

Answers to comments of anonymous referee #1

Thank you very much for your comments and suggestions. Our responses are written in blue in the text.

The manuscript presents 30 years of total column ozone and aerosol optical depth data measured by a Brewer Spectrometer at the Poprad Ganovce station in Slovakia, in Eastern Europe. In addition, tropopause height data are presented from regular radiosonde launches at the same site. In large parts, the paper is an update of a previous paper by Hrabčák et al. (2018), which uses the same instrument and methods. The authors report a significant decreasing trend of aerosol optical depth, in all seasons, a significant increasing trend in tropopause height, throughout most of the year, and little or no trend in total column ozone.

Consistent long-term observations, like the ones present here, are important and deserve publication in a journal like ACP. While the manuscript presents no groundbreaking new results, it still confirms findings of other studies, and helps with our understanding of long-term changes in the atmosphere. I suggest publication in ACP after a few generally minor revisions.

Section 2.4, in my opinion is rather lengthy, difficult to understand, and essentially a complete repeat of what is already presented in Hrabčák et al. (2018). I suggest to remove most of section 2.4, only describe the most salient points, and otherwise refer to Hrabčák et al. (2018). Essentially, to get aerosol optical depth, you need the measured intensity S from the Brewer, the ETC S_0 , and you have to subtract ozone and Rayleigh optical depths times their air-masses. Why not write the relevant Equation that provides aerosol optical depth, and then say that Hrabčák et al. (2018) explain how to get all the parameters in that Equation. If there is anything different from Hrabčák et al. (2018), then explain that. Doing this will reduce Section 2.4 from about 100 lines to 10 or 20 lines, and will make the manuscript much more readable.

The revised version of the manuscript will contain a significantly reduced amount of this section. However, the goal is to move most of the text to the appendix.

Figure 2: you might want to show another panel, which would present the annual cycle of tropopause height in a similar fashion. You might be surprised how closely the annual cycle of tropopause height mirrors the annual cycle of total column ozone.

Thank you for this suggestion. Indeed, the results obtained show the same behaviour as the Reviewer had pointed out. Then, Figure 2a from the article can be substituted by Figure 1 below. In this plot, the inter-annual variation of the tropopause height, computed based on monthly means, has been included next to the annual cycle of TCO. Left vertical axis represents Tropopause height in m, while right vertical axis depicts TCO in DU. It can be noticed that both magnitudes show an opposite behaviour: mean TCO values peak on March (due to Brewer-Dobson circulation, as suggested in the article), coinciding with the minimum in tropopause height. The lowest TCO concentrations are detected in October, but the maximum in tropopause height means occurs in August. Despite the shift, the annual cycle of tropopause height is in remarkable anticorrelation with the annual cycle of TCO.

