Author's Response:

Thanks for all the comments and suggestions. We have carefully revised the manuscript according to these suggestions.

Our point-to-point responses are listed below:

Comments from Referee 1:

Suggestions for technical corrections or reasons for rejection

This work reports observations of ozone in the troposphere and stratosphere obtained from launching ozonesondes in Beijing during 2002-2016. The data provide important information on the ozone variations during this period of drastic changes in the emissions of anthropogenic pollutants in the North China Plains (NCP). The data have been analysed with a stratospheric ozone tracer model and discussed qualitatively in the context of recent emissions information.

I have two major concerns at this stage.

(1) First, it is very surprising to see a huge drop in ozone below 3 km after 2011 which is not convincingly explained by emission changes, raising concern on potential problem with the observation data. I am saying the ozonesonde data has problem for certain, but I advise the author to give more information to eliminate readers' doubt on data quality – after all, this dataset is the basis of the paper. Did other ozone measurements in Beijing give similar result (i.e., sharp and step-wise ozone decrease after 2011)? Was there a sudden and persistent change in large-scale dynamics after 2011?

Author's response: *We give more information about ozonesondes and relative references. The ozonesonde data has been proved reliable and used to validate satellite measurements (Bian et al., 2007) and model products (Wang et al., 2012).*

We think the change in trend is mainly the result of decrease of ozone precursors. So, we add a long-term variation of tropospheric NO_2 from OMI. The huge drop of ozone in 2011-2012 may attribute to the change of transport from stratosphere. Because CLaMS which has no tropospheric ozone chemistry also shows the huge drop.

There was no other ozone measurements in Beijing except satellite data which is not better than ozonesonde measurements below 3 km.

We discussed the possible dynamical factors (ENSO and tropopause) which may be related to the ozone change.

Author's changes in manuscript: *Information about ozonesondes is at Lines 64–76. Added NO_2 from OMI is at Lines 161–168. Sudden decrease caused by transport is at Lines 284–286. The discussion about ENSO and tropapause is at Lines 275–282.*

(2) Second, more in-depth analyses of the data are needed. It is better to use a model with reasonable representations of tropospheric ozone chemistry and/or other chemical (e.g., satellite observations of ozone precursors) and meteorology data to better explain the ozone changes. While the second point can be addressed during the discussion stage, I encourage the author to do this now to boost the rigor of the analysis.

Author's response: *We use the version 3 of Aura Ozone Monitoring Instrument (OMI) Nitrogen Dioxide (NO2) standard product to discuss the influence of precursors on the long-term variation of tropospheric ozone in Beijing. The result shows that the decrease of tropospheric NO2 plays an important role in the decrease of tropospheric ozone.*

Author's changes in manuscript: *Lines 161–168 and Figure 5.*

Other quick suggestions to improve readability:

(1) One or two sentence about the site will be helpful.

Author's response: *The ozonesondes were released from Beijing Observatory (39.81°N, 116.47°E; 31 m above sea level).*

Author's changes in manuscript: *Line 80.*

(2) Give a brief introduction of the chemistry in CLaMS.

Author's response: *We gave a brief introduction of the chemistry in CLaMS in Section 2.2.*

Author's changes in manuscript: *Lines 84–88.*

(3) Line 86, briefly describe how depersonalized ozone is achieved.

Author's response: *We added a brief introduction about how depersonalized ozone is achieved.*

Author's changes in manuscript: *Lines 113–116.*

(4) Fig 1 seems to show seasonal (not deseasonalized) ozone?

Author's response: *No, it is deseasonalized ozone. It is clearer to see the trend after remove the seasonal variation of ozone (Figure A).*

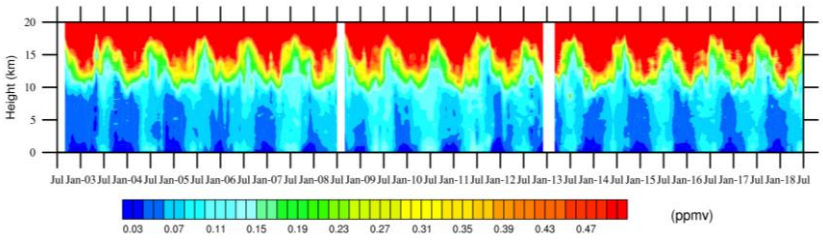

Figure A. Monthly mean ozone mixing ratio (units: ppmv) over Beijing measured by the IAP ozonesonde.

(5) Figure 1b: Why there are no data simulated using the model during July 2008, January, and July 2013?

Author's response: *There was no ozonesonde data during July 2008 and January 2013. In July 2013, we got ozone mixing ratio but lost the information of balloon locations (latitudes and longitudes during the fly). CLaMS is unable to calculate ozone mixing ratio without the information of balloon locations.*

Author's changes in manuscript: *Lines 78–79 and lines 508–509.*

(6) Table 1: Data for 0-3 km simulated by CLaMS are missing.

Author's response: *Since there is no tropospheric ozone chemistry in CLaMS, it is meaningless to give data by CLaMS in 0-3 km which is significantly smaller than data by ozonesondes.*

Comments from Referee 1 in the interactive discussion:

This paper presents ozonesonde observations of ozone in the troposphere and stratosphere in Beijing during 2002-2018. The data are analyzed with a stratospheric ozone tracer model and discussed qualitatively in the context of recent emission changes in Beijing and satellite NO2 data. The ozonesonde data provide valuable information on the ozone variations during this period of drastic changes in anthropogenic pollutants in Beijing and are an important contribution to ozone and climate research. I have some concerns on analysis and interpretation of the results. The paper can be accepted after these concerns have been addressed.

Major comments:

(1) A sudden drop in lower tropospheric ozone (< 3 km) after 2011 is surprising. It is inconsistent with satellite NO2 data shown in Fig 5 (and NOx emission inventory) which indicate gradual decrease in NOx emission after 2011. Recently reported surface measurements at two rural sites near Beijing (Shangdianzi and Gucheng) also did not observe sudden ozone drop around 2011/12 (Xu et al., 2020). I suggest comparing satellite observed tropospheric ozone to verify the sudden change observed in the present study. If no problem is found on data quality, the stepwise change is most likely due to change in large scale dynamics after 2011. The stratospheric model used in this study shows little change in stratospheric contribution to lower tropospheric ozone, but it may be the case that transport within the troposphere played a role. I suggest author add more analysis in this direction. For example, back trajectories can be calculated to see if there was change in transport from other parts of troposphere after 2011.

Author's response: *We compared ozonesonde to OMI observation (Fig. B) as suggested. The data quality of OMI in troposphere is not as good as in stratosphere, especially in lower troposphere where there are often missing values in dataset. Even so, the sudden drop in the period of 2011-2012 are still found in middle troposphere and especially in UTLS. So, we believe the data quality of ozonesonde and the sudden drop we found are reliable. The reason for this sudden drop may mainly due to the changes in UTLS rather than NOx emission. Because the sudden drop is also found in CLaMS simulation which has no tropospheric ozone chemistry.*

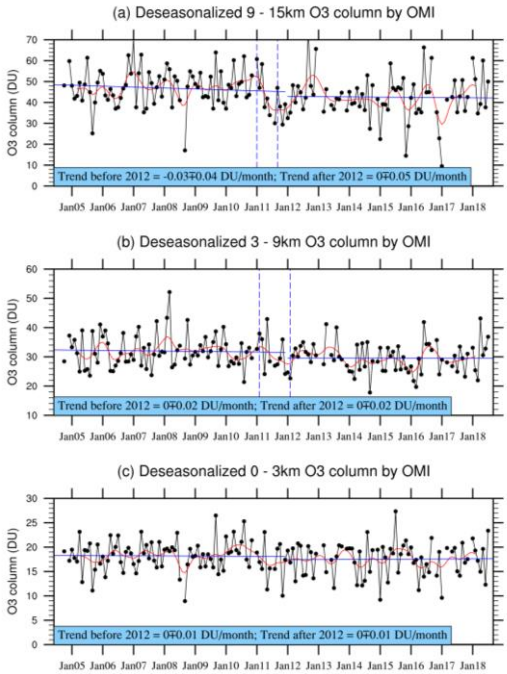

Figure B. Deseasonalized monthly mean partial columns of ozone over Beijing (black solid lines and dots) measured by OMI and the corresponding Gaussian-weighted means using a half-width of 12 months

(2) The trend analysis can be improved; it is not clear why the trend calculation in the main text is different from the linear regression shown in the figures. In addition, the level of statistical significance in trend analysis should be provided.

Author's response: *We checked every value of trend in the main text, and they are the same as the linear regression in the figures. We add statistical significance testing in the O_3 and NO_2 trends, most of them passed the 95% significance criterion. For the seasonal O_3 trends, most ozonesonde trends in lower troposphere and mid-troposphere before 2012 passed the 95% significance criterion. Since there are fewer samples after 2012, some trends only passed the 90% significance criterion.*

Author's changes in manuscript: *We provided statistical significance in Figures 3-5 and in Table 1.*

(3) The lower tropospheric ozone in the present study appeared to have a small positive trend after the 2011 drop (Fig 3). This trend is not supported by author's contention that NOx reduction has decreased ozone. It is instead similar to surface ozone increase observed in many urban areas from the Ministry of Ecology and Environment network since 2013, which has been attributed to the nonlinear chemistry of ozone precursors (NOx emission decrease and VOC emission increase) and aerosol decrease, as well as being affected by meteorological variation (see for example, Li et al., 2019; Liu and Wang, 2020).

Author's response: *We noticed that O_3 trend is still positive after the 2011 drop, but it is much slower than before due to the reduction of NOx. However, there are other precursors which might be responsible for the small positive trend after the 2011 drop. Thanks for showing us the two papers (Li et al., 2019; Liu and Wang, 2020). We added them when we mentioned the possible reasons of meteorological variation in the discussion and conclusions.*

Author's changes in manuscript: *Lines 307-308, 400-401, 271-273 and 469-471.*

Minor comments:

(1) Page 2, line 40-42, "Increasing surface ozone . . ." . Please note that recent studies have shown levelling off/decrease in surface ozone levels in rural areas of eastern China and in outflow of eastern China air masses (Xu et al., 2020; Wang et al., 2019).

Author's response: *We added the recent studies which show the decrease in surface ozone levels in next paragraph.*

Author's changes in manuscript: *Lines 46-48.*

(2) Page 2, line 56, Consider modifying the statement "it is not known. . ." as it contradicts the author's earlier review of Dufour (2018) on the lower tropospheric ozone trend in NCP (which includes Beijing).

Author's response: *We deleted this sentence.*

Author's changes in manuscript: *Line 58.*

(3) Page 2, line 100, Define "average percentage method", and clarify why a different (linear regression method) is used in the figures.

Author's response: *we explained the method. The method is used to remove the seasonality in the time series. As a result, we got deseasonalized O3 (black dots in figures). Linear regression method is applied on the deseasonalized O3 to get the trend of O3. These methods are used on different steps for different purpose.*

Author's changes in manuscript: *Lines 114-115.*

Comment from Referee 2:

Suggestions for technical corrections or reasons for rejection

During the past years, many papers have been published focused on explaining both trends and contribution of tropospheric ozone in some areas. Although this is very interesting for understanding the ozone formation processes and to define strategies for reducing ozone pollution, providing tropospheric ozone estimations is still a challenge because tropospheric ozone is a secondary pollutant and the formation and destruction reactions of it are very complex and difficult to modelize. Besides, it's difficult to isolate the contribution of the different precursors sources. In this paper, Zhang Y. et al. show the trend of ozone observations in Beijing from 2010 to 2018 providing relevant information about the levels of ozone in this area, which has not been studied yet. However, from my point of view, this paper is not very innovative because the applied methodology is very similar to previous works.

