

Reply to Ref. #2

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

General comments

Wang et. al presented a MAX-DOAS observation for tropospheric vertical profiles of NO₂, SO₂, HONO, HCHO, CHOCHO and aerosols in the central- western North China Plain in May and June 2016. The MAX-DOAS results are validated comprehensively by the collocated measurements of ground based lidar, sun-photometer and in situ instrument, as well as overpass aircraft. Besides, characteristics of pollutants distribution and variations were analyzed combined with effects of regional and local transport. As shown in the introduction, there were many studies of the trace gases and air pollutions of NCP in previous, also including the MAX-DOAS measurements. The main concerns is that what is the novelty or unique of this paper compared to the previous. I suggest the authors could highlight these in the manuscript.

Author reply:

Many thanks for the suggestion! We modified the manuscript based on the comments from you and the other two reviewers. The one-to-one replies are given in the following part. For your main concern, we followed your suggestion to highlight the novelty and unique point of the study in the abstract and introduction as follows:

“Note that although several MAX-DOAS measurements of trace gases and aerosols in the NCP area have been reported in previous studies, this study is the first work to derive a comprehensive set of vertical profiles of NO₂, SO₂, HONO, HCHO, CHOCHO, and aerosols from measurements of one MAX-DOAS instrument. Also, so far the validation of MAX-DOAS profile results by comparison with various surface in-situ measurements as well as profile measurements from Lidar and aircraft is scarce. Moreover, the backward propagation approach to characterize the contributions of regional transport of pollutants from different regions was for the first time applied to the MAX-DOAS results of trace gases and aerosols.”

Specific Comments:

- 1) MAX-DOAS spectra analysis: It can be concluded from P5, Line27-28 that the authors used a spectrum measured in the zenith direction closest in time to the off-zenith measurements as a Fraunhofer reference spectrum. So if the telescope scanned in the sequence of 1°, 2°, 3°, 4°, 6°, 8°, 10°, 15°, 20°, 30°, 90°, the DSCDs of lower elevation angle (e.g. 1, 2, 3, 4) should use the zenith spectrum in previous scanning, but the DSCDs of higher elevation angle (e.g. 10, 15, 20, 30) use the zenith spectrum of current scanning. It means that the DSCDs of elevation angles in the same scanning were obtained with different reference spectrum. Any explanation or consideration about this treatment, which may bring some unknown effects in the profile retrieval procedure? Fig.3: why the authors show the CHOCHO spectral analysis in another day compared with other species? And the CHOCHO absorption structure can not be well observed.

Author reply: Regarding the Fraunhofer reference spectrum, thanks for pointing out the obscure elaboration! We modified the description in the revised manuscript as follows: “A sequential Fraunhofer reference spectrum, which is derived from interpolation of two zenith spectra measured before and after an elevation sequence to the measurement time of individual off-zenith measurements, is used in the DOAS fits.”. Regarding the HCHO spectral fit shown in Fig. 3, the CHOCHO dSCD around noon on 27 May is the highest during the whole campaign. As you have seen, CHOCHO fit is quite difficult to analyse, its largest optical depth is only ~0.001, two orders smaller than the optical depth of NO₂. In order to show the best fit, we showed the results on 27 May in Fig. 3. We clarified the point in the revised manuscript for Fig. 3 as follows: “Note that the CHOCHO fit shown in the figure is for the largest CHOCHO dSCD retrieved around noon during the whole campaign period.”

- 2) When you evaluated the DOAS data for HONO, did you consider the impurity of HONO in the NO₂ reference spectra used? There is always some HONO in NO₂ and that is subtracted in the DOAS algorithm. This leads to an underestimation of HONO by ca. 0.5% of the NO₂, which can be significant during daytime and impacts the conclusions in your discussion about HONO/NO₂.

Author reply: We searched the literature regarding measurements of NO₂ cross sections, namely NO₂ reference spectra. However we found no publications reporting the effects of the contamination of HONO in the NO₂ cell. In addition if there are HONO structures in the NO₂ cross section, we can expect an increase of (negative) HONO dSCDs along the increase of NO₂ dSCDs during the day. However we don't see such an increase. Therefore we think the HONO impurity effect on the calculations of the HONO/NO₂ ratio in the study is negligible. If the reviewer knows a publication about the HONO impurity issue, please inform us.

- 3) Aerosol and trace gases retrieval:

How was the vertical grids setting?

How to distinguish the sky condition of high aerosols and clouds?

In section 4.1, since the aerosol retrieval results were poor under the sky conditions of clear sky with high aerosols and cloudy sky (Fig. 6a and b), how to convince the trace gases retrieval are reliable? All the reliable retrieval are the fundamental of the further analysis about effects of regional and local transport of pollutants.

Author reply: The vertical grid is 200 m. The information is added in the revised manuscript.