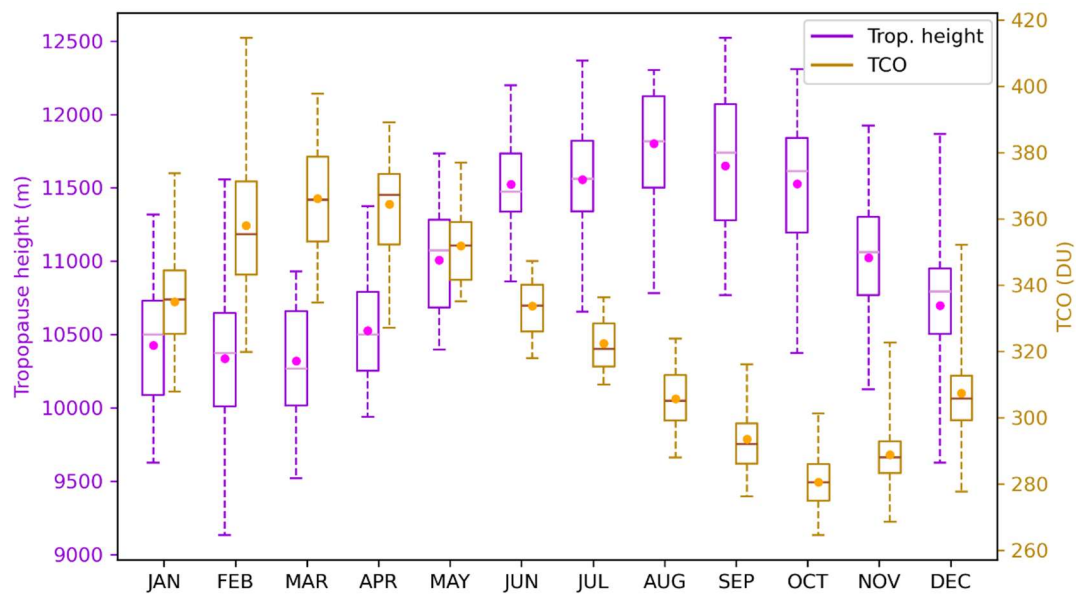


Figure 1 Boxplot showing the statistical distribution of the tropopause height (left vertical axis) and TCO (right vertical axis) for each month based on data from September 1993 to May 2024. The means are represented by solid points. The horizontal lines inside the boxes indicate the medians. The boxes extend from the 25th percentile (**U25**) to the 75th percentile (**U75**). Additionally, the lower and upper whiskers represent the corresponding minimum and maximum values, respectively.

Lines 286 to 292: I would drop this paragraph. It is not needed here.

The authors acknowledge that this paragraph may be overly descriptive and might slow down the pace of reading in the results section. However, they consider it to be quite relevant, as it describes the distribution and transport of ozone in the atmosphere, which is key to interpreting the results. So, these paragraphs:

“TCO varies strongly with latitude over the globe, with the largest values occurring at middle and high latitudes during most of the year. This distribution is the result of the large-scale circulation of air in the stratosphere that slowly transports ozone rich air from high altitudes in the tropics, where ozone production from solar ultraviolet radiation is largest, toward the poles. Ozone accumulates at middle and high latitudes, increasing the vertical extent of the ozone layer and, at the same time, TCO. The TCO is generally smallest in the tropics for all seasons. An exception since the mid-1980s is the region of low values of ozone over Antarctica during spring in the Southern Hemisphere, a phenomenon known as the Antarctic ozone hole (Salawitch et al., 2023).

TCO also varies with season. During spring, it exhibits maxima at latitudes poleward of about 45° N in the Northern Hemisphere and between 45° and 60° S in the Southern Hemisphere. These spring maxima are a result of increased transport of ozone from its source region in the tropics toward high latitudes during late autumn and winter. This poleward ozone transport is much weaker during the summer and early autumn periods and is weaker overall in the Southern Hemisphere (Salawitch et al., 2023). This natural seasonal cycle can be clearly observed in Fig. 2a. Furthermore, it has been reported that the Brewer-Dobson circulation seems to have accelerated during the last years due to the increased presence of greenhouse gases in the atmosphere (Braesicke et al., 2003; Butchart et al., 2006). Other natural atmospheric cycles (e.g., the Quasi Biennial Oscillation, El Niño-Southern Oscillation, Arctic

and Antarctic Oscillations, the solar cycle, etc.) have also been found to influence TCO (Coldewey-Egbers et al., 2022). Since these cycles operate on different timescales, assessing the individual impact of each on TCO is challenging.”

can be added to the Introduction, in a new section describing the Brewer-Dobson circulation and/or other relevant processes related to TCO changes, as well as how and to which extent they affect it.

Tables 3 to 6: It would be good to have additional columns giving uncertainty estimates for the trends.

Indeed. The authors acknowledge that these tables could contain more information relevant for the study. Thereby, they have estimated the uncertainty for the trends and have assessed their significance based on the p-value obtained as output from the Ordinary Least Squares (OLS) regression algorithm in Python.

Table 3 from the manuscript corresponds to Table 1 below. In this case, the uncertainty for the trends is given by the standard error associated to the value of the slope, which is an output from the OLS algorithm. In the case of the TCO, the uncertainties are larger than the value of the slope (except for summer). This, along with the large p-values obtained (0.83, 0.24, 0.77, 0.71 and 0.91 for spring, summer, autumn, winter and annual, respectively), confirms the apparent statistical insignificance of the seasonal trend for the TCO. On the contrary, p-values obtained for the seasonal AOD_{320} indicate highly significant trends: 1.5×10^{-8} , 1.7×10^{-6} , 9.4×10^{-7} , 2×10^{-4} and 8.6×10^{-1} for the spring, summer, autumn, winter and annual analyses, respectively. Figure 3 from the manuscript depicts these behaviours. Fig. 3b clearly shows decreasing trends, while Fig. 3a shows lines with a slight slope.