I first provide some general suggestions/observations regarding the paper and then I list specific comments.

GENERAL COMMENTS

(1) This study is focused on the trends of observed O3 measured in Beijing. A very similar study was done by Wang et.al. (2012) and this manuscript refers to this paper several times. From my point of view this should be highlighted in the introduction and it should be clearer because the applied methodology is really similar. This manuscript could be an extension of the first one.

Author's response: *We highlighted the extension of the work by Wang et. al. in the introduction and it is clearer now.*

Author's changes in manuscript: *Lines 56–61.*

(2) The authors use different names or different nomenclature along the text to refer to the same things. Defining these concepts at the beginning of the paper should be more convenient to avoid repetition of the definition of the different concepts. For example the names of the different layers with heights are in lines 101, 104, 139, 150.

The definition of the month of the year of each season should appear at the beginning, e.g.line 105.

Author's response: *We checked the nomenclatures, especially the layers and the seasons. We gave the definitions when they were firstly mentioned and deleted the repeated description.*

SPECIFIC COMMENTS

Introduction

(1) Line 40: Cooper et.al references doesn't appear in the Reference list.

Author's response: *Cooper, O. R., Parrish, D. D., Ziemke, J., Balashov, N. V., Cupeiro, M., Galbally, I. E., Gilge, S., Horowitz, L., Jensen, J. -F., Naik, V., Oltmans, S. J., Schwab, J., Shindell, D. T., Thompson, A. M., Thouret, V., Wang, Y., and Zbinden, R. M.: Global distribution and trends of tropospheric ozone: An observation-based review, Elem. Sci. Anthr., 2, 000029, doi:http://doi.org/10.12952/journal.elementa.000029, 2014.*

Author's changes in manuscript: *Lines 338–341.*

Data and model

(2) Line 65: The authors say that the data used was measured about once a week. Explain in detail when data was collected (add dates and hours) and which gaps with no data you have if it's possible.

Author's response: *The ozone profiles have been observed about once a week since 2002 at 14:00 local time (06:00 UTC). But we don't have a fixed date in one week. In some intensive observation periods (e.g., 24 March to 10 April 2003), ozonesondes were launched every day. However, there was no observation (gaps in Figure 1) in two periods (July 2008 and January 2013).*

Author's changes in manuscript: *Lines 77–79.*

(3) Line 76: Why 40-year of CLaMS simulation?

Author's response: *We have 40-year of CLaMS simulation, and we use only the period of 2002-2018 which is consistent with ozonesonde data.*

(4) Line 82: According to this manuscript there is no photochemistry in CLaMS model, but in Wang et.al. (2012) the model is executed without ozone chemistry (CLaMS-PO3: passively transported ozone). The model could be executed with and without chemistry? This should be defined more clearly. The definition of the model should be included before the explanation of the configuration.

Author's response: *There is comprehensive photochemistry in CLaMS in stratosphere. However, there are very simple reactions in troposphere. The model could be executed with and without ozone chemistry. To isolate and quantify the long-term trend caused by transport from the stratosphere, a CLaMS simulation without ozone chemistry in troposphere is considered in this paper.*

Author's changes in manuscript: *Lines 95–99.*

(5) Line 96: Define here the concepts of lower troposphere (3-9 km), mid-troposphere (9-15 km) and UTLS (9-15 km). Doing that the authors can use these terms without including the height. This information is repeated several times in the current version of the manuscript.

Author's response: *We removed the repeated description of 0-3, 3-9, 9-15km.*

(6) Line 104-106: T"The CLaMS simulations in the mid-troposphere (3-9) km are much closer to the ozonesonde measurements (Fig.2b). CLaMs seems to overestimate the transport of ozone from the stratosphere to the troposphere, which is strongest during winter and spring".
Comment: If we analyze the figure 2b, spring and summer are the seasons with highest overestimation of O3 values.

Author's response: *After recheck Figure 2b, it is sure that CLaMS overestimate ozone in spring and underestimate ozone in autumn. It is hardly to say overestimate or underestimate in winter and summer except few dots. So, it should be more precise to say "CLaMs overestimates the transport of ozone from the stratosphere to the troposphere, which is strongest in spring".*

Author's changes in manuscript: *Lines 138–139.*

(7) Line 105: Avoid using "seems".

Author's response: *It was changed to "CLaMS overestimates the transport of ozone from the stratosphere to the troposphere, which is strongest during winter and spring".*

Author's changes in manuscript: *Lines 138–139.*

(8) Line 113-114: Delete heights.

Author's response: *Deleted.*

Author's changes in manuscript: *Line 133.*

(9) Line 139: Delete (0-3 km altitude)

Author's response: *Deleted.*

Author's changes in manuscript: *Line 181.*

(10) Line 142: Delete (9-15 km altitude)

Author's response: *Deleted.*

Author's changes in manuscript: *Line 183.*

(11) Line 141: Are there some previous studies analysing the most frequent mesoscale patterns to corroborate this?

Author's response: *I don't understand this question. I am not sure what in Line 141 was asking to be corroborated by previous studies.*

Tropospheric ozone chemistry dominates the trends in the lower troposphere (0–3 km altitude) in summer and
140 autumn. The contribution in CLaMS is so small here that any stratospheric influence can be neglected. We call this range the "troposphere-dominated range". By contrast, the stratospheric influence is dominant in the UTLS (9–15 km altitude) in winter and spring and the tropospheric contribution can be ignored. We call this range the "stratosphere-dominated range". All the other combinations of seasons and altitudes are a superposition of the troposphere- and stratosphere-dominated ranges and we call such combinations the "superposition range".

(12) Line 150: Replace "9-15km" by "UTLS"

Author's response: *"9-15km" has been replaced by "UTLS".*

Author's changes in manuscript: *Line 192.*

(13) Line 156-157: The months of each season should be defined previously.

Replace "before and after the decrease" by "before and after 2012" to be consistent with Figure 3 and 4.

Authors use different nomenclature in the text that doesn't appear in Table 1. For example MO3, TO3, Trel

Author's response: *"Before and after the decrease" has been replaced by "before and after 2012".*

We used MO3, TO3, Trel in Table 1 which consistent with the text.

Author's changes in manuscript: *Table 1.*

(14) Line 171: Why > 20%? Give more detailed information.

Author's response: *Δ_{rel} varies from 0% to 47.7%, we want to show the most significant change of ozone. So we choose Δ_{rel} > 20%. One can also choose > 15%, 25% or 30%... depends on how significant the ozone change was.*

(15) Line 182: Delete (3-9 km)

Author's response: *Deleted.*

Author's changes in manuscript: *Line 223.*

(16) Line 190: Replace "almost neutral" by "almost zero".

Author's response: *"Almost neutral" has been replaced by "almost zero".*

Author's changes in manuscript: *Line 231.*

(17) Line 220: Replace "the increase in ozone has been controlled" by " has been moderated compared to..". What does "been controlled" mean?

Author's response: *We replaced "the increase in ozone has been controlled" by "the increase in ozone has been moderated since 2012".*

Author's changes in manuscript: *Line 262.*

(18) Line 229: Wrong references: Replace "Zheng" by "Zhang"

Author's response: *The reference is "Zheng, B., Tong, D., Li, M., Liu, F., Hong, C. P., Geng, G. N., Li, H. Y., Li, X., Peng, L. Q., Qi, J., Yan, L., Zhang, Y. X., Zhao, H. Y., Zheng, Y. X., He, K. B. and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmos. Chem. Phys., 18(19), 14095–14111, doi: 10.5194/acp-18-14095-2018, 2018."*

Author's changes in manuscript: *Line 497.*

(19) Line 231: Wrong reference: Want et. al. 2019 doesn't appear in the reference section.

Author's response: *Wang, N., Lyu, X., Deng, X., Huang, X., Jiang, F., and Ding, A.: Aggravating O3 pollution due to NOx emission control in eastern China, Science of the total environment, 677, 732-744, doi:10.1016/j.scitotenv.2019.04.388, 2019.*

Author's changes in manuscript: *Lines 466–468.*

(20) Line 234: Wrong reference: Diallo et.al (2018) is from 2019.

Author's response: *It was changed to "Diallo et.al (2019)".*

Author's changes in manuscript: *Line 278.*

Figures

(21) Figure 2: Adding time period and specifying the months for season as the same way as figure 5.

Author's response: *We gave the time period and months for seasons in Figure 2.*

Author's changes in manuscript: *Lines 511–515.*

(22) Figure 3: It's not clear. Replace Mean (DU) by MO3, "Trend" by TO3 and 'Relative trend' by Trel. Add new columns in the table instead of using ',' to separate fields.

Author's response: *We Replace Mean (DU) by MO3, "Trend" by TO3 and 'Relative trend' by Trel in Table 1 which consistent with the text.*

Author's changes in manuscript: *Table 1.*

References

(23) Wang. et. al (2009b): Include this reference after Wang. et. al (2009a)

Author's response: *Wang, T., Wei, X. L., Ding, A. J., Poon, C. N., Lam, K. S., Li, Y. S., Chan, L. Y. and Anson, M.: Increasing surface ozone concentrations in the background atmosphere of Southern China, 1994–2007, Atmos. Chem. Phys., 9(16), 6217–6227, doi:10.5194/acp-9-6217-2009, 2009a.*

Wang, X. S., Li, J. L., Zhang, Y. H., Xie, S. D., and Tang, X. Y.: Ozone source attribution during a severe photochemical smog episode in Beijing, China. Sci. China Ser. B, 52, 1270–1280, doi:10.1007/s11426-009-0137-5, 2009b.

Author's changes in manuscript: *Lines 474–476 and lines 482–484.*

Comments from Referee 3

(1) Suggestions for technical corrections or reasons for rejection

General comment: the ozonesonde location is very near the latitude of the tropopause break, which means it experiences both high-altitude tropopause (i.e., tropical) and low-altitude tropopause (i.e., extratropical) environments throughout the year. One very likely contributing factor to the changes discussed throughout is the time-evolving location relative to the tropopause break and how that relates to i) the degree of stratosphere-to-troposphere transport influence, and ii) the diagnosed UTLS ozone trend. I would recommend the authors include a detailed tropopause analysis to accompany their ozone assessment in order to whittle down the sources of uncertainty in the interpretation of the cause(s) and significance of observed changes. This has implications for the analysis and discussion presented throughout the manuscript.

Author's response: *Thanks for the suggestion. Tropopause analysis would be a good way to investigate the variation of stratosphere-to-troposphere transport. We discussed tropopause in the last section.*

Author's changes in manuscript: *Lines 275–282.*

(2) lines 34-35: should also mention transport of tropospheric ozone into the lower stratosphere.

Author's response: *The transport of tropospheric ozone into the lower stratosphere is mentioned in revised paper.*

Author's changes in manuscript: *Lines 33–35.*

(3) Line 50: "2008–-2012", extra character/hyphen here

Author's response: *Yes. We deleted the extra character.*

Author's changes in manuscript: *Line 51.*

(4) Lines 105-106: Also worth mentioning the potential influence of tropospheric chemistry at this level.

Author's response: *In the mid-troposphere, transport from stratosphere is the main resource of CLaMS ozone because of the lack of tropospheric ozone chemistry in the model.*

Author's changes in manuscript: *Lines 135–137.*

(5) Lines 170-175: If statistical significance was not determined, then I recommend not using the term "significance". Instead, terms like "most apparent" or "largest" could be used to get the same points across without introducing confusion since the meaning of "significant" is more subjective here. The same applies for other places in the paper where "significant" is used.

