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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. #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 This paper presents the temporal variation and vertical distribution characters of the tropospheric aerosol extinction (AE) and trace gases (TGs, including NO2, SO2 and HCHO in this study) derived from relatively long-term (2011-2014) ground-based MAX-DOAS observations in the Wuxi city, located in eastern China. The authors developed a new profile inversion logarithm (PriAM) and applied it to deal with

their MAX-DOAS measurement data in this study. For the retrieval method, the authors find that large, systematic biases of the retrieved AE profiles and thus AOD can be induced if the seasonal variations of temperature and pressure are not considered. They also show that the traditional geometry approximation could lead to larger biases than the profile integration in the retrieval of tropospheric vertical column densities (VCDs) by analyzing the separated processes in the retrieval from their MAX-DOAS measurement data. The authors further analyze the data retrieved from MAX-DOAS measurements and characterize the seasonal, diurnal and weekly variations of NO2, SO2, HCHO and aerosols. One of their results is the finding of a significant annual decreasing of SO2 for both VCD and surface mixing ratio. The study and its results are interesting, providing important information not only for the development in MAX-DOAS technique, but also for further investigations of urban and regional air pollution issues in eastern China. The manuscript can still be improved before publication if some methods could be described more clearly and some results be presented more concisely. Below are my comments and suggestions in detail.

Author reply: Many thanks for the positive assessment! In order to present the results more concisely, 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) In Sect. 2.3, the authors compare the VCD derived by integrating the retrieved vertical profile (VCD_pro) with the VCD calculated by the so-called geometry approximation (VCD_geo). The relative difference is denoted as Diff_total (see Eq. (3)). To identify the dominating difference (which they called "error") source, they split the total difference (Diff_total) between VCD_geo and VCD_pro into two parts: one is the difference between VCD_geo and VCD_geo_m, denoted as Diff_inversion (see Eq. (4)), and another is the difference between VCD_geo_m and VCD_pro, denoted as Diff_geometry (see Eq. (5)), where VCD_geo_m is calculated by applying the geometric approximation to the modelled dSCD. I would say it is a good idea to make such a trial. But one should be noted that the VCD_pro has been taken as a standard value in the comparison and the retrieved profile is assumed to be true in their evaluation. Are the results shown in Fig. 20 for all the measurements including cloudy and haze-foggy conditions? Since both the VCD_pro and VCD_geo_m are calculated from the retrieved profile, a large bias in the retrieved profile may lead to biases in both Diff_inversion and Diff_geometry as well as their relative contributions.

Author reply: Thanks for your general support of our approach. Concerning your suggestion, we think the problem pointed out is not that serious: if the retrieved profile is quite different from the true profile, the Diff_inversion will increase, but the Diff_geometry will not be impacted (both VCD_pro and VCD_geo_m are based on the retrieved profile). Thus the increased Diff_inversion can present the large errors of the

retrieved profile. We added this information to the end of section 2.3 of the manuscript as follows: "One point need to be clarified that the discrepancy of retrieved profile from the true profile doesn't impact the approach, although both and are as function of the retrieved profile. Because in this case, only will increase, but will not be impacted. The increased present the large errors of the profile inversion."

2) Sect. 3.5.2 can be omitted as some discussions are very speculative and the meaningfulness of results seems to be local. The measurements site of this study were made at a suburban site, located in the industrial area. From Fig 1 and Fig. 23, one can see that the dominating winds come from the NE, which is neither in the direction of Wuxi urban center nor in the direction of the large industrial sources. There is no doubt that pollution plumes from the urban center and the larger industrial sources could affect the measurement site, as shown in Fig. 22. Since the characters of the emission sources from the urban area, including the fractions of pollutants and emission heights, can be rather different from those from the industrial area, the concentrations of trace gases, aerosols and its components as well as their vertical distribution might be different for different plumes. These episode effects can be investigated in-depth explicitly in the future studies considering the focus and length of the paper.