Table 2 (which is Table 4 from the manuscript) focuses on the study of AOD_{320} trends for each month of the year. The estimated uncertainties for the slopes of the linear regression have been obtained from the results of the OLS model in Python, as already mentioned. Regarding the p-values, they all indicate very significant trends: 0.028, 1.7×10^{-3} , 1.1×10^{-4} , 1.6×10^{-5} , 1.5×10^{-6} , 9.8×10^{-5} , 9×10^{-4} , 1.1×10^{-4} , 4.7×10^{-4} , 1.1×10^{-5} , 1.5×10^{-4} and 0.017 from January to December, respectively.

Besides the linear regression model, the Python algorithms for the Mann-Kendall test and Sen's slope (*pymannkendall*) were applied. The error of Sen's slope, u_s , has been estimated based on their upper and lower limits at the 95% confidence interval (U_{95} and L_{95} , respectively), in such a way that $u_s = \frac{U_{95} - L_{95}}{2}$. Taking into account the slope values and their uncertainties, the compatibility between the values obtained from the linear regression and the Mann-Kendall test can be confirmed.

The results from the linear regression analysis performed to quantify the seasonal dependence of TCO in tropopause height are summarised in Table 3 (Table 5 from the manuscript). The uncertainty associated with the slope values has been added. Based on the R^2 and p-values (8.2×10^{-9} and 3.0×10^{-6} for May/June/July and November/December/January, respectively), a statistically significant inverse relationship between TCO and tropopause height can be identified.

Finally, Table 4 (Table 6 in the manuscript) presents the results from applying the linear regression and Mann-Kendall tests to deseasonalised monthly means of tropopause height and TCO. The corresponding errors have been determined as mentioned above. For the tropopause height, the errors are higher than the slope values in January, February, March (in the case of the Mann-Kendall analysis), April, May and December. In fact, the corresponding R^2 and p-values (0.50, 0.59, 0.28, 0.37, 0.70 and 0.41,

respectively) from the linear regression analysis indicate the insignificance of the trends. For the remaining months, high p-values are also obtained in July, October and November (0.13, 0.19, 0.10, respectively); while significant dependencies are found in June, August and September (0.04, 0.008, 0.04, respectively), in agreement with the Z values from the Mann-Kendall test representing statistically significant trends at least at a 95 % confidence level (CL).

Regarding the TCO and focusing on the p-values from the linear regression test, only the trend in August can be considered significant with 90 % confidence (p-value = 0.06). For the rest of the year, the p-values are very high (0.19, 0.73, 0.47, 0.43, 0.58, 0.80, 0.65, 0.23, 0.69, 0.34 and 0.99), representing statistically insignificant trends. These conclusions are in line with the results of the Mann-Kendall test, since, according to the Z values, no statistically significant relationship can be found with a 95 % CL in the case of the TCO. In addition, it can be observed that, in both analyses, the slope errors are generally greater than the corresponding value. The exception is August, as well as January and September in the case of the linear regression test.

Table 1 Parameters obtained from the linear regression analysis of seasonal and annual TCO and AOD_{320} , based on weighted means from 1994 to 2023.

	<i>TCO</i>		AOD_{320}	
	Trend (DU/decade)	R ²	Trend (decade ⁻¹)	R ²
Spring	0 ± 2	0.002	-0.074 ± 0.009	0.69
Summer	-1.6 ± 1.3	0.05	-0.068 ± 0.011	0.56
Autumn	-0.5 ± 1.7	0.003	-0.053 ± 0.009	0.58
Winter	1 ± 3	0.005	-0.032 ± 0.008	0.39
Annual	-0.2 ± 1.4	0.0005	-0.057 ± 0.005	0.82

Table 2 Parameters (trends and R²) obtained from the linear regression analysis of AOD_{320} for each month of the year, based on data from September 1993 to May 2024. In addition, results from the Mann-Kendall test are also included. Specifically, **Z** represents the test statistic of the Mann-Kendall test, and **S** denotes Sen's slope. **Z** values indicating statistically significant trends at the 95 % confidence level (**|Z|** > 1.96) have been highlighted in bold.