Author's response: *We replaced the "significant" by "apparent" and "largest".*

Author's changes in manuscript: *Line 213.*

Comments from Referee 3 in the interactive discussion:

General Comments:

(1) The trend quantification in Section 5 is quite underwhelming. It is void of any statistical significance testing, which is necessary to diagnose meaningful changes. Moreover, trends are diagnosed for relatively short time periods, which provides little confidence in the result and leads to overfitting where there is substantial year-to-year variability. Confidence intervals (e.g., 95 and 99 percent) would be espe cially helpful here to demonstrate the degree to which diagnosed long-term changes (and reversals from the first to second half of the time period) are meaningful. Without statistical evaluation here, clear conclusions cannot be made for the diagnosed trends and the use of the term "significant" throughout the paper is inappropriate.

Author's response: *We add statistical significance testing in Section 5. Most ozonesonde trends in lower troposphere and mid-troposphere before 2012 passed the 95% significance criterion. Since there are fewer samples after 2012, some trends only passed the 90% significance criterion.*

Author's changes in manuscript: *Table 1.*

(2) There are at least two claims based on comparison of observations and CLaMS output that are not justified based on the analyses conducted. First, the authors claim at lines 124-125 that CLaMS overestimates transport from the stratosphere to the troposphere. This is based on comparing ozone concentrations in the model to that observed and assuming a certain missing control by tropospheric chemistry. Without additional analysis (or citations to other more thorough evaluation), I do not find this claim to be justified based on the analysis presented in this paper. Second, at lines 261-266 it is argued that a reduction in stratospheric ozone found near the ozonesonde location in CLaMS is a result of ENSO, but there are certainly several alternative explanations for this change that are not acknowledged. Notably, Beijing is near the climatological mean latitude of the tropopause break (the sharp discontinuity in tropopause altitude from tropics to extratropics). Latitudinal migrations of the tropopause break could result in Beijing being more on the tropical side in later years, thus less exposed to downwelling stratospheric air. The latter can certainly be evaluated using the CLaMS output.

Author's response: *We claim that CLaMS overestimates transport from the stratosphere to the troposphere based on not only the comparison between ozonesonde and CLaMS, but also a study by Konopka et al. (2019). Although the current transport scheme in CLaMS shows a good ability to represent transport of tracers in the stably stratified stratosphere, there are deficiencies in the representation of the effects of convective uplift and mixing due to weak vertical stability in the troposphere. We give more explanation here to make it clearer to understand.*

For the second comment, tropopause could also be a reason for the reduction in ozone in UTLS. We add a citation (Chen et al., 2019) in which the tropopause trend across China is investigated. The result shows an upwards trend of tropopause in most part of China including the North China Plain. The uplifted tropopause result in the reduction of ozone in UTLS. Chen, X., Guo, J., Yin, J., Zhang, Y., Miao, Y., Yun, Y., Liu, L., Li, J., Xu, H., Hu, K. and Zhai, P.: Tropopause trend across China from 1979 to 2016: A revisit with updated radiosonde measurements, International Journal of Climatology, 39(2), 1117– 1127. https://doi.org/10.1002/joc.5866, 2019.

Author's changes in manuscript: *Lines 138-140 and Lines 275-278.*

(3) There is substantial repetition in the discussion of the time-evolving role of NOx. Namely, a succinct analysis and discussion is given and then followed shorlty after by a less clear rehashing of essentially the same points while pointing to other work – lines 151-155. Perhaps the authors intend to make a slightly different point, but this is not clear.

Author's response: *We modified this part to make it clearer. First, we cite some studies which revealed the reduction of NOx in many places of China in recent years. Then, we use OMI data to show the long-term variation of tropospheric NO2 over Beijing. After talk about NOx, we were thinking to cite studies to show the change of other precursors. But now we move them to the Discussion and conclusion Section. In Section 4, we revised the description as "The variations in the precursors of tropospheric ozone have dominant roles in the long-term variability of tropospheric ozone. In recent years, the Chinese government has started to invest time and resources in controlling air pollution. A review of 20 years of air pollution control in Beijing (UN Environment, 2019) reported reductions in NOx during the period 2013–2017. A clear decreasing trend in NOx emissions has been observed since 2012 (van der A et al., 2017). Zheng et al. (2018) also reported that emissions of NOx in China decreased by 21% during the time period 2013–2017. Wang et al. (2019) reported that NOx emissions in eastern China decreased by â´Lij25% from 2012 to 2016. Tropospheric NO2, one of the precursors of tropospheric ozone, has gradually decreased over Beijing in recent years (Vu et al. 2019). We use the tropospheric column of NO2 from OMI to discuss the influence of precursors on the long-term variation of tropospheric ozone in Beijing. The deseasonalized tropospheric columns of NO2 measured by OMI from 2004 to 2018 are shown in Figure 5. Tropospheric NO2 was increasing from 2004 to 2010, especially in 2009, leading to the increase of ozone in lower and upper troposphere. As Chinese government start to control air pollutions, tropospheric columns of NO2 were in a condition of relatively large fluctuation in the period of 2010-2013. Tropospheric NO2 over Beijing experienced two major fluctuations in this period, as shown by Gaussianweighted means. Then tropospheric NO2 was gradually decrease since 2013, result in the hiatus of ozone increase in lower and upper troposphere." In the discussion and conclusions section, the description is revised as "The Chinese government has taken action to reduce air pollution since 2012 and the precursors of ozone have decreased gradually in recent years (Vu et al., 2019; Zheng et al., 2018). We show the reduction in tropospheric NO2 by using OMI measurements. Other studies have also shown that the other O3 precursors have decreased in recent years in China, including not only NOx but also SO2 and VOCs (Ma et al., 2016; van der A et al., 2017; Li et al., 2017; UN Environment, 2019; Wang et al., 2019). These reduction in ozone precursors are considered to be the main reason for the hiatus in the increase in ozone in the troposphere, especially in the lower troposphere."*

Author's changes in manuscript: *Lines 153-168, 269-274 and 304-308.*

Specific Comments:

(1) Lines 34-35: delete "which transports stratospheric ozone into the troposphere and tropospheric ozone into the lower stratosphere" as it repeats the previous words in this sentence. Also, this sentence is incomplete. What about the exchange of ozone between the stratosphere and troposphere?

Author's response: *This sentence is changed as "The exchange of ozone between the stratosphere and the troposphere is also important to bring ozone into troposphere (Dufour et al., 2010, 2015; Neu et al., 2014)".*

Author's changes in manuscript: *Lines 33-35.*

(2) Line 66: "relative" should be "previous"

Author's response: *"relative" is replaced by "previous".*

Author's changes in manuscript: *Line 67.*

(3) Line 74: The accuracy and precision of the ozonesonde data should be listed here.

Author's response: *The mean difference in the ozone partial pressure between the IAP and ECC ozonesondes was sphere. The correlation coefficients for profiles by IAP ozonesondes and the ECC are greater than 0.99 (Xuan et al., 2004). The total ozone columns measured by the IAP ozonesonde and the Brewer spectrophotometer were in good agreement with a relative difference of 6%. For the total ozone column, the relative difference and correlation coefficient between IAP ozonesonde and Brewer instrument were 6% and 0.94.*

Author's changes in manuscript: *Lines 71-76.*

(4) Line 86: It is not clear where the "D" comes from in the ASAD acronym. A quick Google search shows that this should be defined here as"A Selfcontained Atmospheric chemistry coDe (ASAD)". Please revise.

Author's response: *We revise it as "A Selfcontained Atmospheric chemistry coDe (ASAD)".*

Author's changes in manuscript: *Line 87.*

(5) Lines 140-148 and Figure5: Details on OMI data used belong in the data and methods section.

Author's response: *We give more details on OMI data in Section 2.*

Author's changes in manuscript: *Lines 101-111.*

(6) Figure 6: It is not clear what exactly is being done to/with the data. Are the time series based on a three-month average of monthly means or something else?

Author's response: *No, the data is still monthly. That means we have 3 samples each year for each season. To make it clearer, we revised it as "Figure 6. Monthly mean column ozone (DU) from the ozonesonde observations (black) and CLaMS simulations (red) in four seasons. Trends of column are calculated before and after the sudden decrease of ozone in 2011-2012. There are 3 monthly values in each year for each season."*

Author's changes in manuscript: *Lines 536-538.*

Comments from Referee 4

Suggestions for technical corrections or reasons for rejection

The presented long term ozone sonde data record itself is of interest for the community. However, the analysis does not meet the scientific standards from my point of view.

The most obvious ones are below:

(1) There are no significance or error estimates of the 'trends' (the authors state, that the the time series is too short for this, which is weird, since the focus of the paper is on trends)

Author's response: *we already added the significance and error estimates in the previous and present version of our paper.*

Author's changes in manuscript: *Figures 3-5 and Table 1.*

(2) The selection of time intervals to calculated trends is rather arbitrary and different in different altitudes. No criteria are given and it is speculated on common reasons for this.

Author's response: *The selection of time intervals to calculate trends is indeed different in different altitudes, but the criteria are not arbitrary. The whole time series is divided by the time of sudden decrease of ozone in Fig. 3 and Fig. 4. It is the time period of sudden decrease that varies with altitudes. The time period of sudden decrease is defined as the period in which the most significant decrease in Gaussian-weighted deseasonalized ozone was observed. So, the time intervals are different in different altitudes dut to the different times of sudden decrease. The time periods in Fig. 6 are consistent with the time intervals in Fig. 3 and Fig. 4. We gave a clearer description in Fig. 6. We gave a clearer description in Fig. 6.*

Author's changes in manuscript: *Lines 150–152. Figure 6.*

(3) There is no link to the meteorology: Does e.g. the monsoon (or a change of the circulation) play a role?

Author's response: *Meteorology may play a role based on the premise that a sudden change of meteorology has been observed around 2012. We didn't find such change in monsoon or circulation so far. Even the related change of meteorology has been found, how can it influence ozone is still a complicated question which is impossible to comprehensively show in this paper. We add discussion about ENSO and tropopause in the discussion and conclusions section. There may be many other meteorological factors which affect variation of ozone, but they are not the key points of this paper and less important than precursors and transport. It is impossible for anyone to investigate all of them in one single paper. We will show the influence of meteorology in our future study when there is enough evidence to support the link between meteorology and ozone variation.*

Author's changes in manuscript: *Lines 275–282.*

(4) They use CLaMS model, which has no tropospheric chemistry to compare ozone (as stated correctly by the authors). The model has no tropospheric chemistry, nor convection included. How can you use this for tropospheric ozone comparisons?

Author's response: *CLaMS is not used to simulate tropospheric ozone and to compare with ozonesonde. We want to isolate and quantify the long-term trends caused by transport from the stratosphere*

and by tropospheric chemistry. No tropospheric chemistry in CLaMS makes it a very qualified model for this work.

(5) Since CLaMS has no tropospheric ozone chemistry they conclude, that differences between the CLaMS ozone variability and observed ozone is driven by stratospheric processes. They do not discuss any reason for stratospheric transport changes or meteorology.

Author's response: *Since CLaMS has no tropospheric ozone chemistry, CLaMS simulation shows the result caused only by stratospheric transport. The difference between the CLaMS ozone variability and observed ozone shows the effect of tropospheric chemistry.*

The reasons for the changes of stratosperhic transport or meteorology are complicated. We are also studying these reasons until a satisfactory result has been achieved. We discussed the possible reasons (ENSO and tropopause) in the last section.

Author's changes in manuscript: *Lines 275–282.*

(6) The do not discuss other ozone sources, which are not included in the model (e.g. lightning NOx, the role of convection, which is not included by CLaMS, impact of wild fires and long range transport e.g. from siberia).

