendix B: Supplementary figures and table

Figure B1 displays the modelled Lauder ozone trend changes due to ODSs, methane, N_2O, and CO_2 including the 2σ uncertainty range. Figures B2 and B3 display the modelled zonal mean ozone trends and the impacts from ODS, combined GHGs, methane, N_2O, and derived CO_2 for the periods of 1987-1999 and 2000-2020, respectively. Table **??** contains the coefficient of determination and the regression coefficients from the multiple linear regression analysis.

Author contributions. RQ, HS, AG, PS carried out Lauder ozonesonde measurements and processed the data. RVM and DP helped with homogenisation of Lauder ozonesonde data, DS and JR provided FTIR ozone time series, GZ and OM performed model simulations and conducted the statistical analysis. GZ led the writing of the paper with inputs from all authors.

Competing interests. The authors declare that they have no competing interests.

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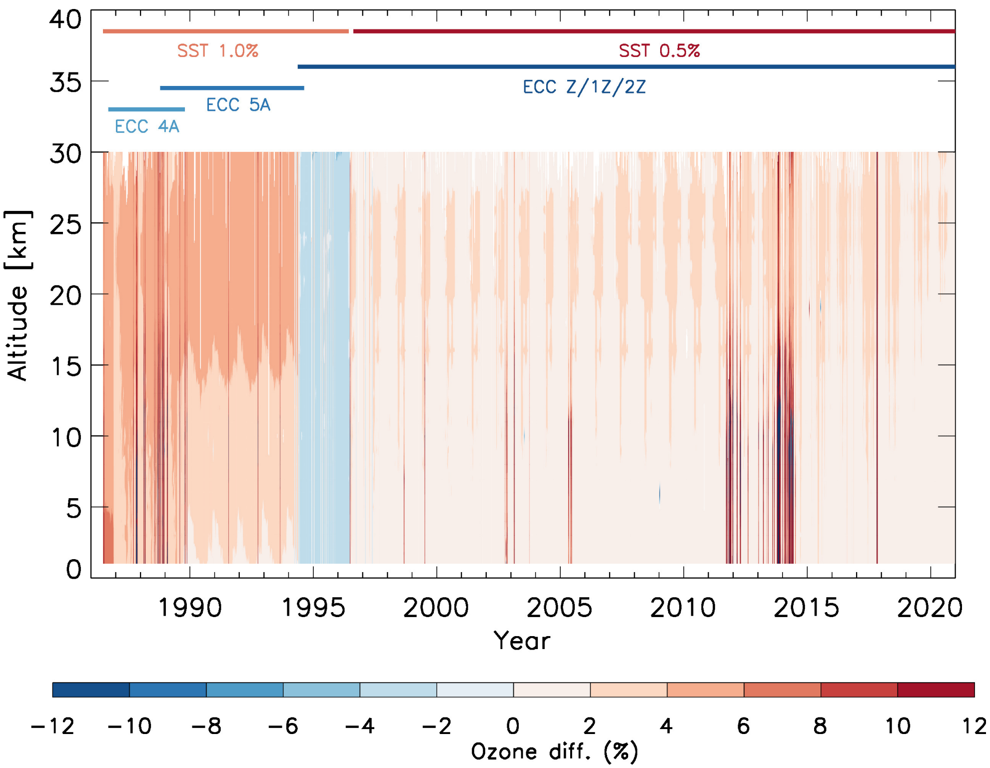

Figure 1. Comparison of ozonesonde timeseries before and after homogenisation over 1987-2020 for all flights, in percentage difference between "homogenised" data and "uncorrected" data (i.e., $100 \times (homogenised - uncorrected)/uncorrected$). Also shown are periods indicating changes in the ozonesonde type and the solution used.