	Linear regression		Mann-Kendall test	
	Trend (decade ⁻¹)	R ²	Z	S (decade⁻¹)
January	-0.021 ± 0.009	0.16	-2.2	-0.018 ± 0.012
February	-0.043 ± 0.012	0.30	-2.8	-0.044 ± 0.019
March	-0.069 ± 0.015	0.42	-4.2	-0.060 ± 0.019
April	-0.085 ± 0.016	0.49	-3.8	-0.09 ± 0.03
May	-0.067 ± 0.011	0.57	-4.6	-0.072 ± 0.013
June	-0.059 ± 0.013	0.42	-3.2	-0.05 ± 0.02
July	-0.062 ± 0.017	0.33	-3.1	-0.06 ± 0.02
August	-0.084 ± 0.019	0.42	-3.4	-0.08 ± 0.03
September	-0.070 ± 0.018	0.36	-3.6	-0.07 ± 0.02
October	-0.052 ± 0.010	0.50	-4.2	-0.049 ± 0.011
November	-0.039 ± 0.009	0.41	-3.1	-0.036 ± 0.014
December	-0.032 ± 0.013	0.19	-2.1	-0.023 ± 0.014

Table 3 Parameters resulting from the linear regression analysis of TCO means computed for different ranges of tropopause height, based on data from 18 August 1993 to 31 May 2024.

Months	Slope (DU/km)	R ²
May/June/July	-11.5 ± 0.6	0.98
Nov/Dec/Jan	-11.7 ± 1.0	0.94

Table 4 Parameters (trend and R²) obtained from the linear regression analysis of tropopause height and TCO for each month of the year, based on data from January 1994 to December 2023. Additionally, results from the Mann-Kendall test are included. Specifically, **Z** represents the Mann-Kendall test statistic, and **S** denotes Sen's slope. **Z** values indicating statistically significant trends at a confidence level of at least 95 % ($|Z| > 1.96$) are highlighted in bold.

	Tropopause height				TCO			
	Linear regression		Mann-Kendall test		Linear regression		Mann-Kendall test	
Month	Trend (m/decade)	R ²	Z	S (m/decade)	Trend (DU/decade)	R ²	Z	S (DU/decade)
January	-60 ± 90	0.016	-1.0	-110 ± 140	5 ± 3	0.06	1.2	4 ± 5
February	70 ± 120	0.011	0.8	100 ± 140	-2 ± 5	0.004	-0.5	-4 ± 7
March	90 ± 80	0.04	1.0	90 ± 130	2 ± 3	0.018	0.6	2 ± 5
April	80 ± 90	0.03	0.8	100 ± 120	-3 ± 3	0.02	-0.9	-3 ± 4
May	-30 ± 80	0.005	-0.4	-20 ± 100	1 ± 2	0.011	1.2	3 ± 3
June	130 ± 60	0.14	2.2	150 ± 80	-0.4 ± 1.7	0.002	-0.5	-1 ± 3
July	120 ± 80	0.08	1.6	200 ± 100	-0.7 ± 1.6	0.008	-0.5	-1 ± 2
August	200 ± 70	0.23	2.9	200 ± 100	-3.6 ± 1.8	0.12	-1.8	-4 ± 2
September	200 ± 100	0.14	2.1	250 ± 150	-3 ± 2	0.05	-1.4	-3 ± 3
October	100 ± 100	0.06	1.5	200 ± 130	-0.8 ± 1.9	0.006	0.1	0 ± 3
November	150 ± 90	0.09	1.9	200 ± 100	2 ± 2	0.03	1.0	2 ± 3
December	90 ± 100	0.02	0.7	70 ± 130	0 ± 3	5·10 ⁻⁶	-0.2	0 ± 4

Figure 6 and Table 6: It would be very interesting to see hypothetical TCO time series and trends, in which the -11.6 DU/km "dependence" on tropopause height has been backed out. Such a hypothetical time series in Fig. 6 might show a TCO increase, and the hypothetical effect of tropopause height changes. In Table 6, the hypothetical TCO trends would mostly become more positive by around 1 DU / decade. In fact, an additional Figure showing the seasonal variation of TCO trend, tropopause height related TCO trend and "hypothetical" TCO trend would be interesting. I suggest that the authors add such a Figure and discuss it. The slightly positive "hypothetical" TCO trend would be inline with ozone increases expected to to declining ODS (possibly enhanced by stronger Brewer Dobson Circulation). The discussion would give more meaning and context for tropopause height / climate change influences on total column ozone, and would round the paper nicely.