Author's response: *We add the result of NO2 from OMI to discuss the influence of ozone precursors. Actually, the convection is included in CLaMS.*

Author's changes in manuscript: *Lines 161–168.*

(7) They use the model and conclude on stratospheric influence without showing a unique stratospheric component (low humidity, other stratospheric species, etc.) to support their conclusion.

Author's response: *CLaMS can simulate not only ozone but also other component. However, we have only ozone measurement by ozonesondes. So, there is no observation of other component can be compared with CLaMS. As for water vapor, we think it not a very good tracer to study stratospheric influence, because it may also come from descending of the Hadley cell.*

Tao, M., Pan, L. L., Konopka, P., Honomichl, S. B., Kinnison, D. E., & Apel, E. C. (2018). A Lagrangian model diagnosis of stratospheric contributions to tropical midtropospheric air. Journal of Geophysical Research: Atmospheres, 123, 9764–9785. https://doi.org/10.1029/2018JD028696.

(8) They do not show any other chemical quantity, which is related to tropospheric (chemical) ozone change, at least satellite data could be compared.

Author's response: *We add NO2, one of the most important ozone precursors which related to ozone change, in revised paper.*

Author's changes in manuscript: *Lines 161–168and Figure 5.*

Comments from Referee 4 in the interactive discussion:

The paper uses ozone soundings in eastern China from 2002 to 2020 to analyze trends in different altitudes, 0-3km, 3-9km and 9-15km. The authors conclude on different trends at these altitudes and particularly increasing

trends before 2011-2012 in the troposphere and decreasing trends afterwards. In the lower stratosphere observations show a slight increase before 2012 and constant values afterwards and some "superposition" of lower tropospheric and stratospheric trends in the free troposphere. To explain potential reason for these trends they compare the data with the Chemical Lagrangian Model of the Stratosphere (CLAMS). Notably CLAMS it is able to simulate the stratospheric ozone distribution, but has no tropospheric chemistry. To do the comparison of trends they calculated the relative changes for each period and season and selected the strongest changes of these tendencies (before and after 2011-2012). They compare the ozone tendencies before and after 2011-2012 in both, model and observations. They conclude, that the stratospheric impact also significantly has contributed to the trends in the free troposphere (They termed 'superposition layer'). They also conclude, that trend changes in the emission strengths are the key driver for ozone changes in the lower troposphere corresponding to NO2 observational trends.

The main problem with the paper, which I see is, that it does not use any robust statistical metrics or error estimate. Trends are evaluated over time periods of very few years (2002-2011/2012 and afterwards) and the year to year variability is high. The tendencies, which are shown and discussed remain vague. E.g. Fig 3a) shows a trend of observed O3 of zero DU after 2012 (9-15km), in the conclusions (l.274/275) the negative trend in the stratospheric dominated regime is mentioned. Also the criteria to define time periods of trend changes are not motivated and seem to differ in different plots (Fig.6).

The observations are interesting in some parts, but the most interesting part, which is the change of ozone in the 9-15km layer, remains unexplained and is not analyzed. The authors discuss some links with ENSO without providing additional analyses and make no link to the tropopause location or jet, tropical widening. Potential tropospheric circulation aspects could in principle also play a role changing tropospheric long range transport of air masses with high stratospheric ozone from non-local downward transport.

As such I do not know what the key finding of the paper is. If so, is it the stratospheric change of trend? Is it its impact on the free troposphere? Given the methods and the coarse analysis I don't see the manuscript meeting the standards of an ACP publication in its current form, although some observations are interesting.

Author's response: *Thanks for all the comments and suggestions.*

We already added the significance and error estimates. The criteria to define time periods in Fig. 6 are consistent with the time intervals in Fig. 3 and Fig. 4 in which sudden decrease is defined as the period in which the most significant decrease in Gaussian-weighted deseasonalized ozone was observed. The periods of sudden decrease are different in different altitudes, so the time intervals in Fig. 6 are different in different altitudes.

In this paper, we focused on the changes of ozone trend which mainly caused by the change of emission and the sudden decrease in 2011-2012 which is largely related to the transport from stratosphere. The other meteorological reason such as ENSO and tropopause might also related to ozone variation as we discussed in the conclusion section. However, they are not the main points of this paper and less important than emission and stratospheric transport. There may be many other meteorological factors like jet and tropical widening, but obviously it is impossible for anyone to investigate all of them in one single paper. We would like to deeper dive on other mechanisms in the future.

As for the key finding of this paper, we think the dataset itself, the trends it revealed and the sudden decrease are the most innovative parts. Based on the only long-term observed ozonesonde data in North China Plain, we revealed the very interesting changes in tropospheric and lower-stratospheric ozone. We use NO2 form OMI to

show the influence of precursor on the change of trend, and we use CLaMS model to show the influence of stratospheric transport on the sudden decrease of ozone in 2011-2012. All of these make this paper an interesting and relatively complete story which we don't agree to call it "coarse". Does a good paper must contain complicated methods or revealed all possible mechanisms?

We have carefully revised the manuscript according to these suggestions. Our point-to-point responses are listed below:

Major comments:

As I said the data record is interesting, but the analysis is more than coarse. The authors should at least provide some statistical valid metrics for the significance of trends.

(1) There are no significance or error estimates of the 'trends' (the authors state, that the the time series is too short for this, which is weird, since the focus of the paper is on trends)

Author's response: *we already added the significance and error estimates in the present version of our paper.*

Author's changes in manuscript: *Figures 3-5 and Table 1.*

(2) The selection of time intervals to calculate trends seems arbitrary and different in different altitudes. Criteria are not clear and seem to differ (Fig. 6).

Author's response: *the selection of time intervals to calculate trends is indeed different in different altitudes (Fig. 6.). But the criteria are not arbitrary, they are consistent with the time intervals in Fig. 3 and Fig. 4 in which sudden decrease is defined as the period in which the most significant decrease in Gaussian-weighted deseasonalized ozone was observed. The periods of sudden decrease are different in different altitudes, so the time intervals in Fig. 6 are different in different altitudes. We gave a clearer description in Fig. 6.*

Author's changes in manuscript: *Lines 150–152 and Figure 6.*

(3) They should also explain more clearly the role of meteorology when interpreting the seasonally resolved trends in the free troposphere (note that the whole manuscript does not contain any mentioning of the monsoon, convection, tropopause, jets).

Author's response: *after the analysis of the long-term trends and the sudden decrease of ozone. We gave the seasonal trends to show in which seasons the significant changes of ozone are observed. In this part, we think that the precursors are the most important factors for the ozone in the troposphere-dominated range, and the transport greatly affects the ozone in the stratosphere-dominated range. It doesn't mean that we can exclude the meteorological reasons such as monsoon, convection, tropopause and jets. They are not the key points of this paper and less important than precursors and transport. Actually, we mentioned ENSO and tropopause in the discussion and conclusions section. There may be many other meteorological factors which affect variation of ozone, but obviously it is impossible for anyone to investigate all of them in one single paper.*

Author's changes in manuscript: *Lines 275–282.*

(4) They use the CLAMS model, which has no tropospheric chemistry to compare ozone (as mentioned correctly by the authors). How do the authors exclude potential changes of tropospheric ozone sources, circulation changes and long-range transport, which could potentially also lead to different variability and trends? The fact that an incomplete model sometimes agrees with observations, does not automatically exclude other processes, which are not included in the model, to explain the observed ozone tendencies.

Author's response: *CLaMS is not used to simulate tropospheric ozone and to compare with ozonesonde. We want to isolate and quantify the long-term trends caused by transport from the stratosphere and by tropospheric chemistry. There is no tropospheric chemistry in CLaMS which you think it is an incomplete model. However, it is the specialty makes it a very qualified model for this work (to isolate and quantify the trends caused by transport and by tropospheric chemistry). We did not exclude potential changes of tropospheric ozone sources, circulation changes, long-range transport and other unknown reasons, but they are not the key points of this paper. No paper can completely include all factors, especially some of them are still unknown. For this paper, we revealed the trends and the sudden decrease of ozone based on the rare ozonesonde dataset, and we found these changes in ozone are related to NO2 and transport. So far, it is a complete and interesting story. Other mechanisms can be investigated deeper and more complete in future works.*

Comments from editor:

Dear authors, Thanks a lot for your reply. The referee made an important comment that should be replied at the begining of your reply because in my opinion it is very relevant: The analysis is mostly straight-forward, but it is not clear to me if this paper amounts to a substantial contribution. Similar recent studies have diagnosed the observed longterm changes using different datasets and have gone into somewhat greater detail to elucidate the processes responsible and the significance of diagnosed trends. I believe this could be a more meaningful contribution with greater communication of its novelty throughout and a bit deeper dive on the mechanisms responsible for observed changes.

Author's response: *We noticed there are similar recent studies which diagnosed the long-term changes of ozone using different datasets. Most of these datasets are satellite observations or surface measurements. The data quality of satellite observations in troposphere are not as good as in stratosphere. Surface measurements are precise, but only surface O_3 are measured. Compared to satellite data, our ozonesonde observations are more precise with much higher vertical resolution. Compared to surface measurements, we have the profiles from surface to ~30km. So, ozonesonde is the best dataset to investigated the ozone variation not only near surface but also in the whole troposphere and lower stratosphere. However, there are only a few stations have ozonesonde data and some of stations only measured for a short time. Beijing ozonesonde data is the longest observation (since 2002) of the ozone profile over the North China Plain. This dataset is once used in Wang et. al. (2012) to show positive trends in the period of 2002-2010 which raises many concerns (more than 60 citations). Now, we extend the observed time series where we used CLaMS to show the trends after 2010. Fortunately, we found a sudden decrease in 2011 which related to stratospheric transport, and we found negative trends in recent years which mainly due to the reduction of precursors. I think the dataset itself, the trends it revealed and the sudden decrease are the most innovative parts of this paper. As for the mechanisms, we discussed the two main reasons which is responsible for the change of trend and the sudden decrease. We use NO_2 form OMI to show the influence of precursor on the change of trend, and we use CLaMS model to show the influence of stratospheric transport on the sudden decrease of ozone in 2011. There are other precursors which need more data and other possible reasons such as changes in meteorology, but they are not the main point of this paper. It is a huge task to investigate all the mechanisms responsible for the O_3 change. We would like to deeper dive on other mechanisms in the future.*

Reference:

Wang, Y., Konopka, P., Liu, Y., Chen, H., Müller, R., Plöger, F., Riese, M., Cai, Z. and Lü, D.: Tropospheric ozone trend over Beijing from 2002–2010: ozonesonde measurements and modeling analysis, Atmos. Chem. Phys., 12(18), 8389–8399, doi:10.5194/acp-12-8389-2012, 2012.

