

Firstly, we thank you very much for your valuable and encouraging comments and suggestions, which lead to a significant improvement of our manuscript. The detailed responses to each comment are listed below:

The text with italics indicates the reviewer's comments, and the normal text is our response.

Anonymous Referee #1

This study analyzes the seasonality and trend of tropospheric ozone over Beijing in ozonesonde observations during 2002-2010 and reports a significant positive trend of 4.6% in the tropospheric ozone column. The relative contributions of two ozone sources (dynamics processes and photochemical production) to the trend in different altitude ranges of the troposphere are investigated in combination with passively transported ozone from the CLaMS model (without tropospheric chemistry); the trend differences generally indicate the contribution from photochemical production. Results show that although transport processes drives most of the tropospheric ozone seasonality, photochemical production contributes significantly to the positive trend especially in the lower troposphere during spring and summer. This paper is generally well organized and written and is suitable for publication on ACP. However, some of the discussions could be improved (see specific comments below). The abstract could be improved to more accurately reflect the main conclusions of this study. I recommend this paper to be published after addressing the following specific comments:

1. *“With a clear positive trend in the maximum summer ozone concentration” in the abstract emphasizes the trend during the summer, but according to Table 2, there is a similar trend (relatively larger) trend during the winter for 3-9 km and 0-3 km. Maybe you should change it to “with a clear positive trend over the last decade”, consistent with what is said in the conclusion section.*

Reply: In Figure 1a, we could see the summer ozone concentration clearly

increasing over the last decade in the troposphere. Based on this phenomenon, we conclude the result. But you are right; there are similar trends during the winter for 3-9 km and 0-3 km. So we accept your suggestion in the abstract and change it to “with a clear positive trend over the last decade”.

2. In abstract, I am not clear about the main purpose of the sentence “This trend is close to the significant trend of . . . 3.4 %/yr-1”. This does not really support the connection between the overall trend and 0-3 km trend during summer. As shown in Table 2, the overall trend is even closer to the trend of 0-3 km ozone during winter (4.6%) and the trend of 3-9 km during the summer (4.8%). I suggest removing it or changing this sentence to what accurately reflects the conclusion.

Reply: Here we hope to emphasize the trend caused by photochemical production. In order to make it clear, we change the two sentences to: The observed significant trend of tropospheric column ozone is mainly caused by photochemical production (3.1 %yr⁻¹ for a mean level of 52 DU). This trend is close to the significant trend of partial column ozone in the lower troposphere (0–3 km) resulting from the enhanced photochemical production during summer (3.0 %yr⁻¹ for a mean level of 23 DU).

3. In abstract, suggest changing “contributed to” to “contributes to” (i.e., use current tense for scientific statement).

Reply: This suggestion has been adopted.

4. Section 2.1, Total ozone from Dobson spectrometer, and satellite observations (when Dobson not available) are used to scale the ozone profile. Since total ozone columns from different sources are used to scale the entire ozone profile and this study focuses on tropospheric ozone, it is worthwhile to check the trend without applying any correction based on recommendations from the SPARC report (1998) and a recent study by Morris et al. (2012). The authors mentioned that there is no significant trend in the correction factors, but it would be more useful to check and report whether the trend is affected and report that how much the trend is changed if

the change is significant.

Reply: The results of our sensitivity study are summarized with the sentence at page 4, line 14 “...with no statistically significant trend”. We also calculated the trend without applying the correction factors. We found the results are almost no difference compared to the results which applied correction factors. The following two figures are the same as Figure 3 and Figure 4 respectively in the paper, but didn’t multiply correction factors.

