Reply to Ref. #3

First of all we want to thank this reviewer for the positive assessment of our manuscript and the constructive and helpful suggestions.

General comments

The paper presents a comprehensive study of vertical distributions of NO2, SO2, HONO, HCHO, CHOCHO and aerosols by MAX-DOAS measurements during a spring/summer period (from 8 May to 10 June 2016) at a suburban site of the North China Plain. The profiles of these gases (volume mixing ratio) and aerosols (extinction coefficient) retrieved by MAX-DOAS are compared with the independent data, including in-situ measurements, Sun photometer, visibility meter, lidar and aircraft measurements. The effects of emissions and transport on the observed results are also analyzed using the backward trajectories and various satellite data. The study is interesting, providing important information to the scientific community on air quality issue in eastern China. The paper is well written and organized, I would recommend the paper to be published subject minor revisions. My major concern is on the comparison of the vertical profiles between ground-based MAX-DOAS and in situ aircraft measurements. While Sect. 5 devotes too much for a discussion about the regional and local transport of pollutants, more detailed analyses and discussions should have be added in Sect. 4.3 for the comparison of MAX-DOAS with aircraft measurements.

Author reply:

Many thanks for the positive assessment! We modified the paper based on the comments from you and the other two reviewers. Please see the replies and modifications regarding your specific comments below.

Specific Comments:

1) - Aerosol extinction and SO2 mixing ratio are underestimated significantly by MAX-DOAS with comparison to the aircraft measurements on 21 May 2016 (black dots in Fig. 8b). Why are the aircraft profiles, instead of MAX-DOAS profiles, converted (or "corrected") for better comparison? Since the airplane flew in a spiral route, were the chemical instruments stable enough to get reliable data with increasing air pressure? What is the vertical resolution (or precision) of the profile inversion by MAX-DOAS? The concept of the smoothing effect of the MAX-DOAS profile inversion should be discussed more in detail. I cannot find sufficient evidences in Sect. 4.2 to support the conclusion "The smoothing effect can cause MAX-DOAS retrievals to underestimate pollutants above 2 km and overestimate below" stated in Page 21, Line 31-32.

Author reply: Thanks for the comment! We give the answers to your individual questions below:

Question 1: "Why are the aircraft profiles, instead of MAX-DOAS profiles, converted (or "corrected") for better comparison?"

Answer: The question might be related to the unclear explanation in the manuscript. We modified the sentence in the revised manuscript as follows:

"Since the limited response of MAX-DOAS profile retrievals to the true profiles, the retrieved profile $\hat{x}$ can be represented as the true profile x, smoothed by the AK according to the equation: $\hat{x} = x_a + AK(x - x_a)$, where xa is a-priori profile used in the profile retrieval of MAX-DOAS. To account for the smoothing effect of the MAX-DOAS profile inversion in the comparisons, the AKs of the MAX-DOAS profile retrievals are applied to the averaged aircraft profiles, which are treated as the true atmospheric profile x to generate the "smoothed profiles" $\hat{x}$. Additionally the combined profiles, derived from the averaged aircraft profile and surface data, are considered as the true atmospheric profile x and converted to "smoothed combined profiles" using the AK of the MAX-DOAS profile retrievals. The "smoothed profiles" and "smoothed combined profiles" are shown in Fig. 8. By comparing the smoothed profiles with the original profiles derived from the aircraft measurements, the smoothing effect of MAX-DOAS retrievals can be evaluated."

Question 2: Since the airplane flew in a spiral route, were the chemical instruments stable enough to get reliable data with increasing air pressure?

Answer: The NO_2 analyzer have internal pressure controllers that maintain the pressure constant at 128 torr, well below the pressure altitudes we flew. So their measurements are not affected by the ambient pressure changes at all. All other trace gas analyzers like ozone, SO_2, NO, and NOy are corrected for pressure and temperature when they reported the final concentrations. So our instruments are stable enough to make reliable measurements during the spiral profiles. All the aircraft instruments have been used for airborne measurements in the United States and China (e.g. Taubman et al., 2006; Dickerson et al., 2007; Hains et al., 2008; He et al., 2012; He et al., 2014; Ren et al., 2018; Salmon et al., 2018).

Taubman, B. F., Hains, J. C., Thompson, A. M., Marufu, L. T., Doddridge, B. G., Stehr, J. W., Piety, C. A., and Dickerson, R. R.: Aircraft vertical profiles of trace gas and aerosol pollution over the mid-Atlantic United States: Statistics and meteorological cluster analysis, Journal of Geophysical Research-Atmospheres, 111, D10s07 10.1029/2005jd006196, 2006.