A List of Major Comments:

	Comments	Referees	Response	Changes
1	significance or error estimates of the 'trends'	1, 3, 4	added the significance and error estimates	Figures 3-5 and Table 1
2	time intervals to calculate trends seems arbitrary and different in different altitudes	4	the criteria are not arbitrary, they are based on the time of sudden decrease which is different in different altitudes. It is defined as the period in which the most significant decrease in Gaussian-weighted deseasonalized ozone was observed.	Lines 150–152.
3	link to the meteorology: e.g. the monsoon, tropopause, a change of the circulation	1, 4	We add the discussion about ENSO and tropopause in the last section. There may be many other meteorological factors, but they are not the key points of this paper and less important than precursors and transport. We will study them in the future, because it is impossible to investigate all of them in one single paper.	Lines 275–282.
4	CLaMS model has no tropospheric chemistry. How can you use this for tropospheric ozone comparisons?	4	CLaMS is not used to simulate tropospheric ozone and to compare with ozonesonde. We want to isolate and quantify the long-term trends caused by transport from the stratosphere and by tropospheric chemistry. No tropospheric chemistry in CLaMS makes it a very qualified model for this work.	
	I advise the author to give more information to eliminate readers' doubt on data quality. Did other ozone measurements in Beijing give similar result? Was there a sudden and persistent change in large-scale dynamics after 2011?	1	The ozonesonde data has been proved reliable, we give more information about ozonesondes and relative references. We think the change in trend is mainly the result of decrease of ozone precursors. So, we add a long-term variation of tropospheric NO_2 from OMI. The huge drop of ozone in	Ozonesonde data is at Lines 64–76. NO_2 from OMI is at Lines 161–168. Sudden decrease

			middle troposphere in 2011-2012 may attribute to the change of transport from stratosphere. Because CLaMS which has no tropospheric ozone chemistry also shows the huge drop. There was no other ozone measurements in Beijing except satellite data which is not better than ozonesonde measurements below 3 km. We discussed the ENSO and tropopause which may be related to the ozone change.	caused by transport is at Lines 284–286. ENSO and tropapause is at Lines 275–282.
	It is better to use a model with reasonable representations of tropospheric ozone chemistry and/or other chemical (e.g., satellite observations of ozone precursors) and meteorology data to better explain the ozone changes.	1	We add the NO_2 from OMI to discuss the influence of precursors on the long-term variation of tropospheric ozone in Beijing. The result shows that the decrease of tropospheric NO_2 plays an important role in the decrease of tropospheric ozone.	The NO_2 from OMI is at Lines 161–168 and in Figure 5.
	The lower tropospheric ozone in the present study appeared to have a small positive trend after the 2011 drop. It is similar to surface ozone increase observed in many urban areas from the Ministry of Ecology and Environment network since 2013, which has been attributed to the nonlinear chemistry of ozone precursors (NOx emission decrease and VOC emission increase) and aerosol decrease, as well as being affected by meteorological variation (see for example, Li et al., 2019; Liu and Wang, 2020).	1	We noticed that O_3 trend is still positive after the 2011 drop, but it is much slower than before due to the reduction of NOx. However, there are other precursors which might be responsible for the small positive trend after the 2011 drop. Thanks for showing us the two papers (Li et al., 2019; Liu and Wang, 2020). We added them when we mentioned the possible reasons of meteorological variation in the discussion and conclusions.	Lines 307–308, 400–401, 271–273 and 469–471.
	It should be clearer highlighted in the introduction that this manuscript is an extension of the work by Wang et al. (2012).	2	We highlighted the extension of the work by Wang et. al. in the introduction and it is clearer now.	Lines 56–61.

	The authors use different names or different nomenclature along the text to refer to the same things. Defining these concepts at the beginning of the paper should be more convenient to avoid repetition of the definition of the different concepts.	2	We checked the nomenclatures, especially the layers and the seasons. We gave the definitions when they were firstly mentioned and deleted the repeated description.	
	The authors claim that CLaMS overestimates transport from the stratosphere to the troposphere. This is based on comparing ozone concentrations in the model to that observed and assuming a certain missing control by tropospheric chemistry. Without additional analysis (or citations to other more thorough evaluation), I do not find this claim to be justified based on the analysis presented in this paper.	3	We claim that CLaMS overestimates transport from the stratosphere to the troposphere based on not only the comparison between ozonesonde and CLaMS, but also a study by Konopka et al. (2019). Although the current transport scheme in CLaMS shows a good ability to represent transport of tracers in the stably stratified stratosphere, there are deficiencies in the representation of the effects of convective uplift and mixing due to weak vertical stability in the troposphere. We give more explanation here to make it clearer to understand.	Lines 138-140
	There is substantial repetition in the discussion of the time-evolving role of NOx.	3	We reorganized the discussion.	Lines 153-168, 269-274 and 304-308.

带格式的：正文

Long-term Variations in Ozone Levels in the Troposphere and Lower Stratosphere over Beijing: Observations and Model Simulations

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Abstract. Tropospheric ozone is both a major pollutant and a short-lived greenhouse gas and has therefore attracted much concern in recent years. The ozone profile in the troposphere and lower stratosphere over Beijing has been observed since 2002 by ozonesondes developed by the Institute of Atmospheric Physics. Increasing concentrations of tropospheric ozone from 2002 to 2010 measured by these balloon-based observations have been reported previously. As more observations are now available, we used these data to analyze the long-term variability of ozone over Beijing during the whole period from 2002 to 2018. The ozonesondes measured increasing concentrations of ozone from 2002 to 2012 in both the troposphere and lower stratosphere. There was a sudden decrease in observed ozone between 2011 and 2012. After this decrease, the increasing trend in ozone concentrations slowed down, especially in the mid-troposphere, where the positive trend became neutral. We used the Chemical Lagrangian Model of the Stratosphere (CLaMS) to determine the influence of the transport of ozone from the stratosphere to the troposphere on the observed ozone profiles. CLaMS showed a weak increase in the contribution of stratospheric ozone before the decrease in 2011–2012 and a much more pronounced decrease after this time. Because there is no tropospheric chemistry in CLaMS, the sudden decrease simulated by CLaMS indicates that a smaller downward transport of ozone from the stratosphere after 2012 may explain a significant part of the observed decrease in ozone in the mid-troposphere and lower stratosphere. However, the influence of stratospheric ozone in the lower troposphere is negligible in CLaMS and the hiatus in the positive trend after 2012 can be attributed to a reduction in ozone precursors as a result of stronger pollution control measures in Beijing.

1 Introduction

Tropospheric ozone is an important pollutant and is detrimental to both human health (WHO, 2006) and the productivity of vegetation (Ainsworth et al., 2012; Emberson et al., 2013; Feng et al., 2015). It is also an important greenhouse gas (IPCC, 2007) and influences radiative forcing (Wang et al., 1976; Lacis et al., 1990; Seinfeld et al., 2006). It is therefore crucial to understand and monitor the long-term variability in tropospheric ozone.

Tropospheric ozone mainly originates from photochemical reactions involving precursors such as nitrogen oxides (NO_x) and volatile organic compounds (VOCs) (Monks et al., 2009; Su et al., 2018; Tan et al., 2018). The exchange of ozone between the stratosphere and the troposphere is also important to bring ozone into troposphere (Dufour et al., 2010, 2015; Neu et al., 2014).

Human activities have significantly increased tropospheric ozone since the industrial revolution as a result of increased concentrations of ozone precursors (Hough and Derwent, 1990; Parrish et al., 2012). The reduction in

surface UV radiation due to high aerosol concentrations has important impacts on the production of photochemical ozone (Deng et al., 2011). Studies have shown a dramatic positive trend in the concentration of tropospheric ozone in China since the 1990s due to rapid economic development and urbanization (Wang et al., 2012; Cooper et al., 2014; Chen et al., 2015; Verstraeten et al., 2015). Increasing ozone concentrations have been observed both at the surface (Cooper et al., 2014; Ma et al., 2016; Wang et al., 2009a) and in the lower troposphere (Ding et al., 2008; Shen et al., 2012; Sun et al., 2016; Wang et al., 2017).

Air quality controls have been implemented in China as a result of the recent increases in atmospheric pollutants, especially in the North China Plain, and the emissions of SO_2 and NO_x have been successfully reduced in recent years (Ma et al., 2016; van der A et al., 2017; Li et al., 2017). Recent studies have shown levelling off/decrease in surface ozone levels in rural areas of eastern China and in outflow of eastern China air masses (Xu et al., 2020; Wang et al., 2019). Using the Infrared Atmospheric Sounding Interferometer (IASI) onboard the European Space Agency's (ESA) MetOp series of polar orbiting satellites, together with surface and ozonesonde measurements, Dufour et al. (2018) reported the trend in tropospheric ozone concentrations over the North China Plain for the time period 2008–2016. They found that there were two distinct periods: (1) 2008–2012 with no significant trend; and (2) 2013–2016 with a significant negative trend in lower tropospheric ozone.

Ozone sounding has been carried out over Beijing on a regular basis since 2002 and is the longest observation period of the ozone profile over the North China Plain (Zhang et al., 2014). As a unique long-term series of ground-based observations, this dataset is the best candidate with which to reveal the long-term variability in tropospheric and lower stratospheric ozone over the North China Plain and especially over Beijing. Wang et al. (2012) used this dataset to show a positive trend in tropospheric ozone during the time period 2002–2010. ~~However, it is not yet known whether the concentration of tropospheric ozone has increased since 2010.~~ As an extension of the work by Wang et. al. (2012), we got more years of data since 2010. In this work, we used the whole time series (2002–2018) of ozonesonde observations to explore the variability in ozone concentrations over Beijing in the last two decades.

2 Data and model

2.1 Ozonesonde

Ozone concentrations from the Earth's surface up to ~30 km were measured over Beijing using an ozonesonde developed by the Key Laboratory of Middle Atmosphere and Global Environment Observation of the Institute of Atmospheric Physics (IAP) (Zhang et al., 2014). The IAP ozonesonde is based on an electrochemical method which is well documented in previous~~relative~~ studies (Wang et al., 2003; Xuan et al., 2004; Zheng and Li, 2005). The ozonesonde has previously been compared with the widely used electrochemical concentration cell (ECC) developed by Komhyr (1969) and the Brewer spectrophotometer (Zhang et al., 2014) and was able to capture the ozone profile. The mean difference in the ozone partial pressure between the IAP and ECC ozonesondes was <0.5 mPa in the troposphere and <1 mPa in the lower stratosphere. The correlation coefficients for profiles by IAP ozonesondes and the ECC are greater than 0.99 (Xuan et al., 2004). The total ozone columns measured by the IAP ozonesonde and the Brewer spectrophotometer were in good agreement with a relative difference of 6%. For the total ozone column, the relative difference and correlation coefficient between IAP ozonesonde and Brewer

instrument were 6% and 0.94. The ozonesonde data has been used to validate satellite measurements (Bian et al., 2007) and model products (Wang et al., 2012).

The ozone profiles have been observed about once a week since 2002 at 14:00 local time (06:00 UTC). In some intensive observation periods (e.g., 24 March to 10 April 2003), ozonesondes were launched every day. However, there was no observation (gaps in Figure 1) in two periods (July 2008 and January 2013). The ozonesondes were released from Beijing Observatory (39.81°N, 116.47°E; 31 m above sea level). The maximum altitude for the ozonesonde profile, which depends on the altitude at which the balloons burst, is between 25 and 35km.

批注 [Y1]: This part has been modified and reorganized.

2.2 Chemical transport model

The Chemical Lagrangian Model of the Stratosphere (CLaMS v1.0) chemical transport model was used to quantify the variation in tropospheric ozone caused by transport from the stratosphere. The CLaMS contains a comprehensive set of reactions of relevance to the stratosphere, including full chlorine and bromine chemistry, 36 chemical species, and 115 reactions (including 27 photolysis and 11 heterogeneous reactions) (McKenna et al., 2002). The model chemistry integrations are based on A Selfcontained Atmospheric Chemistry coDe (ASAD) Carver et al. (1997). The chemical reactions (gas phase and photolysis) are summarized by McKenna et al. (2012). Based on a Lagrangian representation, CLaMS is well suited to simulations of tracer transport (McKenna et al., 2002, Konopka et al., 2004, 2019).

批注 [Y2]: We add more information about CLaMS chemistry.