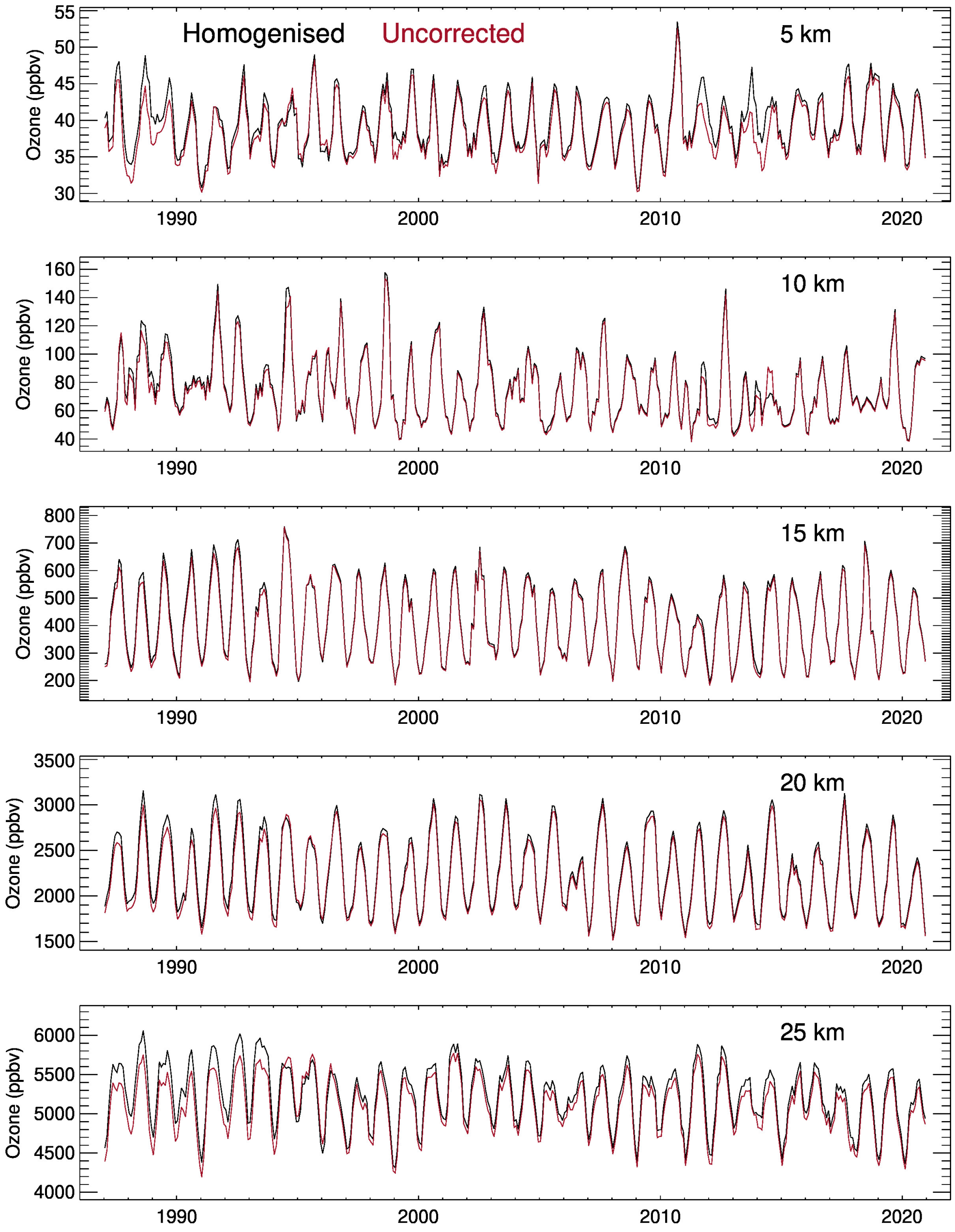

Figure 2. The homogenised and the uncorrected monthly mean ozone values (ppbv) for different vertical layers over 1987-2020. For displaying purposes, the monthly data are smoothed with a ~~3 box-car~~3-month boxcar filter.

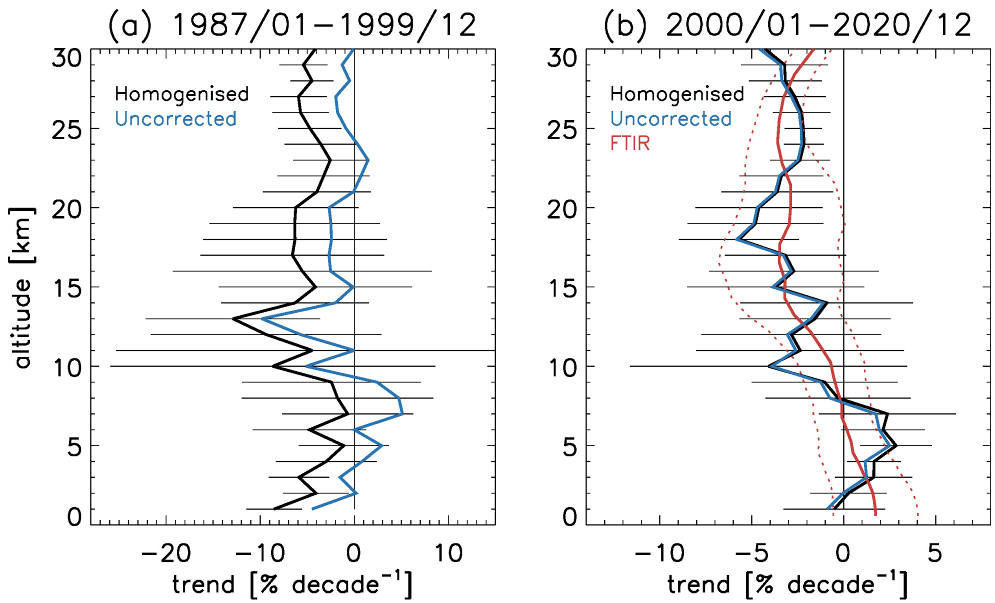

Figure 3. Vertically resolved observed linear trends in monthly mean ozone and their uncertainties ($\pm 2\sigma$) at Lauder over two periods, i.e., 1987-1999 (a) and 2000-2020 (b), from ozonesonde measurements (black: homogenised data; blue: uncorrected data), and from FTIR measurements (red, for the 2001-2021 period). Note the slightly different time period for the FTIR ozone data.