Thank you for your comment. The authors agree that the suggested figure could be a good addition to the article. The procedure followed to separate the contribution from the tropopause height is thus described below.

The series of hypothetical TCO has been computed by subtracting the variation in total ozone related to changes in the height of the tropopause, $TCO_{trop,i}$, from the TCO measurements, $TCO_{obs,i}$:

$$TCO_{hyp,i} = TCO_{obs,i} - TCO_{trop,i}. \quad (1)$$

To determine the $TCO_{trop,i}$ term, the relationship between variations in TCO as a function of tropopause height has been analysed. Based on the methodology described in the manuscript (Figure 5, Table 5), this dependence was studied by distinguishing among four seasons, each consisting of three months: February/March/April, May/June/July, August/September/October and November/December/January. After classifying the data points by season, the TCO values were grouped into height intervals for each season and the corresponding mean was determined. Finally, these points were linearly fitted, as shown in Figure 2. The numerical relationship between total ozone and tropopause height, α , can be estimated from the slope resulting from the linear fit (Table 5).

For simplicity, Figure 5 and Table 5 in the article refer only to the May/June/July and November/December/January seasons. The other two seasons have been added to Figure 2 and Table 5 below. It is important to mention that, taking into account the p-values obtained from the linear fits, trends have been found to be statistically significant for all seasons.

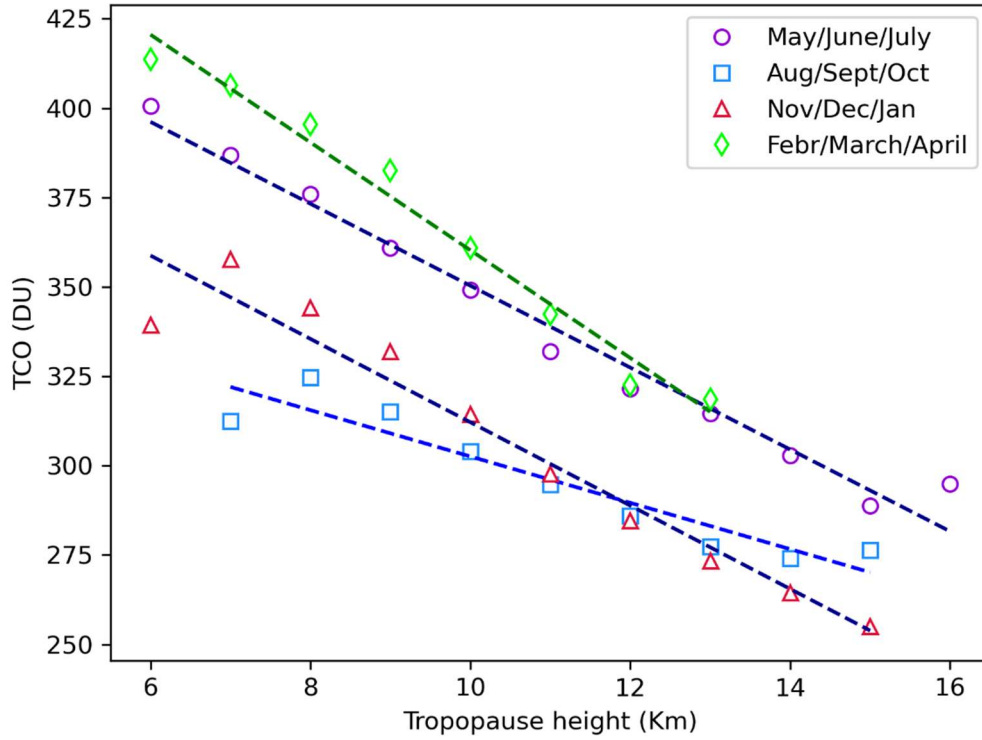


Figure 2 Linear fit of TCO means obtained for different ranges of tropopause height during the May, June, and July (purple); August, September and October (blue); November, December, and January (red); and February, March, April (green) periods. The data set considered for the plot corresponds to days between 18 August 1993 and 31 May 2024, when daily means for both tropopause height and TCO are available.