Hains, J. C., Taubman, B. F., Thompson, A. M., Stehr, J. W., Marufu, L. T., Doddridge, B. G., and Dickerson, R. R.: Origins of chemical pollution derived from Mid-Atlantic aircraft profiles using a clustering technique, Atmospheric Environment, 42, 1727-1741, 10.1016/j.atmosenv.2007.11.052, 2008.

He, H., Li, C., Loughner, C. P., Li, Z., Krotkov, N. A., Yang, K., Wang, L., Zheng, Y., Bao, X., Zhao, G., and Dickerson, R. R.: SO2 over central China: Measurements, numerical simulations and the tropospheric sulfur budget, Journal of Geophysical Research: Atmospheres, 117, doi:10.1029/2011JD016473, 2012.

He, H., Loughner, C. P., Stehr, J. W., Arkinson, H. L., Brent, L. C., Follette-Cook, M. B., Tzortziou, M. A., Pickering, K. E., Thompson, A. M., Martins, D. K., Diskin, G. S., Anderson, B. E., Crawford, J. H., Weinheimer, A. J., Lee, P., Hains, J. C., and Dickerson, R. R.: An elevated reservoir of air pollutants over the Mid-Atlantic States during the 2011 DISCOVER-AQ campaign: Airborne measurements and numerical simulations, Atmospheric Environment, 85, 18-30, 10.1016/j.atmosenv.2013.11.039, 2014.

Ren, X., Salmon, O. E., Hansford, J. R., Ahn, D., Hall, D., Benish, S. E., Stratton, P. R., He, H., Sahu, S., Grimes, C., Heimburger, A. M. F., Martin, C. R., Cohen, M. D., Stunder, B., Salawitch, R. J., Ehrman, S. H., Shepson, P. B., and Dickerson, R. R.: Methane Emissions From the Baltimore-Washington Area Based on Airborne Observations: Comparison to Emissions Inventories, Journal of Geophysical Research: Atmospheres, 0, doi:10.1029/2018JD028851, 2018.

Salmon, O. E., Shepson, P. B., Ren, X., He, H., Hall, D. L., Dickerson, R. R., Stirm, B. H., Brown, S. S., Fibiger, D. L., McDuffie, E. E., Campos, T. L., Gurney, K. R., and Thornton, J. A.: Top-Down Estimates of NOx and CO Emissions From Washington, D.C.-Baltimore During the WINTER Campaign, Journal of Geophysical Research: Atmospheres, 123, 7705-7724, doi:10.1029/2018JD028539, 2018.

Question 3: What is the vertical resolution (or precision) of the profile inversion by MAX-DOAS?

Answer: the vertical resolution can be represented by the averaging kernels. The resolution is shown in the last paragraph of section 2.2.3 and Fig. 4c. In order to introduce the meaning of the averaging kernel more clearly, we modified the sentence in the revised manuscript as follows:

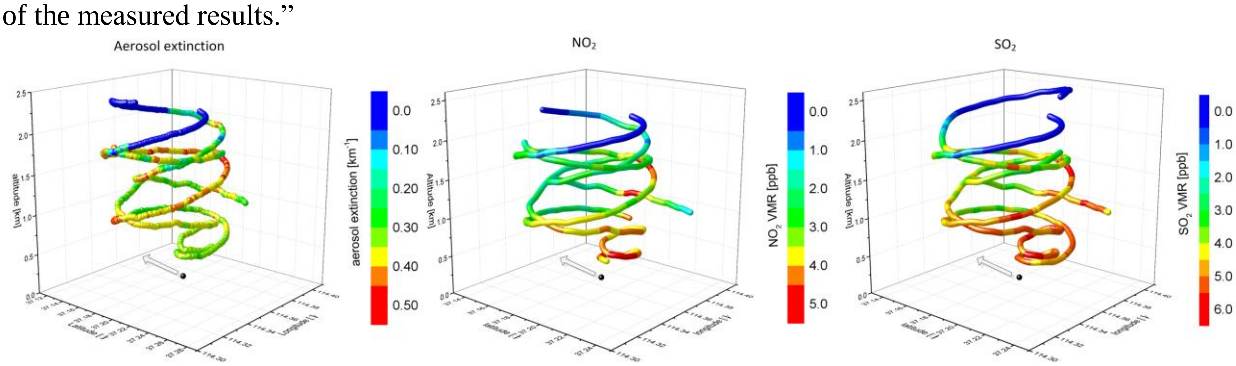
"The vertical resolution and sensitivities of the retrievals at different altitutdes can be quantified by the so-called averaging kernel matrix $AK = \partial \hat{x} / \partial x$, which represents the sensitivity of the retrieved profile x^ as a function of the true atmospheric profile x. The typical AK of the profile inversions shown in Fig. 4c indicate that the sensitivity of the profile retrievals of trace gases and aerosols systematically decreases with altitude. "