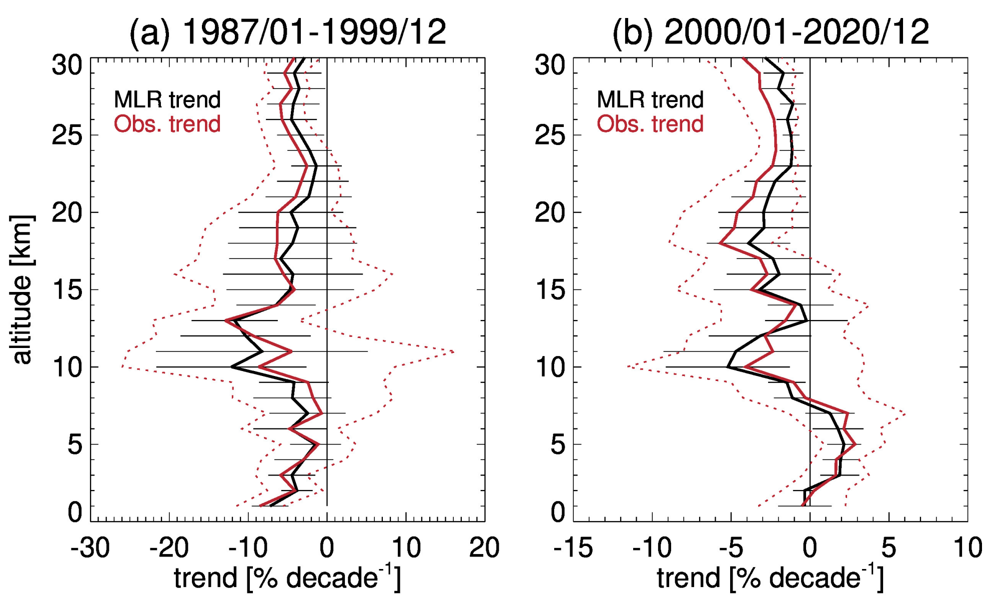

Figure 4. Vertically resolved linear trends in regressed ozone (black) and in the homogenised observed ozone (red) and their uncertainties ($\pm 2\sigma$) at Lauder over two periods, i.e., 1987-1999 (a) and 2000-2020 (b). Data used for linear trend calculations in both cases are annual mean anomalies.