Table 5 Parameters resulting from the linear regression analysis of TCO means computed for different ranges of tropopause height, based on data from 18 August 1993 to 31 May 2024.

Months	Slope (DU/km)	R ²
May/June/July	-11.5 ± 0.6	0.98
Aug/Sep/Oct	-6.5 ± 0.9	0.89
Nov/Dec/Jan	-11.7 ± 1.0	0.94
Feb/March/April	-15.1 ± 0.9	0.98

Once the variation of the TCO as a function of the tropopause height has been quantified, the $TCO_{trop,i}$ can be computed as:

$$TCO_{trop,i} = \alpha (H_i - \bar{H}) , \quad (2)$$

where H_i is each measurement of the tropopause height and \bar{H} is the mean height, in such a way that variations in tropopause height will be considered with respect to this point. Equation 2 has been applied separately to the dataset of each season, so the corresponding \bar{H} is computed in each case. In particular, $\bar{H}_{Feb/Mar/Apr} = 10.4$ km, $\bar{H}_{MayJun/Jul} = 11.4$ km, $\bar{H}_{Aug/Sep/Oct} = 11.7$ km and $\bar{H}_{Nov/Dec/Jan} = 10.7$ km. Furthermore, as it has been already mentioned, α can be found in Table 5 for each season. Taking into account Equations 1 and 2, the hypothetical TCO will be given by

$$TCO_{hyp,i} = TCO_{obs,i} - \alpha (H_i - \bar{H}) . \quad (3)$$

The hypothetical TCO time series obtained has been deseasonalised and smoothed as described in the manuscript. This is shown in Figure 3a. The dashed line in the plot represents the linear fit of the data, the slope being slightly positive ($1.3 \pm 0.3 \text{ DU/decade}$), as predicted by the Reviewer. For completeness, Figure 3b depicts the analogous temporal evolution of TCO attributed to changes in tropopause height. As expected, the slope is negative ($-1.30 \pm 0.10 \text{ DU/decade}$). It should also be noted that it is similar in absolute value to the slope obtained for the hypothetical TCO, to which factors such as natural atmospheric cycles, next climate change influences, and the presence of ODS contribute.

The temporal evolution of the hypothetical and tropopause TCO series has also been approached by analysing their trends over the years for each month, analogous to the manuscript. Both linear regression and Mann-Kendall's tests + Sen's slope have been applied. The results are summarised in Table 6. When comparing the monthly trends of the TCO (Table 4) with those of the hypothetical TCO (Table 6), a weakly increasing trend is evident in the former only in January, whereas in the latter it also appears in March and November. It is noteworthy that not all of the decreasing trend in TCO in August can be explained solely by the tropopause increase, as the TCO_{hypo} trend for this month remains negative. Another reason for the decrease in TCO in August could be related to changes in large-scale circulation patterns in the stratosphere.