Regarding the cloud classification, the difference between “clear sky with high aerosol load” and “continuous clouds” is the spread of the color index at different elevation angles of the MAX-DOAS measurements. The spread is much smaller under “continuous clouds” than under “clear sky with high aerosol load”. The difference between “clear sky with high aerosol load” and “broken clouds” is the temporal variations of the color index measured by MAX-DOAS. Because the cloud coverage can change rapidly under “broken clouds”, the temporal variation is much larger under “broken clouds” than under “clear sky with high aerosol load”. We elaborated the details of the method in our previous publications of “Wagner, T., Beirle, S., Dörner, S., Friess, U., Remmers, J. and Shaiganfar, R.: Cloud detection and classification based on MAX-DOAS observations, *Atmos. Meas. Tech.*, 7, 1289-1320, 2014” and “Wagner, T., Beirle, S., Remmers, J., Shaiganfar, R., and Wang, Y.: Absolute calibration of the colour index and O₄ absorption derived from Multi AXis (MAX-)DOAS measurements and their application to a standardised cloud classification algorithm, *Atmos. Meas. Tech.*, 9, 4803-4823, <https://doi.org/10.5194/amt-9-4803-2016>, 2016.”.

Regarding the cloud effect, since clouds typically located at altitudes above the trace gases, clouds have usually a stronger impact on the O₄ absorptions than on the trace gases. Therefore they impact the aerosol retrievals stronger than the trace gas retrievals.

Under high aerosol load conditions, the discrepancy between the aerosol results from MAX-DOAS and sun-photometer and visibility meter measurements are probably mainly due to inhomogeneous horizontal distributions and different air masses measured by the different instruments. In addition, MAX-DOAS might underestimate aerosols at high altitudes due to the low sensitivity of MAX-DOAS measurements there. Since trace gases typically located at low altitudes, it is not probable that the underestimation of aerosols at high altitudes by MAX-DOAS significantly impacts the trace gas profile retrievals.

Technical corrections:

- 1) P4, Line 28, “10:00 BT” change to “10:00 LT”

Author reply: Thanks for pointing it out. BT is Beijing time. In order to clarify the point, we added a sentence in the revised manuscript as follows: “Since the longitude difference of the station and Beijing is only 2°, the Beijing time is almost the local time.”.

- 2) P5, Line 3-7, the results in Fig. 2 d were obtained from NASA website, however, the data in Fig. 2a, b and c? And the spatial resolution of the satellite products? Did the authors do any treatment or filter with the data? Please specify more clearly.

Author reply: Thanks for pointing out the missing information. We modified the paragraph regarding the satellite data in section 2.1 to add the information in the revised manuscript. The modified paragraph is the following:

“Averaged maps of NO₂ (from DOMINO v2, Boersma et al., 2007 and 2011), SO₂ (from BIRA-IASB, Theys et al., 2015), and HCHO (from BIRA-IASB, De Smedt et al., 2008, 2012 and 2015) derived from satellite observations of the Ozone Monitoring instrument (OMI) (Levelt et al., 2006a and b) for May and June during the period 2012 to 2016 for the same area as shown in Fig. 1a are shown in Fig. 2a, b, and c, respectively. The spatial resolution of the OMI data is 13×24 km² in nadir. Note that the OMI data of the outermost pixels (i.e. pixel numbers 1–5 and 56–60) and pixels affected by the so-called “row anomaly” (see <http://www.temis.nl/airpollution/no2col/warning.html>) were removed. In Fig. 2d a map of the averaged aerosol optical depths (AODs) at 550 nm derived from the Moderate Resolution Imaging Spectroradiometer (MODIS) (Kaufman et al., 2002) for the same period is shown (provided by NASA on <http://ladsweb.nascom.nasa.gov/data/search.html>). The spatial resolution of the MODIS AOD data is 5×5 km². In order to exclude cloud contaminated data, for both OMI and MODIS data, only the data with cloud fractions smaller than 30% are included for the generation of the maps. A grid interval of 0.02 ° is used to generate the averaged maps of the OMI and MODIS data by binning the satellite data of pixels around each grid with distance weightings.”

- 3) Fig. 2a, c, d, poor resolution. Please correct.

Author reply: Since the pixels of OMI satellite instruments cover an area of 13×24km², the map resolution can not be further improved.

- 4) Fig. 7, I suggest the author present a panel plot of the differences of AE between MAX-DOAS and Lidar for more clearly and apparent comparison results.

Author reply: We followed the idea and added the panel in the new Fig. 7 in the revised manuscript.

- 5) Acknowledgements:

MAX-DOAS, LP-DOAS and etc. in Wuxi station? But the measurements was in NCP area.

WINDOAS software? But you used QDOAS

Author reply: Thanks for pointing out the mistakes! The mistakes are corrected in the revised manuscript.