We used the 40-year CLaMS transient simulation starting on 1 January 1979 and driven by horizontal winds and the diabatic heating rates (vertical velocities) derived from the ERA-Interim reanalysis (Dee et al., 2011). The configuration and model initialization followed the model setup described in Wang et al. (2012) and Pommrich et al. (2014) (100 km horizontal/400 m vertical resolution around 380 K). The first 10 years of the CLaMS transient simulation can be considered as the model spin-up time. To isolate and quantify the long-term trend caused by transport from the stratosphere, a CLaMS simulation without ozone chemistry in troposphere is considered. The ozone values in the lowest model layer were set to zero. CLaMS comprises three main modules: Lagrangian advection; mixing; and stratospheric chemistry. Because there is no tropospheric photochemistry in CLaMS, the tropospheric ozone simulated by CLaMS mainly descends from the stratosphere. The daily output of CLaMS was interpolated at the ozonesonde locations in Beijing for all observed profiles.

2.3 Nitrogen Dioxide from OMI

To discuss the long-term variation of tropospheric ozone precursor in Beijing, we use the version 3 of Aura Ozone Monitoring Instrument (OMI) Nitrogen Dioxide (NO_2) standard product (Krotkov et al., 2018). NO_2 is an important chemical species in troposphere where it is a precursor to ozone production. OMI is a contribution of the Netherlands's Agency for Aerospace Programs (NIVR) in collaboration with the Finnish Meteorological Institute (FMI) to the Aura mission. The Earth is viewed in 740 wavelength bands along the satellite track with a swath large enough to provide global coverage in 14 orbits (1 day). Due to its unprecedented spatial resolution and daily global coverage, OMI plays a unique role in measuring trace gases important for the ozone layer, air quality, and climate change (Levelt et al., 2018). It measures the total ozone and other atmospheric parameters related to ozone chemistry and climate such as NO_2, SO_2, and aerosols. In this study, we select OMI tropospheric columns of NO_2 one degree around Beijing.

3 Comparison between the ozonesonde data and CLaMS simulation

The concentration of tropospheric ozone has strong seasonal variations with a minimum in winter and a maximum in summer. To better estimate the ozone trend, we calculated the contribution of each month to the annual O_3 ~~used the average percentage method~~ to remove the seasonality from the time series, leaving the deseasonalized ozone data. Figure 1 therefore shows the deseasonalized ozone mixing ratio measured by the IAP ozonesondes and simulated by CLaMS in the troposphere and lower stratosphere during the time period 2002–2018. The ozonesonde observations (Figure 1a) show that the concentrations of tropospheric ozone increased in the period 2002–2012. This positive trend is consistent with the study of Wang et al. (2012) in which the same ozonesonde data were used. They suggested, in agreement with other studies (Wang et al., 2006; Wang et al., 2009b; Chou et al., 2009), that photochemical ozone production is the primary reason for the increase in ozone concentrations in the troposphere. A sudden decrease in the ozone mixing ratio occurred in the upper troposphere and lower stratosphere (UTLS) from late 2011 to early 2012. The concentrations of tropospheric ozone have not increased since 2012. The tropospheric ozone mixing ratios have remained stable and are almost equal to the levels in 2005–2006.

The ozone mixing ratio simulated by CLaMS (Figure 1b) captures the main characteristics of the deseasonalized ozonesonde observations in the mid-troposphere (3–9 km) and the UTLS (9–15 km). In particular, the CLaMS ozone mixing ratio below 10 km in the period 2009–2012 was larger than the ratio before and after this period. However, the CLaMS simulations in the lower troposphere (0–3 km) were much smaller than the observations.

To quantify the differences between the ozonesonde measurements and the CLaMS simulations, Figure 2 shows the correlations between the respective partial columns (9–15, 3–9 and 0–3 km) in four seasons (winter: December–January–February; spring: March–April–May, summer: June–July–August and autumn: September–October–November). In the lower troposphere ~~(0–3 km)~~, the ozone columns simulated by CLaMS were much smaller than the those measured by the ozonesonde (Figure 2c). This is because there is no tropospheric ozone chemistry, the main source of ozone in the lower troposphere, in CLaMS (Monks et al., 2009). In the mid-troposphere, transport from stratosphere is the main source of CLaMS ozone because of the lack of tropospheric ozone chemistry in the model. The CLaMS simulations in the mid-troposphere are much closer to the ozonesonde measurements (Figure 2b). CLaMS overestimates the transport of ozone from the stratosphere to the troposphere, which is strongest in spring. This is because CLaMS has deficiencies in the representation of the effects of convective uplift and mixing due to weak vertical stability in the troposphere (Konopka et al., 2019). The destruction of ozone in the mid-troposphere is not completely included in the model as a result of the absence of tropospheric ozone chemistry. A larger production of ozone is expected as a result of reactions involving water vapor, hydrogen peroxy and hydroxyl radicals (Stevenson et al., 2006). The CLaMS simulations in the UTLS agree well with the ozonesonde observations and only slightly overestimate the observations in spring (Figure 2a).

4 Long-term variations in ozone concentrations

To quantify the long-term variations in ozone concentrations, the deseasonalized partial column of ozone observed by the ozonesonde and simulated by CLaMS are shown in Figures 3 and 4 for the lower troposphere, mid-troposphere and UTLS. Figure 6 explores the seasonal dependence of the respective partial columns and their trends. Based on the ozonesonde observations, two features determine the variations in ozone concentrations after

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2012: (1) the sudden decrease in ozone from late 2011 to early 2012; and (2) the hiatus in the positive trend after 2012. The sudden decrease is defined as the period in which the most significant decrease in Gaussian-weighted deseasonalized ozone was observed (the period between the two blue dashed lines).

The variations in the precursors of tropospheric ozone have dominant roles in the long-term variability of tropospheric ozone. In recent years, the Chinese government has started to invest time and resources in controlling air pollution. A review of 20 years of air pollution control in Beijing (UN Environment, 2019) reported reductions in NO_x during the period 2013–2017. A clear decreasing trend in NO_x emissions has been observed since 2012 (van der A et al., 2017). Zheng et al. (2018) also reported that emissions of NO_x in China decreased by 21% during the time period 2013–2017. Wang et al. (2019) reported that NO_x emissions in eastern China decreased by ~25% from 2012 to 2016. Tropospheric NO_2, one of the precursors of tropospheric ozone, has gradually decreased over Beijing in recent years (Vu et al. 2019).

We use the tropospheric column of NO_2 from OMI to discuss the influence of precursors on the long-term variation of tropospheric ozone in Beijing. The deseasonalized tropospheric columns of NO_2 measured by OMI from 2004 to 2018 are shown in Figure 5. Tropospheric NO_2 was increasing from 2004 to 2010, especially in 2009, leading to the increase of ozone in lower and upper troposphere. As Chinese government start to control air pollutions, tropospheric columns of NO_2 were in a condition of relatively large fluctuation in the period of 2010-2013. Tropospheric NO_2 over Beijing experienced two major fluctuations in this period, as shown by Gaussian-weighted means. Then tropospheric NO_2 was gradually decrease since 2013, result in the hiatus of ozone increase in lower and upper troposphere.

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Even if a massive reduction in the precursors of tropospheric ozone results in a local decrease in ozone in the lower troposphere, the transport of ozone from the stratosphere to the troposphere is widely considered to be an important source of tropospheric ozone. We used the CLaMS simulations to show the role of the transport of ozone from the stratosphere in modulating the concentration of tropospheric ozone (Figure 4). Because there is no photochemical reaction in the troposphere in CLaMS, the simulated variations in tropospheric ozone can only be transported from the stratosphere. As a result, the ozone columns in the lower troposphere simulated by CLaMS (Figure 4c) are much smaller than those measured by the ozonesonde (Figure 3c). The observed sudden decrease in ozone in the mid-troposphere and UTLS is also clear in the CLaMS simulations, which means that this decrease originates in the stratosphere. Because the decrease in ozone in the stratosphere and the start of pollution control measures in Beijing occurred at roughly the same time, we have to separate the two variations to understand the trends in ozone concentrations. An exploration of the seasonal dependence of the respective partial columns (Figure 6) will help to narrow down this problem.

Tropospheric ozone chemistry dominates the trends in the lower troposphere ~~(0–3 km)~~ in summer and autumn. The contribution in CLaMS is so small here that any stratospheric influence can be neglected. We call this range the “troposphere-dominated range”. By contrast, the stratospheric influence is dominant in the UTLS ~~(9–15 km)~~ in winter and spring and the tropospheric contribution can be ignored. We call this range the “stratosphere-dominated range”. All the other combinations of seasons and altitudes are a superposition of the troposphere- and stratosphere-dominated ranges and we call such combinations the “superposition range”.

The sudden decrease in ozone concentrations in the troposphere-dominated range in 2011–2012 ended the positive ozone trend that had been observed since 2002 and was most prominent in lower troposphere during summer and autumn (Figure 6g and 6j). The trend after the decrease was almost neutral, indicating that air pollution control

measures effectively reduced the concentration of ozone in the lower troposphere. In the stratosphere-dominated range, both the ozone trends observed by the ozonesonde and those simulated by CLaMS became negative after 2013, most clearly during winter and spring in ~~9–15 km~~UTLS (Figure 6c and 6f). In the superposition range, most of the observed increasing trends weakened rapidly after 2012 (all other panels in Figure 6). Almost all the trends simulated by CLaMS became negative. The increasing trend simulated in the mid-troposphere by CLaMS in spring was almost parallel to the trend observed by the ozonesonde before the decrease (Figure 6e). However, the trend simulated by CLaMS became negative after the decrease when the trend observed by the ozonesonde was still slightly positive due to the upward influence of the positive trend in the lower troposphere. Thus changes in the trends in the superposition range can only be understood as an interaction between the impact of air pollution control and the changing influence of the stratosphere.

Table 1 summarizes the (linear) ozone trends before and after 2012 in ozone concentrations calculated for all four seasons. The first column gives the mean values of ozone (M_{O3}, units: DU) observed by the ozonesonde and simulated by CLaMS. All the numbers are broken down to the altitude range and season considered. The ozone trends (T_{O3}) are calculated in DU/year and the relative ozone trends (T_{rel}) are defined as the percentage of T_{O3} in M_{O3}, i.e., $T_{rel} = T_{O3}/M_{O3}$ (%).

The change in the ozone trend (Δ_{rel}) was calculated as the difference between the relative trends after and before the decrease, i.e., Δ_{rel} (%) = T_{rel} (after the decrease) − T_{rel} (before the decrease). For example, in the lower troposphere, where the most significant change in ozone occurred in the autumn, the mean concentration of ozone was 13.18 DU. The trend was 3.34 DU/year (25.3% relative to 13.18 DU) before the decrease and −0.33 DU/year (−2.5% relative to 13.18 DU) after the decrease. The reversal of the relative trend can therefore be quantified as the absolute value of Δ_{rel}, i.e., $|\Delta_{rel}| = |-2.5\% - 25.3\%| = 27.9\%$.

In this paper, we only discuss time evolution of the ozone columns with absolute values of Δ_{rel} >20%, which typically describe the reversal in trend from positive before 2012 to neutral or negative after 2012. Thus, the most apparent change in the lower troposphere occurred in autumn (27.9%). In the mid-troposphere, the largest changes in trend were observed in spring (19.6% by ozonesonde and 47.7% by CLaMS) and autumn (27.4% by ozonesonde and 36.2% by CLaMS). In the UTLS, the highest values were observed in spring (23.0 and 34.2% by ozonesondes and CLaMS, respectively).

