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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ground-based MAX-DOAS observations of tropospheric aerosols, NO_2, SO_2 and HCHO in Wuxi, China, from 2011 to 2014" *by* Y. Wang et al.

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Reply to Ref. #1

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

General comments This paper presents long-term (May 2011-November 2014) MAX-DOAS observations of tropospheric aerosols, NO2, SO2 and HCHO in Wuxi, China. Vertical profiles of trace gas concentrations and aerosol extinctions are retrieved using a new inversion algorithm called PriAM. It is based on the Levenberg-Marquardt modified Gauss-Newton numerical procedure and uses the SCIATRAN radiative trans-

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fer model as forward model. In the first part of the paper, MAX-DOAS observations and the PriAM algorithm are described and the following issues are investigated: impact of the surface pressure and temperature seasonality regularly observed in Wuxi on O4 VCD and aerosol retrieval, observed differences between VCDs derived using the geometric approximation and the profiling algorithm, impact of the sky conditions on the aerosol and trace gases retrievals and on the agreement between MAX-DOAS and correlative measurements (AERONET, LP-DOAS). In the second part of the paper, MAX-DOAS data are used to characterize the seasonal, diurnal, and weekly variations of NO2, SO2, HCHO and aerosols. This is a very interesting study of high scientific quality which fits well with the scope of ACP. I recommend its final publication after addressing the following comments.

Author reply: Many thanks for the positive assessment!

Major comments

-Although clearly structured, the manuscript is difficult to read due to the large number of figures and panels in the manuscript itself (29) and in the supplement (28). The authors should improve the readability of their paper for the final publication in ACP by focusing on the main results only and asking themselves which figures are needed to best illustrate these results.

Author reply: We minimized the number of figures to only show the necessary figures in the main manuscript. In the revised version there are only 15 figures in the main part of the manuscript and in total fewer subplots in the manuscript and supplement than in the original version. To make the paper more focused on the most important new findings, we moved the original section 2.2.4 into the supplement as new section 3.2 (the main conclusion of the original section 2.2.4 is summarized at the end of section 2.2.2. We also moved the original Fig. 5 into the supplement as new Fig. S10. The original Fig. S11 is removed from the supplement. We made the new Fig. 6 in order to replace the original Figs. 8-12. The key information including the mean differences, standard

deviations, R, slopes, intercepts and numbers of observations in the original figures are now all plotted in the new Fig. 6. We made the new Fig. 9 in order to replace the original Figs. 15-19 to only show the key information. In the new supplement the new Figs. S21-24 summarize the key information from the previous Figs. S20-S23. We also moved the original section 3.1 and Fig. 21 and 22 (containing the meteorology data) into the supplement (a yellow arrow is added in Fig.1b of the revised version to show the dominant wind direction). We also removed the original Fig. 4a and only keep Fig. 4b in the revised version (because the information in Fig. 4a is already well presented in Fig. 4b). The original Fig. 7 is removed. Although the current supplement still contains many figures, we think it is good for the readers who want to learn the details of the study. Thus we paid more effort to shorten the main manuscript.

Specific Comments:

1) Sect. 2.2.3, page 6: One important result is the impact on the seasonal variation of the pressure and temperature profiles on the retrieved AODs and aerosol extinctions. So far most MAXDOAS groups were using US Standard Atmosphere in their aerosol extinction profile retrievals from O4 slant column densities. The authors show in their study that ignoring this systematic seasonal variation in Wuxi can cause a 20-30% bias on the retrieved AODs and near-surface aerosol extinctions, possibly yielding to unrealistic seasonal variations of these quantities. Is such a large effect only specific to the Wuxi region or can we expect similar features in other parts of China, especially the Beijing area where several MAXDOAS instruments are currently in operation?

Author reply: The variation of O4 VCD depends on the systematic seasonal variation of temperature and pressure. The seasonal variation of temperature occurs in many locations of the world (especially outside the tropics). However the temporal variation of the pressure is usually more complex and can be very different at different locations. The variation of pressure in Wuxi (and also many other parts of Eastern China) is related to the East Asian Monsoon and shows a systematic seasonal pattern. The monsoon is a general phenomenon in the eastern China. The pressure in the conti-

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nent is systematically lower and higher than that in the ocean in summer and winter, respectively. Thus a similar seasonal variation of the O4 VCD is expected in general in Eastern China including Beijing. We added this information at the end of section 2.2.3 of the revised manuscript.