Question 4: The concept of the smoothing effect of the MAX-DOAS profile inversion should be discussed more in detail. I cannot find sufficient evidences in Sect. 4.2 to support the conclusion "The smoothing effect can cause MAX-DOAS retrievals to underestimate pollutants above 2 km and overestimate below" stated in Page 21, Line 31-32.

Answer: In order to discuss the smoothing effect better, we added a sentence in Section 4.3 in the revised manuscript as follows: "Generally, pollutants above 1km are significantly underestimated due to the smoothing effect of MAX-DOAS profile retrievals.” And the sentence in the conclusion section is modified as “The smoothing effect of MAX-DOAS profile retrievals can cause a reshaping of box-profiles below 2km towards exponentially decreasing profiles. This effect can cause MAX-DOAS measurements significantly underestimate pollutants located at altitudes above 1km.”.

2) - It is stated that “the deviations between the MAX-DOAS and aircraft measurements can probably also be attributed to inhomogeneous horizontal distributions of pollutants and their temporal variation during a period of aircraft measurements” in Sect. 4.3 (Page 13, Line 14-16). Did you find any regular horizontal distribution patterns of aerosols and gases from aircraft measurements? Will the comparison improve if only the aircraft measurements in the area that the MAX-DOAS instrument was pointed to are selected?

Author reply: A large variability of the original data from the aircraft measurements at individual altitudes can be seen in Fig. 8. This finding indicates inhomogeneous horizontal distributions of the pollutants. In order to show the phenomenon more clearly, we plotted 3D distribution of aircraft data on 21 May in the following figures. In the figure, the colors indicate aerosol extinctions or VMRs of NO_2 and SO_2. The black dots on the surface represent the location of the MAX-DOAS instrument, and the arrows point to the direction of the MAX-DOAS telescope. The figures clearly indicate strong horizontal gradients of the pollutants. The new figures given below are not included in the manuscript because its information can be well shown with the variability of original aircraft data in Fig. 8. Since in the pointing direction of the MAX-DOAS telescope only a few aircraft measurements are available, we didn’t do the comparisons only for these data. Another important aspect is that the aircraft results are from in-situ measurements, whereas MAX-DOAS measurements represent averages of the pollutants along an effective light path of ~ 5 to 10 km. Therefore the different air masses measured by the two techniques can be seen as one important reason for the differences of the results. The effect of different probed air masses was not clearly pointed out in the previous manuscript. Therefore we added the following sentences in the revised manuscript:
“In addition, aircraft results represent in-situ measurements along the spiral route, whereas MAX-DOAS results represent averages of pollutants over an effective light path of ~5 to 10 km. The different air masses measured by the two techniques can be seen as one important reason for the observed differences of the measured results.”



3) - In addition to co-author’s research group, other aircraft measurement work in the NCP region should be credited, e,g. Ma et al. (2012) and Zhang et al. (2014); so did the MAX-DOAS measurement, e.g., Jin et al. (2016).

Author reply: Thanks for reminding these references! We cited them in appropriate positions in the introduction section of the revised manuscript.

Technical corrections:

1) Page 2, Line 32. What does “East-Aire” mean ?
Author reply: It is the abbreviation of “East Asian Study of Tropospheric Aerosols: an International Experiment”. The full name is given in the revised manuscript.

2) Page 4, Line 23: What is the terrain height of the station?
Author reply: The terrain height of the station is ~200 m asl. The information is given in the revised manuscript.

3) Page 5, Line 15-20: The direction for the measurement should be mentioned.
Author reply: The telescope was pointed to an azimuth angle of 25 ° northeast. The information is given in section 2.2.1 in the revised manuscript.

4) Page 7, Line 12. Please check the punctuation here as well as elsewhere in the manuscript.
Author reply: the punctuations are checked.

5) Page 8, Line 7-8. There are two references of Zhang et al., 2018. Please distinguish them when citing.
Author reply: One of the tow references was wrongly cited. Therefore we deleted this one in the revised manuscript.

6) Page 9, Line 15-17. Please delete the repeating word of “be”.
Author reply: It is corrected in the revised manuscript.