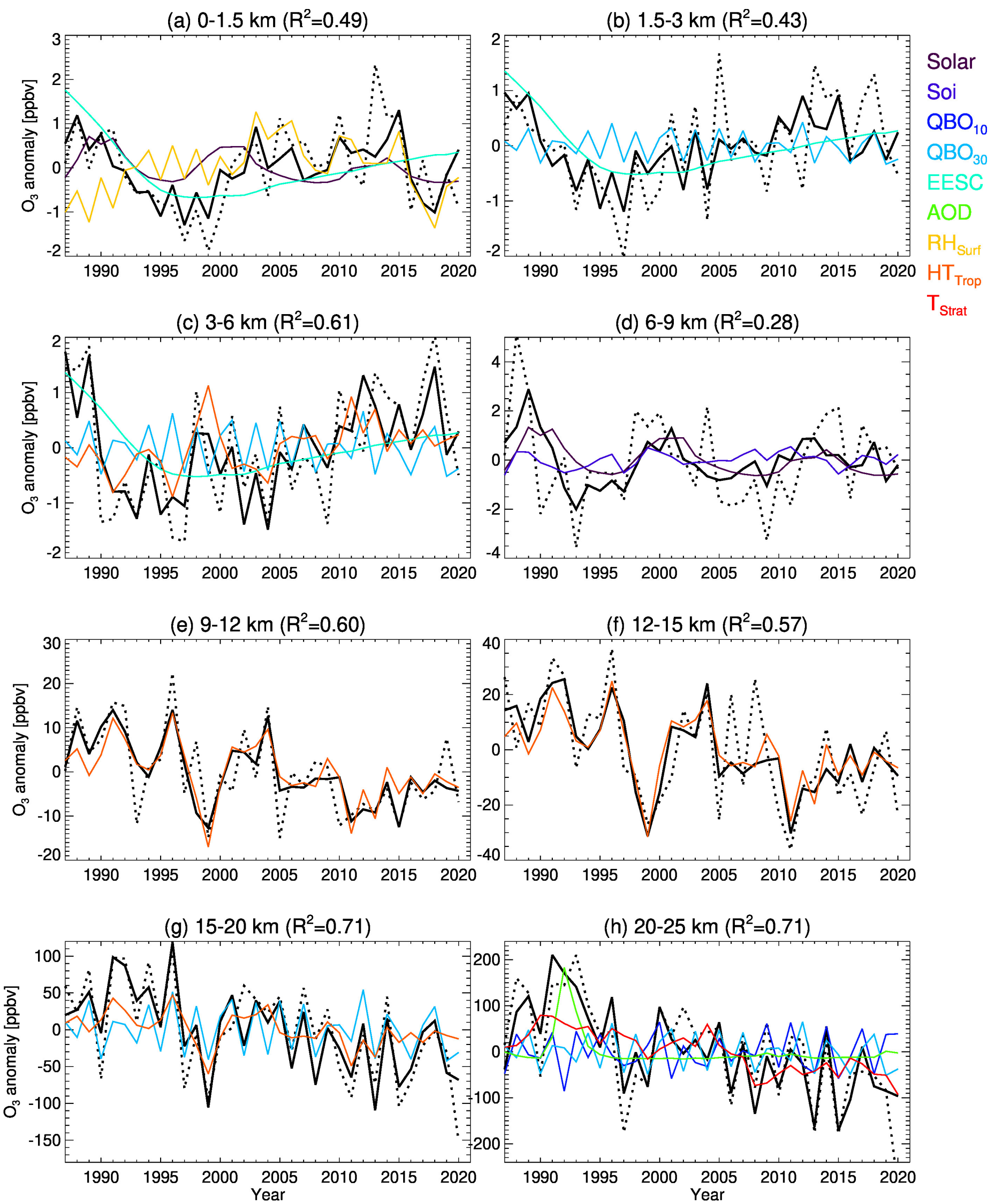

Figure 5. Regressed ozone anomalies (black curves) and Observed ozone anomalies (black dotted curves, homogenised data) at Lauder (1987-2021) for eight vertically averaged layers. Contributions from Leading regressors for each layer are displayed in coloured curves (colour keys in the right of the plot.

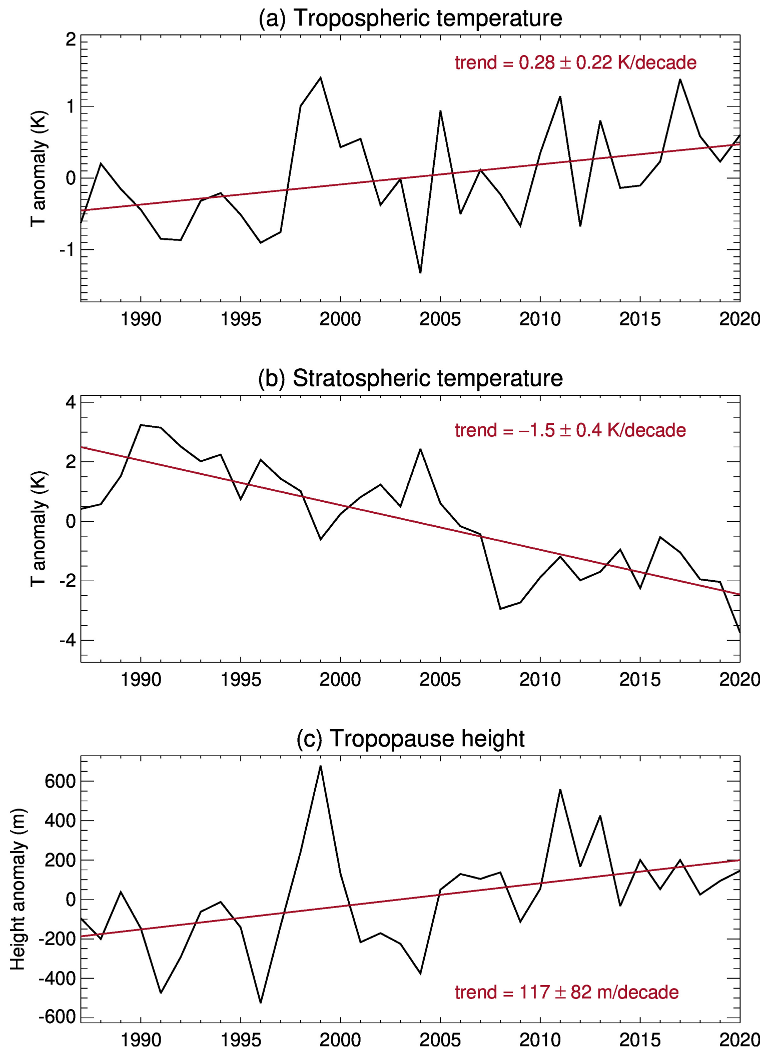

Figure 6. Annual mean anomalies of Observed tropospheric temperature (averaged below 5 km) (a), stratospheric temperature (averaged between 22 km and 30 km) (b) and the tropopause height (c), and their linear trends ($\pm2\sigma$) at Lauder. ~~Monthly mean time series are smoothed using a 12-boxcar filter for displaying purposes. Linear trends $\pm2\sigma$ shown in the plot are calculated based on monthly mean data without smoothing.~~