5 Quantified ozone trends

The quantified trends show us how significant the variations in ozone were over Beijing during the time period 2002–2018. The trend after 2012, the period and the magnitudes of the decrease varied with the atmospheric layers (Figure 3). The ozone columns measured by ozonesonde in the lower troposphere showed an abrupt decrease of 5 DU from late 2011 to mid-2012, which is estimated by the Gaussian-weighted means of the deseasonalized ozone concentrations (red curves). The increasing trends, estimated as 0.06 DU/month before the decrease, slowed to 0.02 DU/month (Figure 3c). A similar magnitude of decrease occurred in the mid-troposphere ~~(3–9 km)~~ from mid-2011 to 2012 (Figure 3b). The hiatus in the increase of ozone was more pronounced at this level. The ozone column in this level showed a rate of increase of 0.08 DU/month before the decrease and this positive trend became neutral (0 DU/month) after the decrease. The mean ozone columns in both the lower and mid-troposphere after 2012 returned to levels almost equal to the columns in 2005–2006. The decrease in the ozone column in the UTLS (Figure 3a) occurred in a period from mid-2011 to the end of 2011 and the ozone levels recovered to those

observed before the decrease in early 2012. The increasing trend of 0.05 DU/month became neutral (0 DU/month) after the decrease.

The ozone trend simulated by CLaMS was almost zero (-0.01 DU/month) in the UTLS until a sudden decrease in 2011. The concentration of ozone in the UTLS then decreased at a rate of −0.11 DU/month. In the mid-troposphere, where ozone is a result of downward transport, the increasing trend was 0.05 DU/month before the decrease (Figure 4b). This increasing trend was slower than its counterpart measured by the ozonesonde (0.08 DU/month) as a result of the absence of tropospheric ozone photochemistry in CLaMS. For the same reason, instead of remaining neutral as in the ozonesonde measurements (Figure 3b), the CLaMS ozone in the mid-troposphere decreased at a faster rate of −0.07 DU/month after the decrease in 2011 (Figure 4b). The ozone columns simulated by CLaMS in the lower troposphere (Figure 4c) were much smaller than those measured by the ozonesonde (Figure 3c) as a result of the absence of tropospheric photochemistry. This comparison indicates that the decrease in ozone and the hiatus in the increase of ozone in this troposphere-dominated range is the result of the air pollution control measures started by the Chinese government.

The increasing trends in the troposphere-dominated range became slower after the decrease in 2011–2012 (Δ_{rel} = −17.3% in summer and −27.9% in autumn). The trends changed from 3.91 to 0.39 DU/year in summer and from 3.34 to −0.33 DU/year in autumn, proving the influence of air pollution control measures on the hiatus in the increase in ozone. The positive ozone trends observed by ozonesondes in the stratosphere-dominated range became negative, with Δ_{rel} = −9.6% in winter and −23.0 in spring. The changes in the trends simulated by CLaMS were more dramatic (Δ_{rel} = −23.0% in winter and −34.2% in spring).

Most of increasing trends weakened rapidly (negative values of Δ_{rel}) in the interaction range or even began to decrease after 2012, except for the trend in the lower troposphere in winter, which increased by 9.4%. Of all the interaction ranges, the most significant change in ozone concentrations occurred in the mid-troposphere in spring and autumn. These trends observed by ozonesondes changed by about 5 DU/year with Δ_{rel} = −19.6% in spring and −27.4% in autumn. Their counterparts simulated by CLaMS changed even more dramatically, with Δ_{rel} = −47.7% in spring and −36.2% in autumn. All the trends simulated by CLaMS became negative, except in winter in the mid-troposphere when the trend was 0.67 DU/year. However, although they decreased a lot after the decrease in ozone concentrations, most of the actual trends simulated by ozonesonde were still positive. These comparisons between the ozonesonde and CLaMS results shows that the concentrations of ozone should have decreased since 2012 if we only consider the influence of transport from the stratosphere. However, the actual trends after 2012 in the mid-troposphere did not decrease as much as in CLaMS because they were affected by the much slower, but still increasing or almost neutral, trends in the lower troposphere.

In general, the quantified ozone trends revealed the hiatus in the increase in ozone over Beijing. Although influenced by chemistry and stratospheric transport, the changes in the ozone trend varied with both altitude and season. In most situations, the increase in ozone has been moderated since 2012.

6 Discussion and conclusions

We observed tropospheric and lower stratospheric ozone columns in Beijing once a week from 2012 using ozonesondes developed by the IAP. Using these data, Wang et al. (2012) found a positive ozone trend during the time period 2002–2010. We extended these data to 2018 and found that the evolution of this trend after 2010 was

strongly determined by two factors: (1) a sudden decrease in mainly stratospheric ozone from late 2011 to early 2012; and (2) a decrease in mainly tropospheric ozone caused by reduction in air pollution in the Beijing region.

The Chinese government has taken action to reduce air pollution since 2012 and the precursors of ozone have decreased gradually in recent years (Vu et al., 2019; Zheng et al., 2018). We show the reduction in tropospheric NO_2 by using OMI measurements. Other studies have also shown that the other O_3 precursors have decreased in recent years in China, including not only NOx but also SO2 and VOCs (Ma et al., 2016; van der A et al., 2017; Li et al., 2017; UN Environment, 2019; Wang et al., 2019). These reduction in ozone precursors are considered to be the main reason for the hiatus in the increase in ozone in the troposphere, especially in the lower troposphere.

The decrease in stratospheric ozone has a more global origin. Chen et al. (2019) investigated the long-term variation (1979-2016) of tropopause in China by using the newly released quality-controlled radiosonde data from China Meteorological Administration. The result shows an upwards trend of tropopause in most part of China including the North China Plain. The uplifted tropopause may result in the reduction of ozone in UTLS. Diallo et al. (2019) reported that a strong La Niña phase of the El Niño–Southern Oscillation around 2011 caused an anomalous increase in ozone in the lower stratosphere of the tropics. The related weaker upwelling in the tropics coincided with weaker downwelling in extra-tropical regions and, as a consequence, less transport of ozone from the stratosphere to the troposphere, especially at the latitude of Beijing. Because there is no tropospheric ozone chemistry in CLaMS, this is clearly manifested as a decrease in ozone in the CLaMS simulation interpolated along the ozonesonde profiles (Figure 4a and 4b). The observed sudden decrease of ozone in the mid-troposphere and UTLS in the period of 2011-2012 is also clear in the CLaMS simulations, which means that this sudden decrease originates in the stratosphere. The CLaMS simulations were much closer to the ozonesonde measurements in the mid-troposphere, but CLaMS seemed to overestimate the transport of ozone from the stratosphere to the troposphere in the UTLS.

Because pollution control measures in Beijing began around the same time as the decrease in stratospheric ozone, it is difficult to separate quantitatively their contribution to the observed reversal in trend. However, it is possible, at least qualitatively, to separate two ranges using CLaMS: (1) a tropospheric range in which the influence of stratospheric ozone is negligible; and (2) a stratospheric range where this influence is substantial. Thus, the control of air pollution in the troposphere-dominated range effectively reduced the concentration of ozone after 2012. By contrast, the ozone trends in the stratosphere-dominated range observed by the ozonesondes or simulated by CLaMS ~~both~~ became zero or negative after 2012. In the superposition range, most of the observed increasing trends weakened rapidly after 2012. Almost all the trends simulated by CLaMS became negative. The changes in the trends in the superposition range can be understood as an interaction between the impact of air pollution control and the changing influence of the stratosphere.

We conclude that the abrupt decrease and the deceleration of the increase in ozone in the troposphere and lower stratosphere is mainly the result of two overlapping effects: (1) the environmental protection measures implemented in recent years; and (2) variations in the transport of ozone from the stratosphere. Although the reduction in tropospheric ozone precursors played a dominant part, the effect of the transport of ozone from the stratosphere to the troposphere should not be ignored. Recently, there are studies indicate the surface ozone trends are affected by meteorological variation. Liu et al. (2020) recently found that higher temperatures after 2013 led to an increase in O3 concentrations in northern China via an increase in biogenic emissions. They assessed the effects of changes in meteorology (temperature, specific humidity, wind field, planetary boundary layer height,

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clouds, and precipitation) on ozone levels. Li et al. (2019) indicate that an important factor for ozone trends in the North China Plain is the ~40% decrease of fine particulate matter (PM2.5) over the 2013–2017 period. More observations are needed to investigate the variations in tropospheric ozone precursors and in related meteorology to fully understand the long-term variations in tropospheric and lower stratospheric ozone. More details of the transport of ozone from the stratosphere are expected to be revealed by comparing the CLaMS simulations of ozone with other observations. The observations of ozone over Beijing by IAP ozonesondes will be continued and we expect more improvements in reducing tropospheric ozone pollution.

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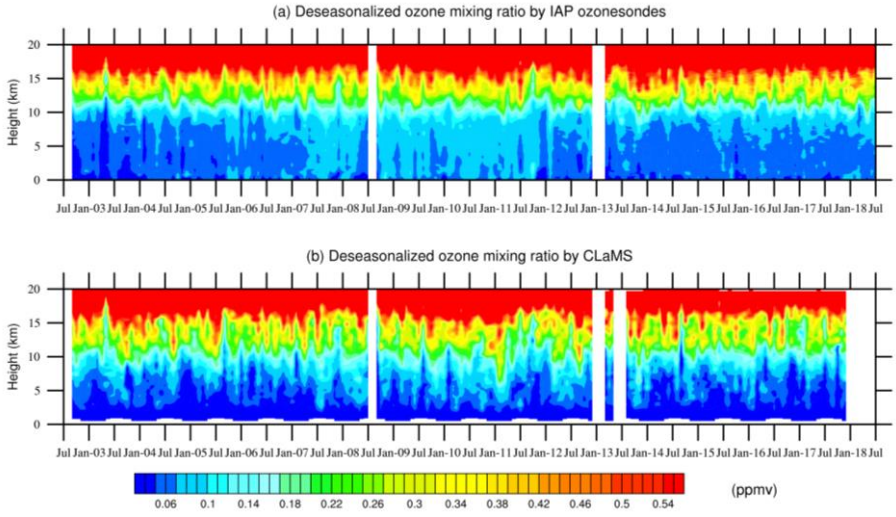

Figure 1. Deseasonalized monthly mean ozone mixing ratio (units: ppmv) over Beijing (a) measured by the IAP ozonesonde and (b) simulated by CLaMS. There was no ozonesonde observation in July 2008 and January 2013. CLaMS is unable to calculate ozone mixing ratio in July 2013, because the information of balloon locations were lost during this period.

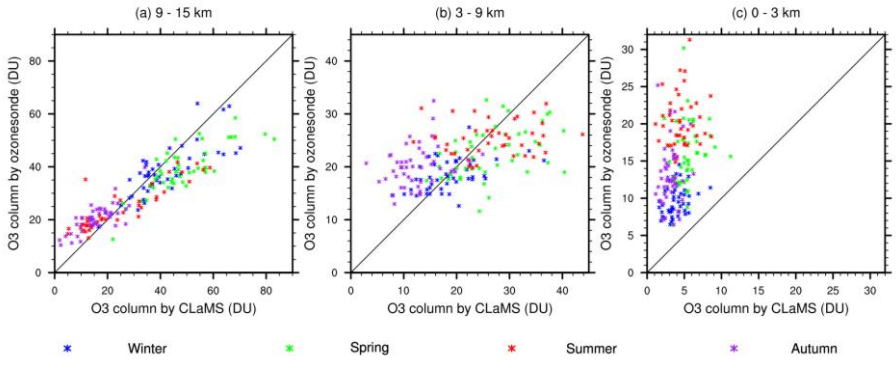

Figure 2. Validation of ozone columns simulated by CLaMS (in Dobson units, DU) by comparison with the IAP ozonesonde in (a) the UTLS, (b) the mid-troposphere and (c) the lower troposphere during 2012–2018. Each point represents the average of one month of measurements. Measurements in different seasons (winter: December–January–February; spring: March–April–May, summer: June–July–August and autumn: September–October–November) are shown as different colors.