2) Sect. 2.2.4, page 6 and Sect. 2.2.5, page 8: The evaluation of the internal consistency of the inversion algorithm and the validation of the retrieval results are performed for favourable measurement conditions, i.e. 'clear-sky with relatively low aerosols (average AOD of about 0.6)'. 'Average AOD of about 0.6' is for me very vague. Is it the daily or hourly average? Looking at Figure 9 showing the scatterplots of the MAX-DOAS versus AERONET AODs, MAXDOAS AOD values much larger than 0.6 (see scatterplot for Spring) are selected while this figure is supposed to illustrate the agreement between MAXDOAS and AERONET in low aerosols (and clear-sky) conditions. I think a clarification is needed here.

Author reply: Thanks for pointing out the misleading description. 'Clear-sky with relatively low aerosols' belongs to one category of the sky conditions identified by MAX-DOAS observations. Please see section 2.2.5. To clarify the point, we added a sentence at the end of the first paragraph of section 2.2.5: "Another point which needs to be clarified is that distinguishing "low aerosols" and "high aerosols" is based on the colour index observed by MAX-DOAS. Thus there is not an explicit AOD value which distinguishes both aerosol categories. The studies of Wang et al., 2015, however, demonstrated that the AODs observed by the Taihu AERONET sun photometer are mostly smaller and larger than 0.6 for the "low aerosols" and "high aerosols", respectively. In addition to the cloud effect, also the effect of high aerosol loads is evaluated (due to the unrealistic assumption of the pdf of the atmospheric state in the OE algorithm for high aerosol loads (see Eq. (1))." Note that in the revised version of the manuscript the original section 2.2.4 was moved to the supplement as new section 3.2. In the beginning of section 3.2 in the supplement, we deleted "average AOD of about 0.6", but write "the sky condition is directly identified by MAX-DDOAS observations,

see section 2.2.5".

3) Sect. 2.2.5, page 8: MAXDOAS AODs are validated using AERONET data from the Taihu station, which is located at 18 km south west of the MAXDOAS instrument location. Based on the coordinates mentioned on the AERONET website, the sun photometer is located in the south of the Taihu mountains, at the edge the Taihu Lake (west of the lower left corner of the orange rectangle in Figure 1b), which seems to be a much more remote area than the one from where the MAXDOAS instrument is operating (see also figure 1b). The question is therefore how representative is the Taihu station compared to the location of the MAXDOAS instrument? Did the authors consider this point for the interpretation of their comparison results?

Author reply: Thanks for pointing out this potential source for differences. We added the following discussion to section 2.2.4: "Here it should be noted that AERONET Taihu station is located in a more remote area (from downtown Wuxi) than the MAX-DOAS at Wuxi station. The different locations could contribute to a systematic bias between both data sets. However the long residence time of up to several days (Ahmed et al., 2004) and the relatively homogeneous horizontal distribution of aerosols (implied by the weak dependence of AOD on wind direction, see section 3.4.2) implies that the differences between both measurements should be small."

4) Sect. 2.2.5, pages 8-9: the validation results are discussed only in terms of absolute differences between MAXDOAS and correlative data. It would be useful for the reader to have also an idea about the corresponding relative difference values.

Author reply: We added the relative differences (compared to the average values) to the manuscript in section 2.2.4 and the conclusions.

5) a) Sect. 2.2.6, pages 9-10: Is it really useful to show the scatterplots of MAXDOAS versus correlative data and histograms of the absolute differences between MAXDOAS and correlative data for all sky conditions, seasons, and trace gas (TG) and aerosol variables (Figs 15-19 and S20-23) ? I think the manuscript could be simplified here. My

suggestion for the final publication is to remove all the histograms from the manuscript and to present the linear regression and correlation results in a table. The latter could also be presented in a panel like the ones usually used in MAXDOAS intercomparison campaigns for summarizing the slant column density comparison results (see e.g. Figure 6 in Roscoe et al., Atmos. Meas. Tech., 3, 1629–1646, 2010; the authors would show for each TG or aerosol variable, the correlation coefficient, slope, and intercept values in three different subplots, replacing the x-axis by the sky conditions and the elevation angles (colored circles) by the seasons).

Author reply: Great thanks for your good suggestions! We followed your suggestions to re-plot the figures. Please see the description in the reply to your “Major comments”.

b) Based on these comparison results (Figs 15-19 and S20-23), the authors have developed recommendations for determining under which sky conditions which TG and aerosol data products can be used or not. This filter scheme is presented in Table 3. However, nothing is said on the criteria (e.g. which threshold values on correlation coefficients and/or slopes, etc) used in practice by the authors to develop these recommendations. Maybe a panel summarizing the linear regression results as suggested in comment 5a could also support the discussion here.