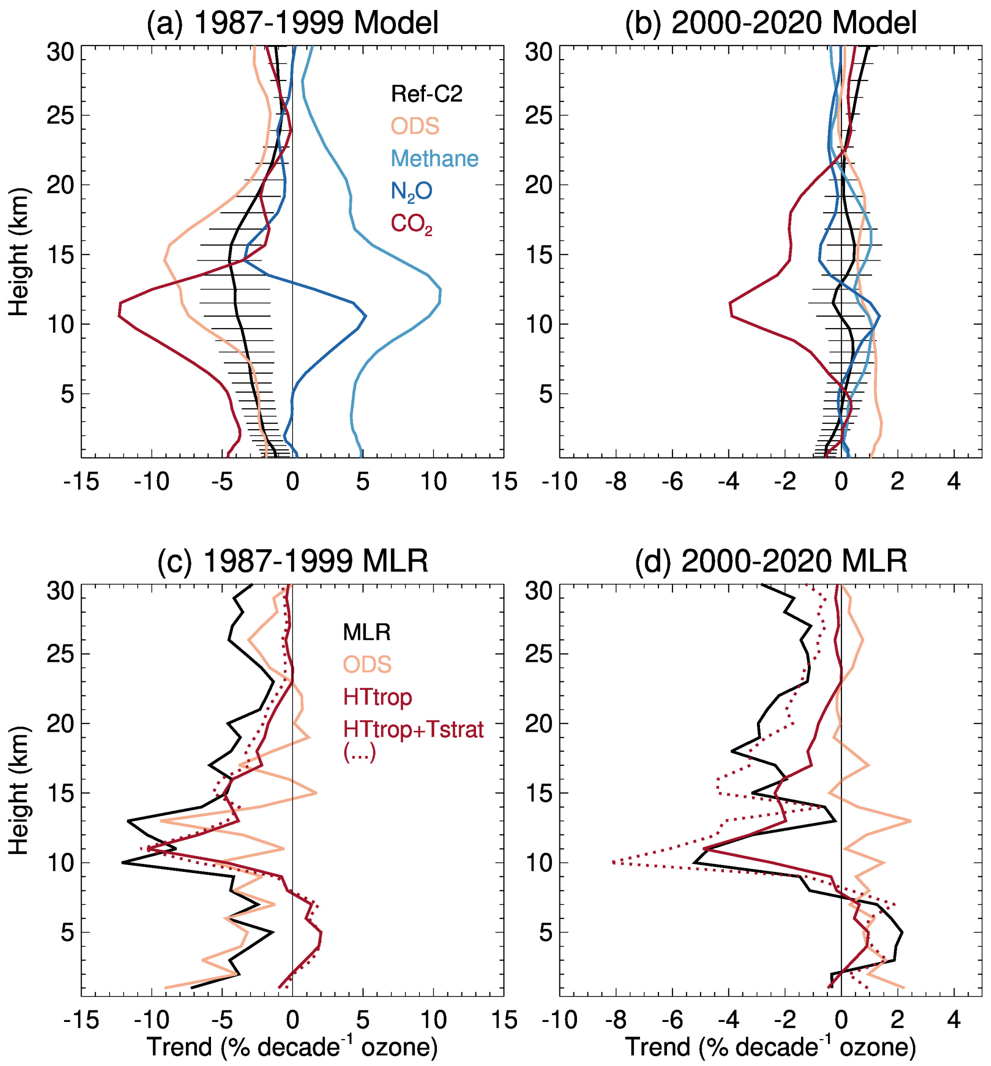

Figure 7. Upper panel: Vertically resolved trends in modelled annual mean ozone anomalies averaged in the area of 160°E-180°E and 40°S-50°S (representing the location of Lauder) for the periods of 1987-1999 (a) and 2000-2020 (b) from the NIWA-UKCA CCMI RefC2 simulation (Ref-C2) with the 2σ uncertainty range (black), and the trend changes due to changes in the ozone depleting substances (ODSs), methane, nitrous oxide (N_2O), and CO_2 over the same period. Lower panel: Vertically resolved ozone trends in the predicted ozone (at 1 km resolution) by the multiple linear regression (black) and the contribution from EESC (orange) , the tropopause height (red), and the combination of the tropopause height and the stratospheric temperature changes (dotted red), for the periods 1987-1999 (c) and 2000-2020 (d).