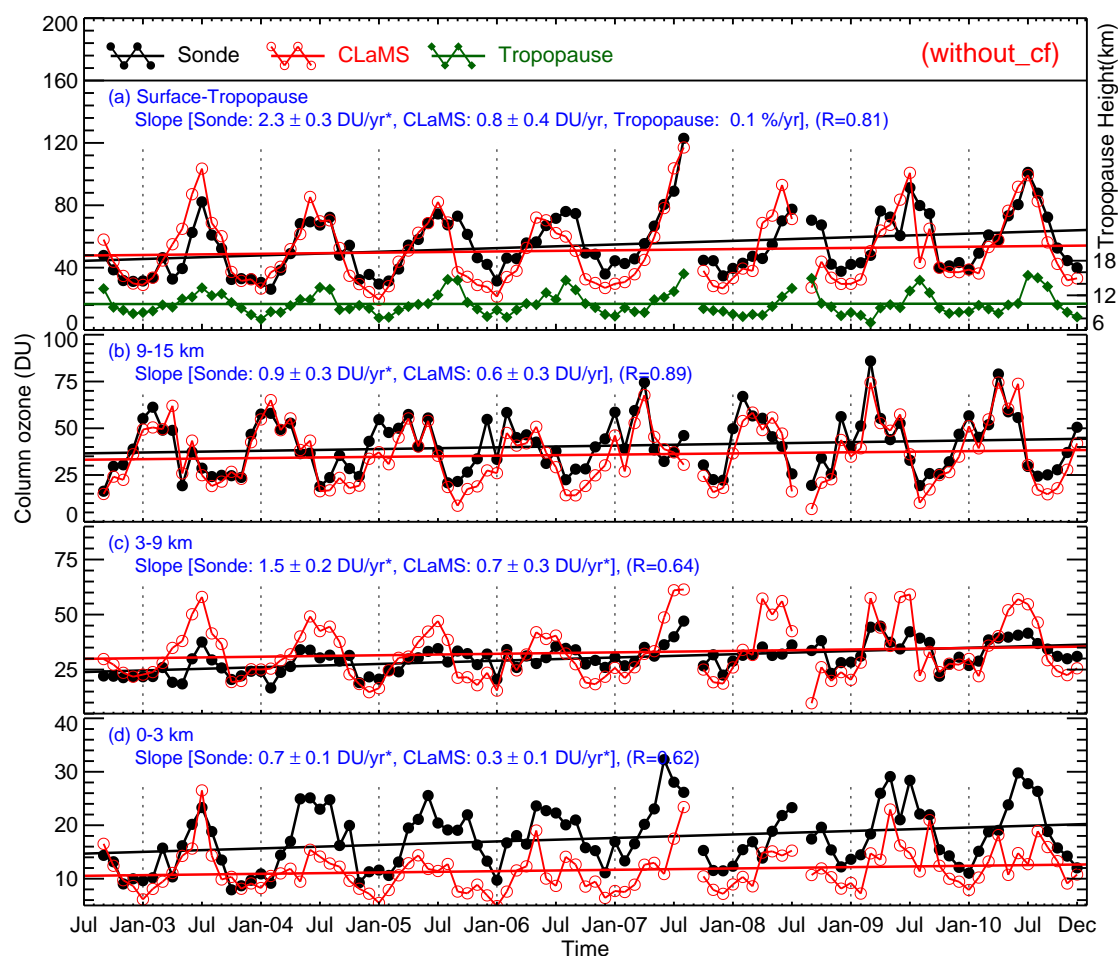


Fig. 3. Trends of monthly mean partial column ozone for (a) the whole troposphere (surface–tropopause), (b) 9–15 km layer, (c) 3–9 km layer and (d) 0–3 km layer from observation (Sonde) and simulation (CLaMS) during 2002–2010, together with the first (LRT1) thermal tropopause. The numerical values for the slopes with symbol “*” pass the 95% significance criterion. The number R is the correlation coefficient between observation and simulation. There were no observations available during September 2007 and August 2008.