Author reply: Although this study is local and rough, it still shows several general and important results: 1) the dependence of the measured TG VMRs on the wind direction indicates that the dominating sources of the pollutions are local, but not from the long range transport. Also, strong horizontal gradient appears. Because of the expected similar life time, meteorological conditions and emission sources, the conclusion probably fits to the whole YRD region. 2) The study provides an example on how to use ground-based MAX-DOAS observations to find strong emission sources in an urban-size area. 3) The seasonality of the wind dependence of the trace gases, especially for HCHO, indicates the different sources in different seasons. Because of these reasons, we prefer to keep this section. Further explicit studies could be done in the future to further investigate these findings. We added this statement to the end of section 3.4.2

of the revised manuscript.

Technical issues:

P1, L15-16: Change “spatial distribution” to “vertical distribution” and remove “using vertical profiles”. The measurements were made only at one station and might not be used to characterize the (3-D or 2-D) spatial distribution.

Corrected

P1, L28: Change “from the aerosol results” to “for the aerosol results”.

Corrected

P1, L30 – P2, L2: The sentences here need to be rewritten. The phrase “are found” or “is found” occurs so many times here. Better to use them only for the most important findings. The result on wind direction dependency can be skipped as it is only locally meaningful with little information for general chemistry and transport.

We rewrote the paragraph based on the suggestion. We prefer to keep the results about the wind direction dependency (see our reply to the second specific comment).

P2, L8-9: Add “respectively” after “nitrate and sulfate”. Remove “and methane” as methane also belongs to VOCs.

Corrected

P2, L18: Actually, photochemistry of precursor gases was not discussed in the paper of Huang et al. (2014).

We corrected the sentence to “Recent studies found that in megacities in different regions of China most of aerosol particles are from secondary sources, e.g. formed through photochemistry of precursor gases, during haze pollution events (Crippa et al., 2014 and Huang et al., 2014).” Huang et al, 2014 characterized the percentages of different compositions in total aerosols.

P2, L23: Change “Since about 15 years” to “Since about 15 years ago”.

Corrected

P3, L10: The “stability” and “flexibility” issues can be explained a little bit, taking the OE and look-up table methods as example

We add the explanation as “Here good stability means that an inversion approach is robust with respect to the effects of measurement noise. Good flexibility means that it can well retrieve diverse profile shapes.”

P4, L9-12: Use “Section” or “Sect.”, and the same for other places in the manuscript.

Corrected

P4, L13: Change “discussed” to “summarized”.

Corrected

P5, L13-17: I would suggest to rewrite this paragraph as “The PriAM algorithm was originally introduced by Wang et al. (2013a and b). Below we summarize the basic concept of the PriAM algorithm and its implementation settings for this study, while details can be found in Sect. 2 of the supplement. Like for other algorithms, a two-step inversion procedure is used in PriAM. In the first step, tropospheric vertical profiles (in the layer from the ground to the altitude of 4 km) of aerosol extinction are retrieved from the O4 dSCDs. Afterwards, the profiles of NO2, SO2 and HCHO volume mixing ratios (VMRs) are retrieved from the respective dSCDs in each MAX-DOAS elevation angle sequence”.

We rewrote the part based on the suggestion.

P5, L24: “Fig. 7” appears earlier than “Fig. 3”.

We agree with your suggestion to correct the order. However, as described above, we already removed the Fig. 7 in the revised manuscript.

P5, L29: To do (simulate) what with RTM?

We correct it as “the RTM simulations of weighting functions are done at 370 nm”.

P5, L32: For the single scattering albedo, a fixed value of 0.9 is used, or it is allowed to change between 0.85 and 0.95 in the retrievals?

A fixed value is used. To be clear, we corrected the sentence as “The fixed single scattering albedo of 0.9 and asymmetry factor (Henyey and Greenstein, 1941) of 0.72 are chosen according to average inversion results from the Taihu AERONET station from 2011 to 2013 (the data in 2014 is unavailable).”