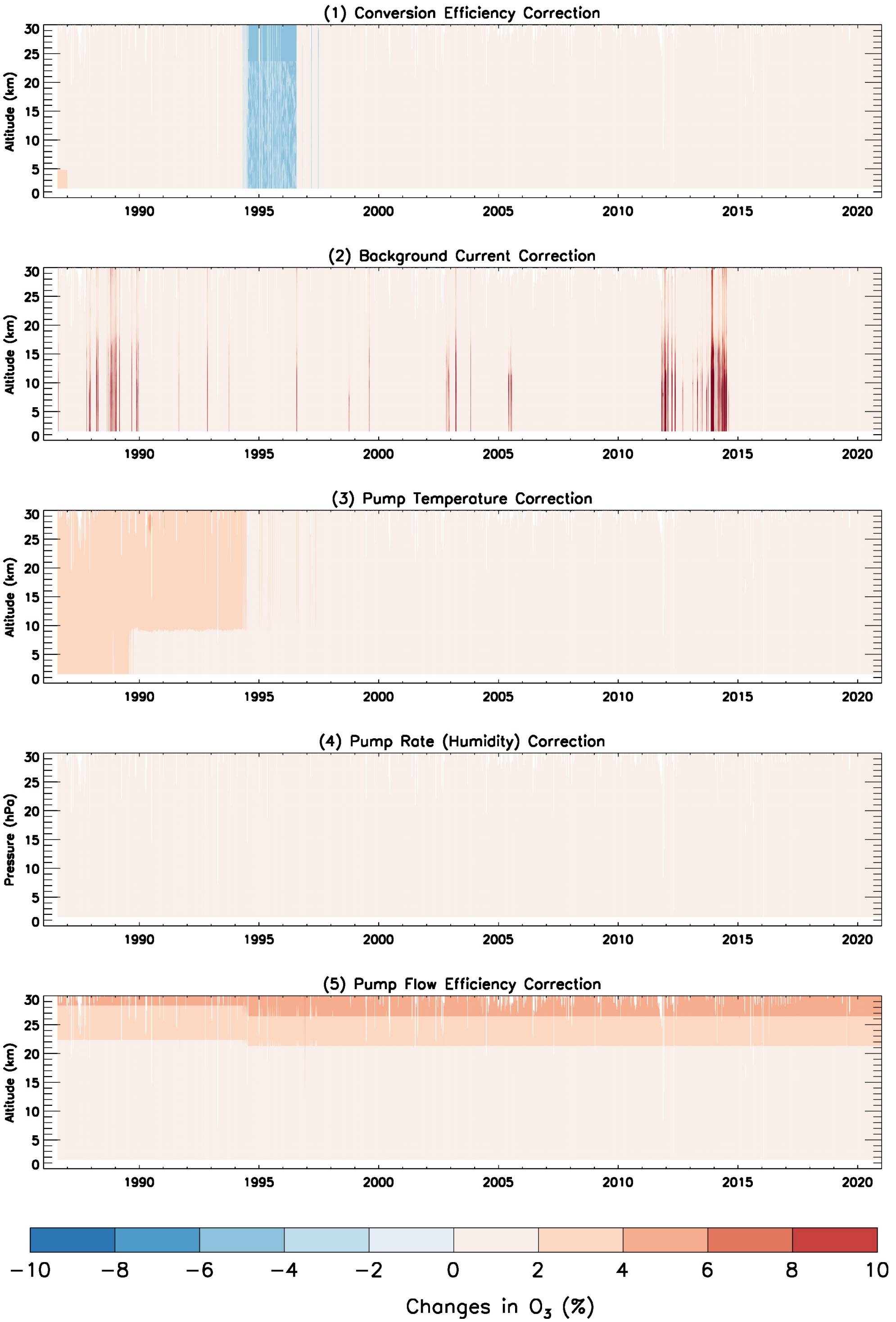

Figure A1. Effect of various corrections on Lauder ozonesonde time series, expressed in percentage changes in ozone.

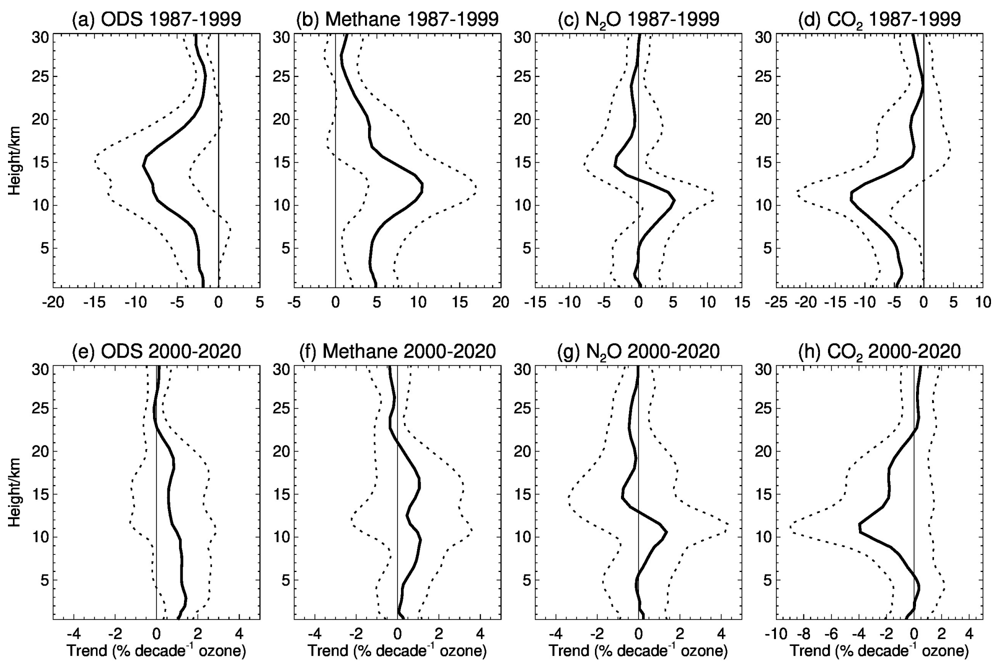

Figure B1. Ozone trend changes due to changes in the ozone depleting substances (ODS), methane, nitrous oxide (N_2O), and CO_2 for the periods of 1987-1999 (up) and 2000-2020 (bottom) simulated in NIWA-UKCA CCMI simulations. The 2σ uncertainty range is marked by dotted lines. Trends in ozone are averaged in the area of 160°E-180°E and 40°S-50°S (representing the location of Lauder).