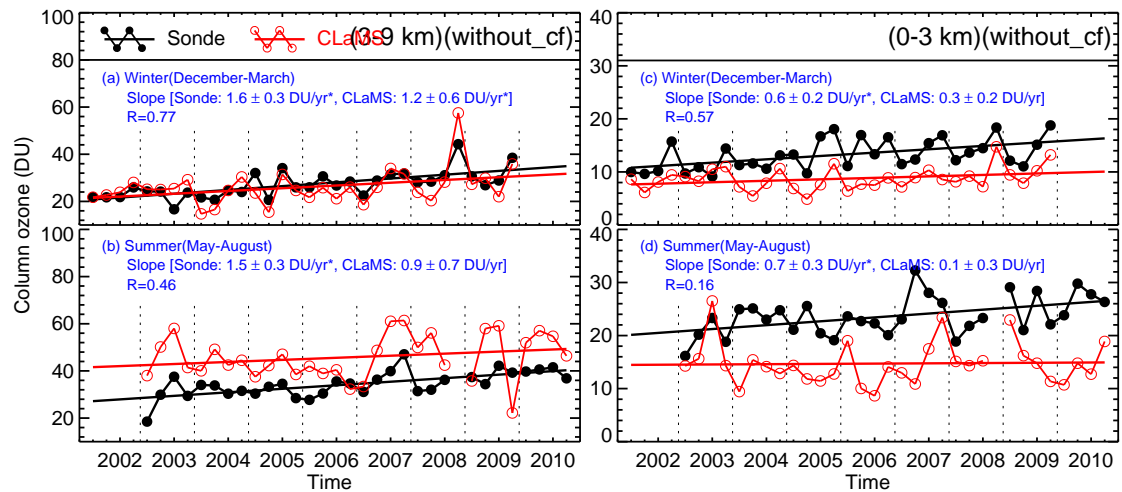


Fig. 4. Trends of monthly mean partial column ozone for 3–9 km layer and 0–3 km layer in winter (December–March) and summer (May–August) from observation (Sonde) and simulation (CLaMS) during 2002–2010. The numerical values for the slopes with symbol “*” pass the 95% significance criterion. The number R is the correlation coefficient between observation and simulation. There were no observations available during August 2008.

5. P11179, line 10, please specify which OMI total ozone product is used as there are two products: OMT03 and OMD0A03.

Reply: Thank you very much for this suggestion. The OMI total ozone product used here is OMT03. We also add this information in the manuscript at Page 4, Line 18.

6. P11180, line 1, change “near to the surface” to “near the surface”.

Reply: This suggestion has been adopted.

7. In Figure 1 caption, it would be better to define LRT1 and LRT2 as the readers understand them without reading the text.

Reply: This suggestion has been adopted.

8. P1181, first paragraph, what does LRT1 and LRT2 mean? The two tropopause levels in case of double tropopause? I think that more description of their physical

meaning is needed here.

Reply: The individual temperature profiles are used to determine the location of the first lapse-rate tropopause (LRT1) and, if present, the second tropopause (LRT2), using the definition from the World Meteorological Organization (WMO, 1957).

(a) The first tropopause is defined as the lowest level at which the lapse rate decreases to 2 °C/km or less, provided also the average lapse rate between this level and all higher levels within 2 km does not exceed 2 °C/km.

(b) If above the first tropopause the average lapse rate between any level and all higher levels within 1 km exceeds 3 °C/km, then a second tropopause is defined by the same criterion as under (a). This case accords to double tropopause (or tropopause folding).

We add this information in the manuscript at Page 6, Line 7-12.

9. P11181, line 23: LRT2 does not seem to have a strong seasonal cycle.

Reply: We modified this to (**in the manuscript at Page 6, Line 26-27**): LRT1 (Fig. 1) reveal strong seasonal cycles with low and high tropopause in winter and summer, respectively. The seasonal variation of LRT2 (Fig. 1) is relative small compare to LRT1.

10. P11182, line 20, change “chemistry” to “tropospheric chemistry”.

Reply: This suggestion has been adopted.

11. P11183, line 8, change “exactly” to “almost” as there are some differences.

Reply: This suggestion has been adopted.

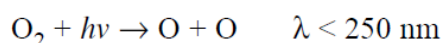
12. P11183, line 16, I would not call them consistent if aircraft and ozonesonde observations peak at different time. Do you have any idea about their discrepancy (ozonesonde data peak in June and aircraft data peak in spring)?

Reply: Sorry for this mistake. The tropospheric ozone over Beijing also exhibits a narrower early summer (June) peak in the lower troposphere based on aircraft

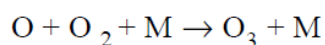
observation (Ding et. al., 2008). It's the same with our result from ozonesonde observation. We revised this sentence in the text (**in the manuscript at Page 8, Line 19**).

13. In section 3.2 and Figure 2, there is a major difference between ozonesonde and CLaMS-PO3: in August and September, low ozone reaches much higher altitude, above some high ozone in the middle troposphere (Fig. 2b), causing more negative ozone gradients in Fig. 2d than in Fig. 2c. Please comment on this.