Author reply: It is hard to quantify the cloud effects on the MAX-DOAS results of aerosols and TGs. Here we combine the results shown in Fig. 8 and 9 (in the revised version) to qualitatively discuss the effect of clouds and give our recommendation. We added more discussion of the cloud effects (as shown in the three figures in the end two paragraphs of section 2.2.5) in order to make our selection more clear.

6) Sect. 2.2.7, page 12, lines 12-13: the total error budgets of aerosol retrievals are simply reported as error of TGs related to the errors of aerosols. This is not correct because the relationship between the aerosol extinction profiles and the TG retrievals is not linear, i.e. a 15% difference in the aerosol extinction profile used in the TG retrievals does not lead necessarily to a 15% difference in the retrieved TG profiles.

This point should be further investigated and corrected in the revised manuscript.

Author reply: We agree with the reviewer. But the quantification of the aerosol effects on TG results is difficult because it depends on the aerosol profile, aerosol properties, profiles of TGs and even observation geometries. Many simulation studies need to be done to acquire a more reasonable estimation on the effects. Although the topic is very interesting and new, but it should be done in a separated work in the future. Thus we would only coarsely estimate the relevant errors of TGs using the assumption of linear propagation of the errors of aerosol retrievals. And the following clarification is given in the manuscript: "The estimations of aerosol relevant errors are rough. A further studies need to be done to acquire a more reasonable estimation by considering aerosol properties, profiles of aerosols and TGs and observation geometries."

7) Section 3.2: Interpreting the retrieved profile shapes should be done with caution given the fact that the average DFS is only around 2. This particularly the case for the HCHO profiles which show a secondary maximum around 1km. A possible explanation is of course the transport of longer-lived VOCs to higher altitudes but with a DFS around 2, one should not totally exclude the possibility of a retrieval artifact.

Author reply: We agree with the reviewer. We added the following clarification in section 3.1 of the revised version: "However it should be noted that VMRs of HCHO at high altitudes are strongly constrained by the a-priori profiles because of the low sensitivity of MAX-DOAS retrievals at these altitudes. More comparisons studies with aircraft measurements need to be done in the future to further quantify the retrieval sensitivities for elevated layers." In my own opinion, we still have confidence on the extensive vertical distribution of HCHO retrieved by MAX-DOAS because of two reasons: 1) the Fig. S9 in the supplement of the revised version indicates the higher vertical extension can still be partly represented even for using an exponential a-priori profile; 2) the large variation amplitude of HCHO VMRs at the altitude around 1km is retrieved from MAX-DOAS observations. It indicates the sensitivity of MAX-DOAS retrievals to the layers at around 1km is still well. Thus we add the following comment in section 3.1:

“Nevertheless we still have confidence on the extensively vertical distribution of HCHO retrieved by MAX-DOAS because of two reasons: 1) the Fig. S9 in the supplement indicates the higher vertical extension can be partly represented even for using an exponential a-priori profile; 2) the large variability of HCHO VMRs at the altitude around 1km is retrieved from MAX-DOAS observations. It indicates the sensitivity of MAX-DOAS retrievals to the elevated layers is still well.”

Suggestions for technical corrections:

*Page 1, line 16: ‘extinctions’ -> ‘extinction’

Corrected

*Page 2, line 26: ‘ground based’ -> ‘ground-based’; this should be corrected throughout the manuscript.

Corrected

*Page 3, line 8: ‘Clemer’ -> ‘Clémer’; this should be corrected throughout the manuscript

Corrected

*Page 3, line 22: ‘humidity controlled’ -> ‘humidity-controlled’

Corrected

*Page 3, line 34: ‘so called’ -> ‘so-called’

Corrected

*Page 5, line 26: ‘long term’ -> ‘long-term’

Corrected

*Page 6, line 11: ‘session’ -> ‘section’

Corrected

*Page 12, line 21: ‘budges’ -> ‘budget’ or ‘budgets’

Corrected

*Page 18, line 10: remove ‘.’ after ‘found:’

Corrected

*Page 38, Figure 7: the ‘2’ of ‘NO2’ on the x-axis label of subplot (b) is cut.

Corrected

*Page 39, Figure 9: remove the ‘,’ after ‘Figure 9:’

Corrected

*Supplement, page 8, Figure S2: There is a problem with y-axis labels of subplots (a) and (b).

Corrected

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