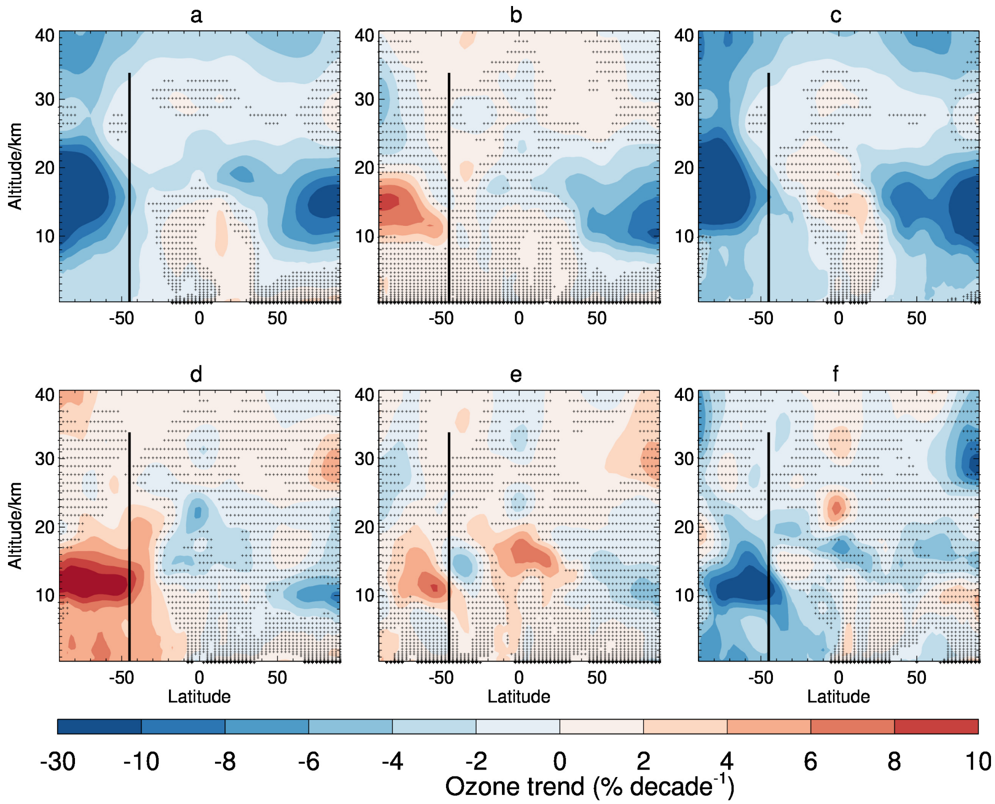

Figure B2. Trends in zonal mean ozone between 1987 and 1999 from the NIWA-UKCA CCMI RefC2 simulation (a), and the change in zonal mean ozone trend due to changes in b) greenhouse gases (GHGs), c) ozone depleting substances (EESC), d) methane (CH_4), e) nitrous oxide (N_2O), and f) CO_2 over the same period. Black vertical lines indicate the latitude of Lauder Station.