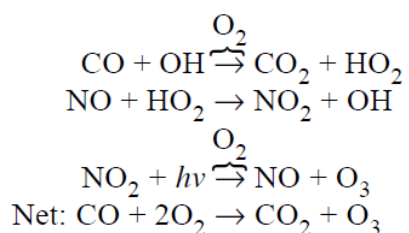
Reply: This because there is no ozone chemistry both in stratosphere and troposphere in the model simulation. In the stratosphere the principal reaction that leads to the production of ozone is photolysis by sunlight with wavelengths below 250 nm which breaks the O₂ bond (John C. McConnell and Jian Jun Jin, 2008):



The O atoms formed thus combine rapidly with O₂ to form O₃:



In the troposphere and lower stratosphere where O₂ cannot be photodissociated, O₃ is formed by chemical reactions that result in the chemical breaking of an O₂ bond. The prime reaction sequence is (John C. McConnell and Jian Jun Jin, 2008):



Due to the lack of ozone production in the stratosphere, the downward transport of ozone is also underestimated in the model simulation. This point is partially discussed in the manuscript at the page 12.

14. P11184, lines 2-4, as the positive trend is not caused by enhanced ozone, this sentence may be rephrased to “there is a positive trend during periods (spring and summer) of enhanced lower tropospheric ozone concentrations over the last decade”.

Reply: This suggestion has been adopted. We rephrased to “We show in the next section that, for Beijing during spring and summer, there is a positive trend of enhanced lower tropospheric ozone concentrations over the last decade.”

15. P11184, line 8, change “each profiles” to “each profile”.

Reply: This suggestion has been adopted.

16. P11184, line 13, what is the source of OMI TCO?

Reply: OMI TCO data are made by Liu et. al., (2010) (in the manuscript at Page 9, Line 16-17).

17. Fig. 3a shows the trend in tropopause but there is no discussion related to this. Since the tropopause trend is in different units, it is not clear about the contribution of tropopause to the overall TCO trend. To evaluate its impact, I suggest calculating TCO using the all-year average monthly mean tropopause and check the change in the TCO trend.

Reply: As your recommendation, we calculated trends of the all-year averaged TCO and tropopause height (LRT1) and found no significant change compare to the results using monthly mean data. The following figure shows the main results.

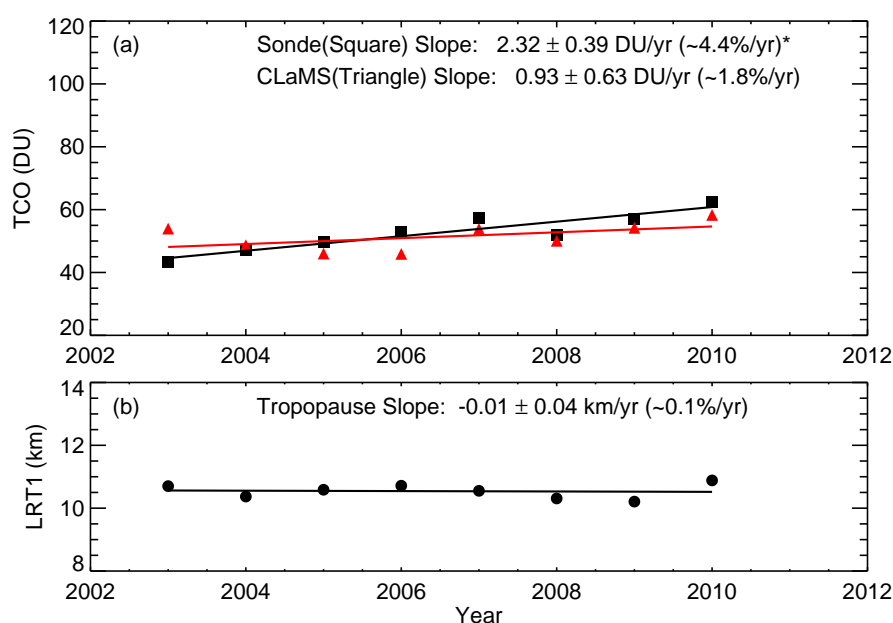


Fig. Trends of all-year average monthly mean tropospheric column ozone (TCO) from observation

(Sonde) and simulation (CLaMS) during 2003–2010, together with the first (LRT1) thermal tropopause. The numerical values for the slopes with symbol “*” pass the 95% significance criterion.

We add one sentence **in the manuscript at Page10, Line 10-11**: Because there is no trend for LRT1 (0.1 \%yr^{-1} with no statistical significance), these TCO trends can’t originate from the long-term variation of LRT1.