P6, L18: Fig. S10 should be relabeled as Fig. S8 (its position should also be moved to the front in the Supplement).

We followed this suggestion. We put the Fig. S16 before Fig. S17 in the revised version.

P7, L24: Change “shown” to “as shown”.

Corrected

P8, L16: How are clear sky conditions classified, by AERONET data or by MAX-DOAS data?

The sky condition is identified by MAX-DDOAS observations as described in section 2.2.5 in the revised version.

P9, L24-25: It makes me confusing that the retrieval is based on a “forward model”. Do you mean a “radiation transfer” model or you still have a “backward” model?

We changed “forward model” to “RTM”. Because the SCIATRAN RTM used in this study is a forward model. Both of “RTM” and forward model represent the same thing in this study.

P11, L12-13: I would suggest moving Figs. 16-19 to the Supplement.

Please see the reply to your general comment. We use the new Fig.9 in the revised version to replace the original Fig. 16-19.

P12, L10: Change “45 k” to “45 K”.

Corrected

P14, L5-13: Sect. 3.1 can be skipped.

We moved section 3.1 and Fig. 21 and 22 in the revised version into the supplement. We added a yellow arrow to show the dominant wind direction in Fig. 1b.

P15, L1-2: This sentence can be rewritten, e.g., as “The observed seasonal variations of the different species are related to various processes: the seasonal variations of source emissions, chemical formation and destruction, dry and wet deposition, and atmospheric transport”.

We correct it based on the suggestion as “The observed seasonal variations of the different species are related to various processes: the seasonal variation of source emissions, chemical (trans-) formation and destruction, dry and wet deposition, and atmospheric transport (Wang et al., 2010; Lin et al., 2011)”.

P15, L10-12: It is difficult to understand that there is no seasonal variation for NOx while the SO2 emissions vary by about 20%. Note that the boiler for domestic heating could also make a contribution to the NOx emissions.

Because the contribution of boilers to NOx is only about 5%, but to SO2 is about 20% based on the study of Huang et al., 2011. We agree that we can’t totally ignore the effect of seasonal use of domestic heating on the seasonality of NOx. But its effect on NOx is much weaker than SO2. Thus we modified the test as “It is assumed that about 94% of total NOx emission in the Wuxi region is emitted from the power plants, industrial fuel combustions and vehicles (Huang et al., 2011), which emit similar amounts in different seasons. The contribution of boilers for the seasonal use of domestic heating to NOx is only about 5% (Huang et al., 2011). Thus the seasonal variation of the MAX-

DOAS results cannot be explained by the variation of the NOx emissions. However, the SO2 emissions might vary by about 20% due to the significant contribution of boilers (Huang et al., 2011)."

P17, L33 - P18, L3: Similar to the seasonal variation (P15, L1-2), the diurnal variation can be affected by various factors. The explanation here seems to be very speculative and can be skipped.

We agree on your opinion. Thus we skip the speculative explanation and rewrite it as "Their diurnal variations can be affected by various factors, e.g. the diurnal variation of the emission sources, as well as secondary formation, deposition and dispersion."

P18, L16: The title of Sect. 3.5.1 can be omitted since Sect. 3.5.2 has been suggested to be skipped.

Because we decide to keep the section 3.4.2 in the revised manuscript (section 3.5.2 in the original version), the title of section 3.4.1 (3.5.1 in the original version) needs to be kept.

P18, L17-19: I would suggest to skip over the sentences "Huang et al. (2014) : : :: : : The aerosol in Wuxi close to Shanghai is expected to have similar properties". There are many kinds of properties for aerosols. It is not clear what properties of aerosols are referred to here. The statement that the aerosols in Wuxi have similar properties with those in Shanghai is very speculative.