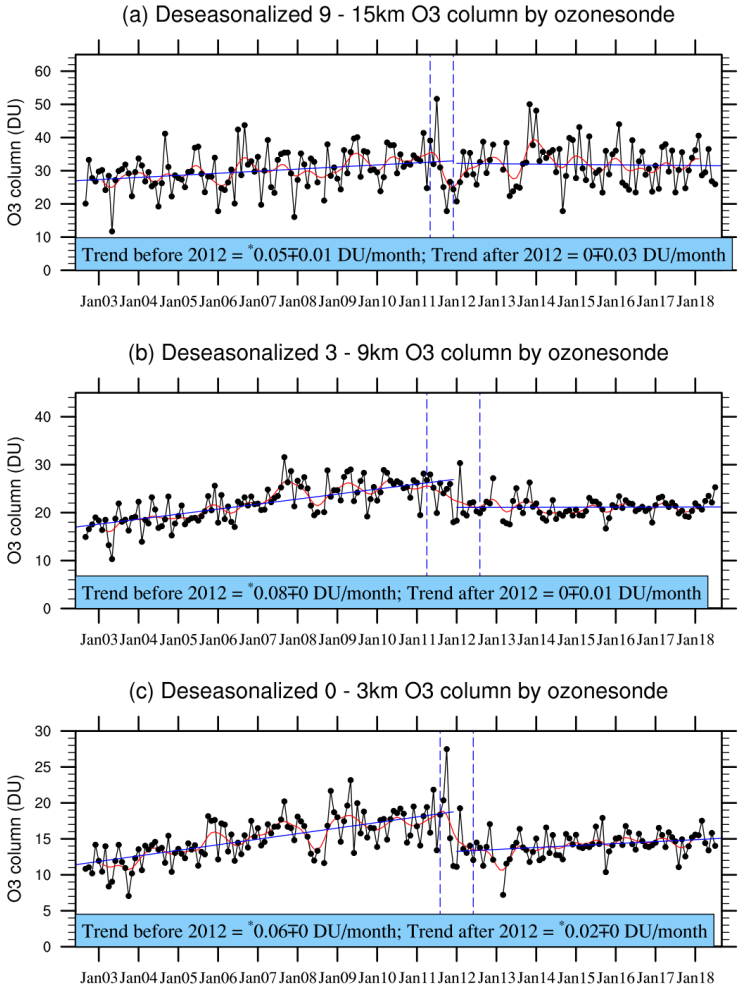

Figure 3. Deseasonalized monthly mean partial columns of ozone over Beijing (black solid lines and dots) measured by the IAP ozonesonde and the corresponding Gaussian-weighted means using a half-width of 12 months (red curves). The blue solid lines estimate the linear trends (slope ∓ standard error) before and after the decrease in ozone during late 2011 and early 2012. The periods of decrease are represented by the time between the two blue dashed lines. The trends with "*" passed the 95% significance criterion.

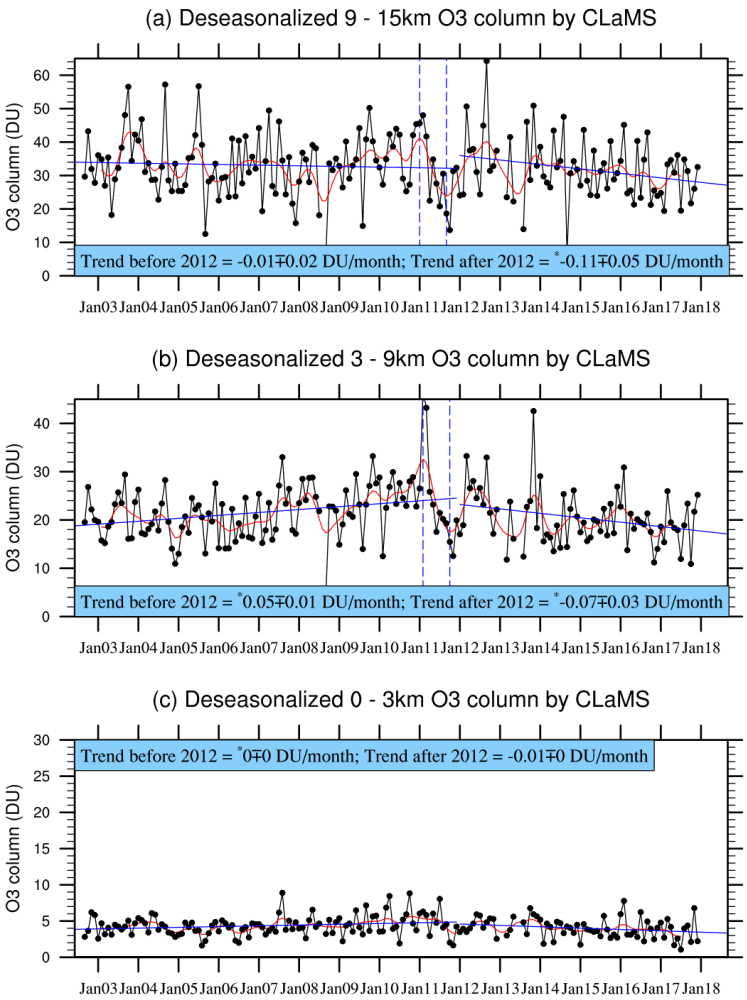

Figure 4. Deseasonalized monthly mean partial columns of ozone over Beijing (black solid lines and dots) simulated by CLaMS and the corresponding Gaussian-weighted means using a half-width of 12 months (red curves). The blue solid lines estimate the linear trends (slope ∓ standard error) before and after the decrease in ozone during late 2011 and early 2012. The periods of decrease are represented by the time between the two blue dashed lines. The trends with "*" passed the 95% significance criterion.

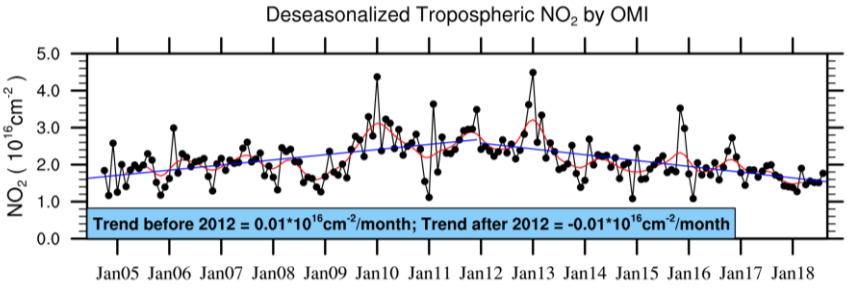

Figure 5. Deseasonalized monthly mean columns of tropospheric NO_2 (black solid lines and dots) and the corresponding Gaussian-weighted means using a half width of 12 months (red curves). The blue lines estimate the linear trend before and after the sudden decrease of ozone during late 2011 and early 2012.

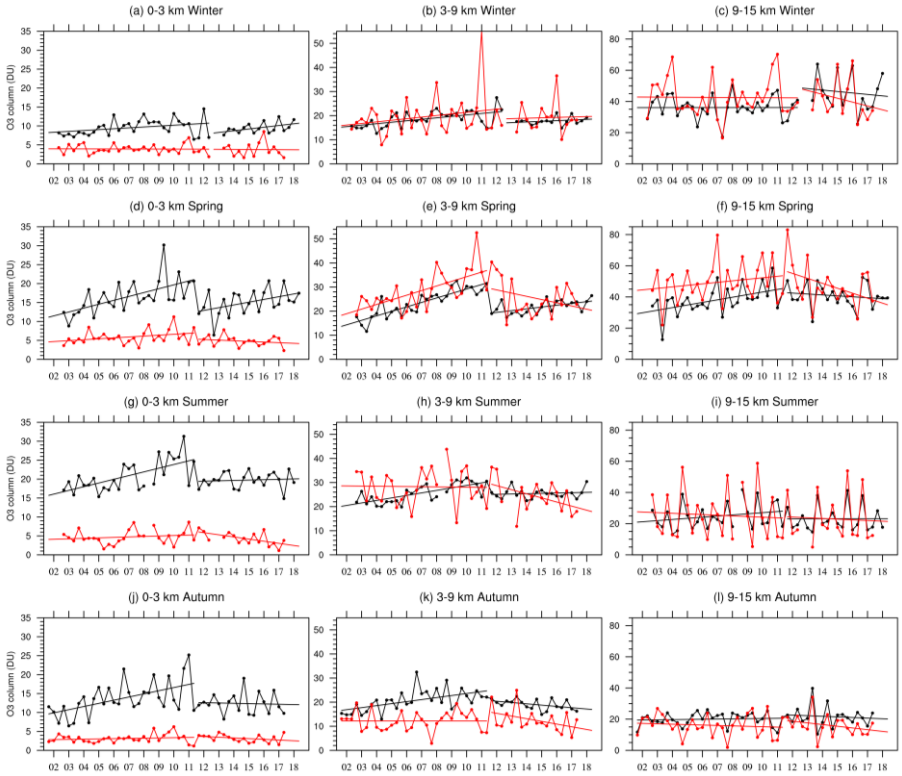

Figure 6. ~~Trends of M~~onthly mean column ozone (DU) from the ozonesonde observations (black) and CLaMS simulations (red) in four seasons. Trends of column are calculated before and after the sudden decrease of ozone in 2011-2012. There are 3 monthly values in each year for each season.

Table 1. Trends in the monthly mean partial column ozone (units: DU/year) at three altitudes in four seasons. The trends with "*" passed the 95% significance criterion. The trends with "※" passed the 90% significance criterion.

		Ozonesonde						CLaMS					
		M_{O3}	Before 2012		After 2012		Δ_{rel}	M_{O3}	Before 2012		After 2012		Δ_{rel}
			T_{O3}	T_{rel}	T_{O3}	T_{rel}			T_{O3}	T_{rel}	T_{O3}	T_{rel}	
9–15 km	Winter	39.22	0.02	0.0	−3.73	−9.5	−9.6	42.15	−0.20	−0.5	−9.90	−23.5	−23.0
	Spring	39.26	*6.66	*17.0	−2.37	−6.0	−23.0	48.48	3.87	8.0	※−12.71	※−26.2	−34.2
	Summer	24.09	2.92	12.1	0.02	0.1	−12.0	24.50	−1.65	−6.8	−1.77	−7.2	−0.5
	Autumn	20.61	0.49	2.4	−1.57	−7.6	−10.0	16.05	−1.07	−6.7	−4.35	−27.1	−20.5
3–9 km	Winter	18.38	*2.43	*13.2	1.08	5.9	−7.3	19.46	2.64	13.6	0.67	3.4	−10.1
	Spring	22.77	*7.43	*32.6	*2.97	*13.0	−19.6	27.30	*7.68	*28.1	−5.34	−19.6	−47.7
	Summer	25.57	*4.16	*16.3	0.55	2.1	−14.2	26.69	−0.27	−1.0	※−6.80	※−25.5	−24.5
	Autumn	20.04	*3.35	*16.7	※−2.15	※−10.7	−27.4	12.34	−0.01	−0.1	※−4.47	※−36.2	−36.2
0–3 km	Winter	9.55	※0.93	※9.7	※1.83	※19.2	9.4	3.87					
	Spring	15.94	*4.10	*25.7	*2.90	*18.2	−7.5	5.46					
	Summer	20.38	*3.91	*19.2	0.39	1.9	−17.3	4.57					
	Autumn	13.18	*3.34	*25.3	−0.33	−2.5	−27.9	3.14					