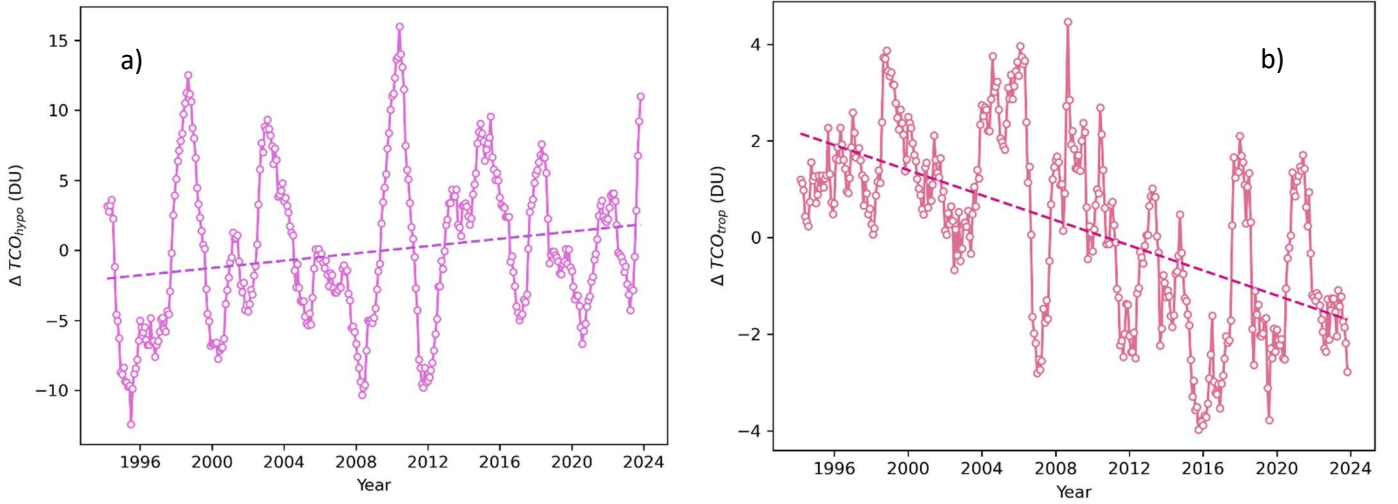


Figure 3 Representation of the time evolution of deseasonalised monthly running means of hypothetical TCO (a) and TCO related to the height of the tropopause (b) from March 1994 to November 2023 (circles). Dashed lines represent linear fits to the data.

Table 6 Parameters (trend and R²) obtained from the linear regression analysis of hypothetical and tropopause TCO for each month of the year, based on data from January 1994 to December 2023. Additionally, results from the Mann-Kendall test are included. Specifically, **Z** represents the Mann-Kendall test statistic, and **S** denotes Sen's slope. **Z** values indicating statistically significant trends at a confidence level of at least 95 % ($|Z| > 1.96$) are highlighted in bold.

Month	<i>TCO_{hypo}</i>				<i>TCO_{tropo}</i>			
	Linear regression		Mann-Kendall test		Linear regression		Mann-Kendall test	
	Trend (DU/decade)	R ²	Z	S (DU/decade)	Trend (DU/decade)	R ²	Z	S (DU/decade)
January	4 ± 3	0.06	1.2	4 ± 4	0.6 ± 1.1	0.009	0.4	0.5 ± 1.8
February	0 ± 4	0.0004	-0.2	-1 ± 5	-1 ± 2	0.016	-0.9	-2 ± 3
March	4 ± 3	0.08	1.6	5 ± 3	-2.0 ± 1.5	0.06	-1.2	-2 ± 2
April	-1 ± 3	0.007	-0.6	-1 ± 4	-1.4 ± 1.5	0.03	-0.9	-2 ± 2
May	1 ± 2	0.004	0.6	2 ± 3	0.7 ± 0.9	0.019	0.8	0.9 ± 1.3
June	1.2 ± 1.7	0.018	0.5	1 ± 3	-1.7 ± 0.7	0.17	-1.96	-1.5 ± 1.1
July	0.1 ± 1.5	4·10 ⁻⁵	-0.4	-0.4 ± 1.7	-1.1 ± 0.9	0.05	-1.14	-1.2 ± 1.3
August	-2.7 ± 1.7	0.08	-1.5	-3 ± 2	-1.3 ± 0.5	0.21	-2.5	-1.4 ± 0.6
September	-1.2 ± 1.8	0.015	-1.0	-1 ± 2	-1.6 ± 0.7	0.17	-2.7	-2.0 ± 0.9
October	0.5 ± 1.5	0.004	0.6	1 ± 2	-1.3 ± 0.7	0.12	-1.8	-1.4 ± 1.0
November	4.5 ± 1.7	0.20	1.93	4 ± 2	-2.3 ± 1.2	0.12	-2.1	-2.3 ± 1.6
December	2 ± 2	0.017	0.7	2 ± 3	-1.7 ± 1.6	0.04	-1.0	-1.6 ± 1.8

When comparing the results of both tests, an agreement between the values of the linear regression slopes and Sen's slopes can be noticed. Furthermore, it is important to mention that no statistically significant trend has been found except in November, where the corresponding p-value in the linear regression analysis is 0.015. For the other months, p-values are quite high: 0.19, 0.92, 0.14, 0.66, 0.74, 0.48, 0.97, 0.12, 0.52, 0.66 and 0.50 for January to October and December, respectively).