We agree that there are many kinds of properties for aerosols. Thus we specified the statement as "The aerosols in Wuxi (which is close to Shanghai) are expected to be similarly dominated by secondary aerosol". We prefer to keep it because the previous study motivated our study here. Because the distance between Wuxi and Shanghai is 130km. And the life time of aerosols in the boundary layer is up to several days. The aerosols can be mixed well in the region including Wuxi and Shanghai through a transport of 1-2 days. Thus the dominant aerosol sources can be expected to be

similar.

P19, L18-21: Too speculative.

We think it is reasonable to say that the wind dependence of HCHO indicates the anthropogenic sources of HCHO. Because the natural sources are rather homogenously spread around the measurement site, but the factories which emit VOCs are located in specific areas.

P20, L2: Change "3 km" to "4 km".

Corrected

P21, L18-24: This paragraph can be omitted.

Because we decide to keep section 3.4.2 (see the reply to the second specific comment), the relevant paragraph is needed in the conclusion.

P22-30: Use indented lines for each reference.

Corrected

P31, Table 1: The format of this table looks not good and needs to be rearranged. Try to avoid using the same items in both column and row. For instance, use species for each column, and for the row use Cross section, Fitting interval, Polynomial degree, Intensity, and so on.

We modified the table based on the suggestion.

P34, L9: Change "mean maps of" to "maps of mean".

Corrected

P37, Fig. 5; P39, Fig. 8; P44, Fig. 15: Better to reduce the absolute maximum/minimum values in Y-Axis appropriately so that the differences can be seen more clearly.

We re-plotted the figures as Fig. 6 and 9 in the revised version.

P39-41: Figs. 9-12 can be merged into one figure, with 4 columns for different seasons.

We merged the Figs. 9-12 into one figure (as Fig. 6 in the revised version).

P41-42, Fig. 13: It might be difficult to understand the top panels (colored) of this figure if one had not read the manuscript carefully. It can be more helpful if some words like "primary sky" and "secondary sky" are added, e.g., to the legend, in the figure.

We modified Fig. 13 (Fig. 7 in the revised version) based on the suggestions.

P45-46: It is suggested to move Figs. 16-19 to the Supplement.

We replace Figs. 16-19 by the new Fig. 9 in the revised version (see general comments above).

P48-49, Figs. 21-22: These two figures can be omitted or be moved to the Supplement. Since the seasonal variability of wind directions is not so high, you may consider making a wind rose diagram averaged for all the experiment period and adding it to Fig. 1.

We modified them based on your suggestion.

P58, Fig. 27: The positions of the characters in X-Axis need to be adjusted.

Corrected

P59-60, Fig. 28: This figure can be omitted or be moved to the Supplement.

Because we decide to keep section 3.4.2 (see the reply to the second specific comment), the relevant figure (Fig. 15 in the revised version) is needed to show.

Supp.-P1, L31: With what do NO2 and O4 dSCDS show a systematic increase or decrease?

With the increase of SZA. We correct the sentence.

Supp.-P3, L12: The dSCDs shown here read not as large as two times of the mean RMS.

We corrected the text: They are assumed as two times of the mean RMS.

Supp.-P6, L16: Fig. S9 should be renumbered as its position be moved the place after Fig. S23. Change "the for elevation angles" to "the elevation angles".

We correct it as "the AODs for elevation angles of" in the revised version.

Supp.-P7, Fig. S1; P8, Fig. S2; Fig. 10, Fig. S5: Both RAA and SAA are used. Please check if they refer to the same variable.

We change all "RAA" as "SAA". Because the instrument is pointed to the north, the SAA is the same as RAA.

Supp.-P12, Fig. S8: I did not find a place in the main manuscript as well as in the Supplement that this figure is referred to.

We corrected the manuscript. Fig. S8 (Fig. S17 in the revised version) is cited in section 2.2.3.

Supp.-P15, Fig. S10: This figure should be moved to the front.

We moved it to the front.

Supp.-P19, L3: Change "ds" to "DoF"?

Corrected

Supp.-P7-37: Please try to let the main body figure and its caption to be in the same page.

We followed the suggestion.

Supp.-P39: Use indented lines for each reference.

Corrected

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