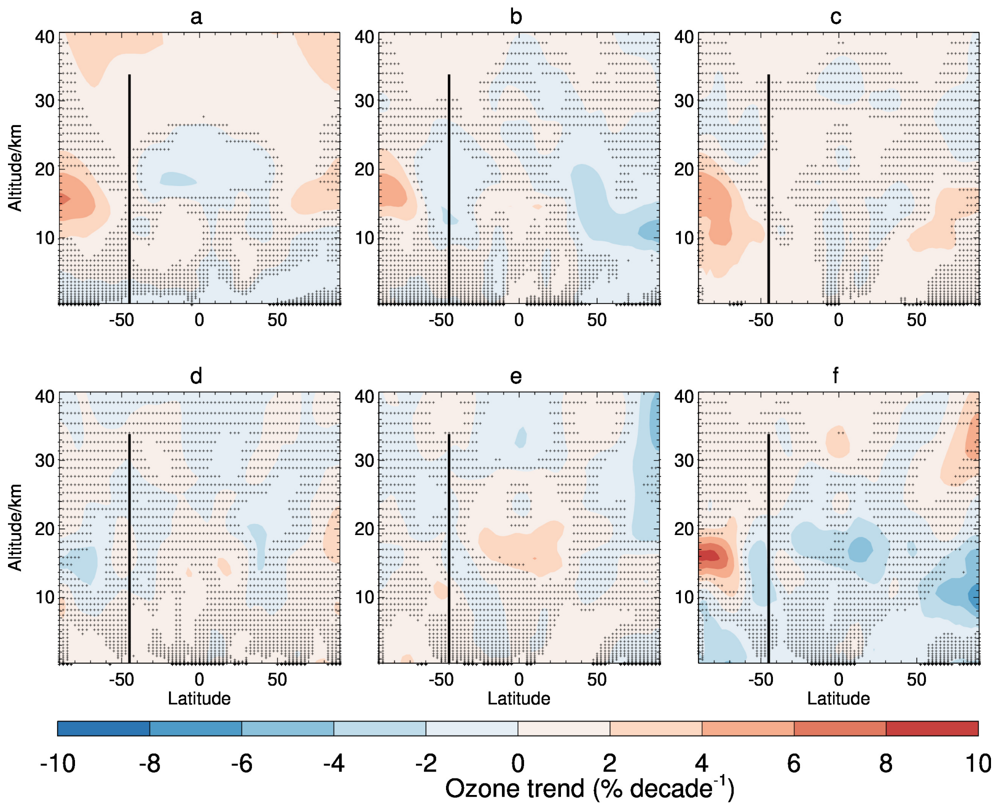

Figure B3. As Figure B2, but for the period of 2000-2020.

Table 1. Changes in ozonesonde types and solutions.

Ozonesonde type changes at Lauder		
Science Pump	ECC 4A	August 1986 to October 1989
(4A/5A/6A)	ECC 5A	1988 (3), August 1989 to 1995, 1996 (2), 1997 (2)
	ECC 6A	1997 (2)
EnSci	ECC 1Z	May 1994 to 2016
(1Z/2Z/Z)	ECC 2Z	2000 (1), 2001 to present
	ECC Z	2007 (2), 2008 - present
Sensing solution changes at Lauder		
KI 1.0% SST		August 1986 to Jul 1996 (incl. 3 dual flights for comparison)
KI 0.5% SST		August 1996 to present
Note: 2.5 ml instead of 3 ml of cathode solution was used in 1986. 1.5 ml of anode solution is always used.		

Numbers in brackets indicate the numbers of flights in these conditions.

Table 2. Forcings for regression model.

Variable	Description	Source
$Solar(t)$	Monthly mean 10.7 cm solar flux	https://psl.noaa.gov/data/correlation/solar.data
$SOI(t)$	Multivariate ENSO Index Version 2 (MEI.v2)	https://www.psl.noaa.gov/enso/mei
$QBO\_10(t)$	Orthogonalised Singapore winds at 10 hPa	https://acd-ext.gsfc.nasa.gov/Data_services/met/qbo/
$QBO\_30(t)$	and 30 hPa	QBO_Singapore_Uvals_GSFC.txt
$EESC(t)$	Equivalent Effective Stratospheric Chlorine	RCP6.0 Scenario (World Meteorological Organization (WMO), 2011)
AOD	Stratospheric Aerosol Optical Depth	NASA/LARC/SD/ASDC (2022) (https://asdc.larc.nasa.gov/)
$HT_{Trop}(t)$	Tropopause height	WMO lapse rate definition (WMO, 1957)
$T_{Strat}(t)$	Stratospheric temperature	Measured
RH_{surf}(t)	Relative humidity at the surface	Measured

Table 3. Coefficient of determination, R^2, for each altitude band (km), and the standardised regression coefficients $\pm 2\sigma$ (Standard error).

Height	R^2	Standardised Regression Coefficients (a_{1-9})								
(km)		Solar	SOI	QBO10	QBO30	EESC	AOD	HT_{Trop}	T_{Strat}	RH_{surf}
0-1.5	0.49	0.32±0.34	-0.0±0.36	0.02±0.30	-0.0±0.29	**-0.62±0.35**	-0.06±0.31	-0.19±0.37	-0.34±0.42	**-0.63±0.34**
1.5-3	0.42	0.14±0.33	-0.02±0.35	0.14±0.29	-0.25±0.28	**-0.48±0.34**	-0.05±0.30	0.11±0.36	-0.25±0.41	-0.23±0.34
3-6	0.61	-0.01±0.34	-0.03±0.36	0.26±0.30	**-0.39±0.29**	**-0.48±0.35**	-0.17±0.31	**0.43±0.37**	-0.16±0.42	0.13±0.34
6-9	0.28	0.60±0.79	-0.31±0.83	0.36±0.69	-0.11±0.68	-0.41±0.81	-0.35±0.72	0.07±0.85	-0.14±0.97	0.13±0.80
9-12	0.60	-0.18±3.06	-2.75±3.22	0.42±2.70	1.67±2.63	-1.02±3.15	0.50±2.78	**-6.52±3.31**	2.32±3.77	0.41±3.11
12-15	0.57	-1.13±6.40	-1.36±6.73	3.02±5.63	3.01±5.49	-2.46±6.58	0.84±5.80	**-12.1±6.9**	3.13±7.87	0.93±6.49
15-20	0.71	-5.62±17.8	-4.27±18.7	-8.15±15.7	**-32.0±15.3**	-1.12±18.3	11.1±16.2	**-23.0±19.3**	20.6±21.9	4.85±18.1
20-25	0.71	11.5±30.5	-16.3±32.1	**-40.7±26.9**	**-38.5±26.2**	-7.3±31.4	**37.5±27.7**	-15.5±33.0	**45.7±37.5**	-0.14±30.9