Regarding *TCO_{tropo}*, the trends are negative or close to 0, as expected, showing the strong correlation between the decrease in TCO and the increase in tropopause height. In this case, statistically significant trends are observed in June, August, September, October (for the linear regression analysis) and November, with corresponding p-values from the linear regression analysis of 0.025, 0.011, 0.023, 0.059 and 0.063. For the other months, these values are high: 0.63, 0.51, 0.19, 0.34, 0.47, 0.26 and 0.28 for January to May, July and December, respectively.

Finally, Figure 4 below is the one suggested by the Reviewer. This plot clearly illustrates what has been observed throughout this discussion. The hypothetical TCO shows a positive evolution over time, while the *TCO_{tropo}* trend is negative. When combined, the overall trend is slightly positive, but statistically insignificant. Therefore, it can be concluded that the TCO in the atmosphere is governed by two main components: a decrease associated with the rising tropopause height (Figure 6b), represented by *TCO_{tropo}*, and an increase represented by *TCO_{hypo}*. The second factor can be attributed, on the one hand, to the implementation of policies aimed at reducing ODS emissions. On the other hand, the acceleration of the Brewer–Dobson circulation due to climate change probably contributes to the increase in TCO by enhancing the transport of ozone to mid-latitude sites such as Poprad-Gánovce.

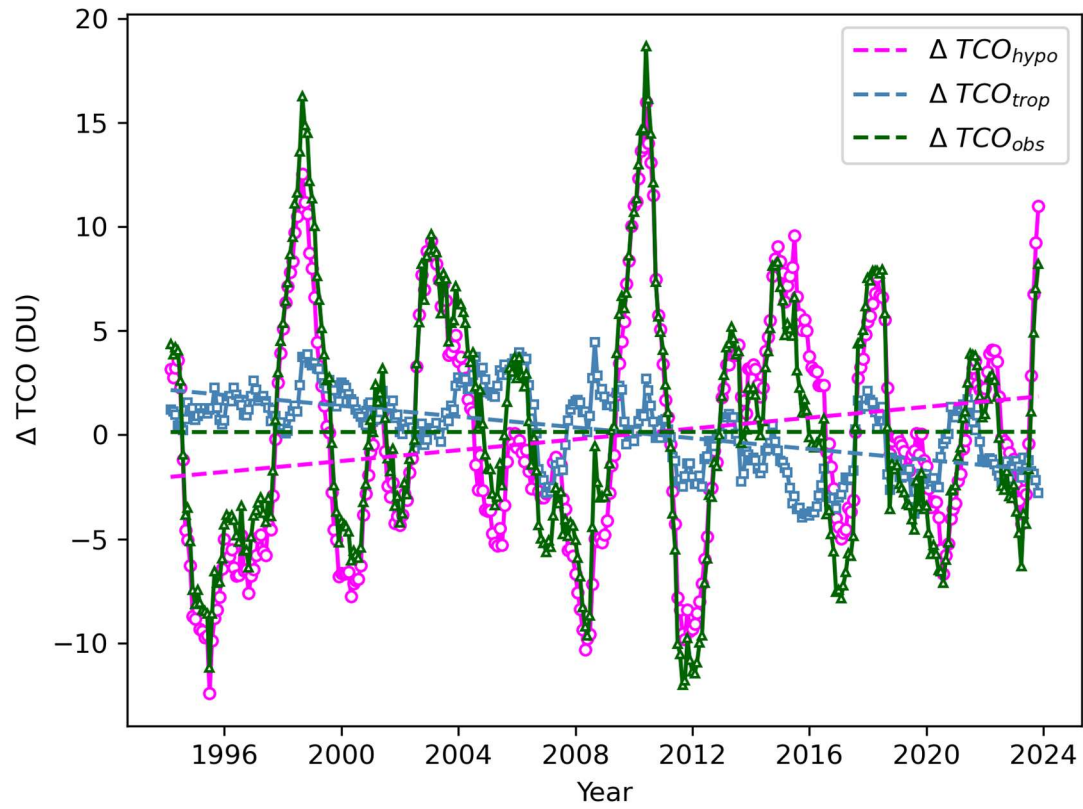


Figure 4 Representation of the time evolution of deseasonalised monthly running means of hypothetical (fuchsia), tropopause (blue) and observed (green) TCO from March 1994 to November 2023. Dashed lines represent linear fits to the data.

The authors consider this discussion to be of great interest, and all or a part of it will be added to the article. Furthermore, Figure 4 and Table 6 will be added to the manuscript to support the discussion.