18. Footnotes in Tables 1 and 2, change “slop” to “slope”.

Reply: This suggestion has been adopted.

19. Table 2 should also include the trend comparison for the TCO as well as 9-15 km layer to make the discussion/abstract/conclusion clearer as there are discussions of the trends/mean values of TCO and upper layer ozone between winter and summer in the text (e.g., P11186, lines 15-18, abstract, conclusion).

Reply: This suggestion has been adopted. We add the trends of TCO and 9-15 km layer in the table 2. In the following is the new Table 2.

Table 2. Trends of monthly mean partial column ozone from observation (Sonde) and simulation (CLaMS) in winter (w: December–March) and summer (s: May–August) during 2002–2010^a

Layer	Mean (DU)		Absolute trend (DU)		Relative trend (%)			R
	Sonde	CLaMS	Sonde	CLaMS	Sonde	CLaMS	Diff	
TCO (w)	38	34	*2.1±0.9	1.1±1.1	*5.5	2.9	2.6	0.70
9–15 km (w)	49	43	0.5±0.9	0.2±0.9	1.0	0.4	0.6	0.87
3–9 km (w)	26	26	*1.6±0.3	*1.2±0.6	*6.2	*5.8	0.4	0.79
0–3 km (w)	13	9	*0.6±0.2	0.3±0.2	*4.6	2.3	2.3	0.61
TCO (s)	70	77	*3.4±0.8	1.4±1.3	*4.9	2.0	2.9	0.54
9–15 km (s)	34	36	1.6±0.9	1.5±1.2	4.7	4.4	0.3	0.92
3–9 km (s)	33	46	*1.6±0.3	0.9±0.7	*4.8	2.7	2.1	0.40

0–3 km (s)	23	15	*0.8±0.2	0.1±0.3	*3.4	0.4	3.0	0.08
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^aAbsolute trends (slope±standard error) for the whole troposphere (TCO), 9–15 km layer, 3–9 km layer and 0–3 km layer are shown in the table, together with the correlation coefficients (R) between Sonde and CLaMS. The trends with symbol “*” pass the 95% significance criterion. The relative trend column list Absolute(trend)/Sonde(mean)×100% and the difference of relative trends (Diff) between Sonde and CLaMS.

20. P11187, line 22: “the largest . . . during summer” seem to contradict to the sentence in line 19 (3.1 % yr⁻¹), please clarify this or modify the sentence.

Reply: We revised the sentence (**in the manuscript at Page 12, Line 21-22**) to: For seasonal analysis of the partial column ozone in different altitudes, the largest photochemically produced trend occurs in the lower troposphere (3.0 %yr⁻¹) during summer.

21. P11188, line 5, the sentence “This constitutes with . . .” is confusing. I suggest changing it to “Most of the trend is caused by photochemical production (~3%)”

Reply: This suggestion has been adopted.

References:

- Ding, A. J., Wang, T., Thouret, V., Cammas, J. P., and Nédélec, P.: Tropospheric ozone climatology over Beijing: analysis of aircraft data from the MOZAIC program, *Atmos. Chem. Phys.*, 8, 1-13, 2008.
- John C. McConnell and Jian Jun Jin, Stratospheric Ozone Chemistry, *ATMOSPHERE-OCEAN*, 46 (1), 2008, 69–92, doi:10.3137/ao.460104.
- Liu, X., Bhartia, P. K., Chance, K., Spurr, R. J. D., Kurosu, T. P. Ozone profile retrievals from the Ozone Monitoring Instrument [J]. *Atmos. Chem. Phys.*, 2010, 10(5): 2521-2537.
- Morris, G. A., Labow, G., Akimoto, H., Takigawa, M., Fujiwara, M., Hasebe, F., Hirokawa, J., and Koide, T.: On the use of the correction factor with Japanese ozonesonde data, *Atmos. Chem.*

Phys. Discuss., 12, 15597-15638, doi:10.5194/acpd-12-15597-2012, 2012.

SPARC-IOC-GAW: Assessment of Trends in the Vertical Distribution of Ozone, SPARC report No. 1, WMO Global Ozone Research and Monitoring Project Report No. 43, Geneva, 1998.

WMO: Meteorology - A three dimensional science: Second session of the Commission for Aerology, WMO Bulletin, IV, 134-138, 1957.