7) Page 12, Line 25-26. The agreement of the aerosol profiles from MAX-DOAS and lidar above 500m is not obvious, especially on 16 May, 2016. It is better to alter the color bar to show this point more clearly.
Author reply: Since aerosol extinction at high altitudes is much lower than those at ~500m, we think it is not necessary to highlight these differences. We prefer to use the current color bar, which can balance the requirement to show structures of high concentrations and low concentrations at different altitudes.

8) Page 12, Line 30. The content should move to the section 4.1.
Author reply: We added the following clarification in the section 4.1: “In addition, the sun-photometer measured the air masses in the direction of the sun. The different air mass measured by the two techniques can contribute to the differences of the AOD results.”
We also clarified the same statements for the other instruments in section 4.1 as follows: “Note that the visibilitymeter and the in-situ measurements of NO2, SO2 and HCHO represent air masses close to the instruments, whereas the MAX-DOAS measurements represent averages of pollutants along the effective horizontal light path of ~5 to 10 km in the vertical grid from the surface up to 0.2 km. Therefore different probed air masses can be seen as one important reason for the differences of results.”

9) Page 13, Line 20-22. This paragraph seems to be redundant.
Author reply: We prefer to keep the paragraph to give readers an impression on the contents of section 5 before they go to the details.

10) Page 14, Line 4. The language expression needs to be improved.
Author reply: The sentence was modified in the revised manuscript as follows: The mountain-plain topography causes a daily cycle with downslope (northeast winds) and upslope (southeast winds) winds.

11) Page 16, Line 6. Please note the subscript.
Author reply: They are corrected in the revised manuscript.

12) Page 16: Sect. 5.2.2. It is known that the MAX-DOAS measurements are per- formed during the daytime. However, the sorting here is mainly based on the nighttime trajectories. In addition, there are large differences between nighttime and daytime in Fig.S5, especially for the southerly trajectories.
Author reply: We clarified the reason why we use the nighttime trajectories to separate the results in section 5.1 as follows "Therefore we can expect that the gas pollutants, e.g. NOx, SO2 and HONO, can be transported to a farther distance during nighttime than daytime. Thus nighttime regional transport of pollutants from the Wuan area could significantly pollute the entire measurement area. " and in section 5.2.2 "Considering that the life times of the observed trace gases are typically longer during night time than day time (because of lower OH radical concentrations), the measurement data are sorted mainly based on the nighttime trajectories". Note that generally the difference between the nighttime and daytime trajectories in Fig. S5 is not big. But the difference is also the reason why we only use night time trajectories to sort the data.

13) Page 17, Line 34. Please change "Fig.12i" to "Fig.12I".
Author reply: It is changed in the revised manuscript.

14) Page 22, Line 23, 26. Please check the names of the station "Wuxi" and software "WINDOAS".
Author reply: They are all corrected in the "Acknowledgements" of the revised manuscript.

15) Figure 6: Please clarify the temporal resolution of the data used in Fig.6.
Author reply: we added the information for the figure in the revised manuscript as follows:
"All independent data are averaged over the individual time intervals of the MAX-DOAS measurements."

16) Table 2: "outliers"?
Author reply: We used the filters given in the table to get rid of "outliers". We do not apply any other filters to delete specific "outliers".

17) References
Jin, J., Ma, J., Lin, W., Zhao, H., Shaiganfar, R., Beirle, S., and Wagner, T.: MAX-DOAS measurements and satellite validation of tropospheric NO_2 and SO_2 vertical column densities at a rural site of North China, Atmospheric Environment, 133, 12-25, http://dx.doi.org/10.1016/j.atmosenv.2016.03.031, 2016.
Ma, J. Z., Wang, W., Chen, Y., Liu, H. J., Yan, P., Ding, G. A., Wang, M. L., Sun, J., and Lelieveld, J.: The IPAC-NC field campaign: a pollution and oxidization pool in the lower atmosphere over Huabei, China, Atmos. Chem. Phys., 12, 3883-3908, 10.5194/acp- 12-3883-2012, 2012.
Zhang, W., Zhu, T., Yang, W., Bai, Z., Sun, Y. L., Xu, Y., Yin, B., and Zhao, X.: Airborne measurements of gas and particle pollutants during CAREBeijing-2008, Atmos. Chem. Phys., 14, 301-316, 10.5194/acp-14-301-2014, 2014.

Author reply: Thanks for reminding these references. We cited them at appropriate positions in the introduction section of the revised manuscript.