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Interactive comment

Interactive comment on "Validation of OMI, GOME-2A and GOME-2B tropospheric NO_2, SO_2 and HCHO products using MAX-DOAS observations from 2011 to 2014 in Wuxi, China" *by* Yang Wang et al.

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

General comments a) Wang and co-authors investigate the quality of satellite retrievals of NO2, SO2, and HCHO over Wuxi in polluted China via a detailed comparison with ground-based col-umn measurements obtained with the MAX-DOAS technique. This technique is sen-sitive to pollution in the lower atmosphere, and Wuxi in the Yangtze River area faces pervasive high levels of pollution from these gases and aerosols. The

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three years of MAX-DOAS measurements collected in Wuxi thus provide a very interesting data set to test the satellite retrievals, and provide guidance on how to use and possibly improve the retrievals. The authors report that the KNMI OMI NO2 (DOMINO v2) product agrees very well with the MAX-DOAS NO2 columns in Wuxi, especially in situations with few clouds. But the KNMI NO2 products from the GOME-2 sensors tend to be overestimated. Because of this overestimation of GOME-2 NO2, also the satellite-derived NO2 diurnal cycle, while correct in sign, is overestimated. Satellite retrievals of SO2 and HCHO from BIRA and NASA tend to be underestimated by tens of per-cents relative to the MAX-DOAS measurements. These findings are relevant to the many users of satellite data interested in obtaining a better understanding of Chinese air pollution. The paper then addresses some of the critical assumptions made in the satellite re-trievals on: the a priori trace gas vertical distribution in the retrievals, the cloud correc-tions made, the aerosol correction, and to what extent this proceeds implicitly via the cloud retrievals that are sensitive to aerosol effects (Leitao et al., 2011; Boersma et al., 2011; Castellanos et al., 2015; Chimot et al., 2016). The comparison of MAX-DOAS and (CTM-derived) a priori profile shapes is a strong and innovative element of the study, and it is interesting to see how replacing the CTM-profiles with the actually observed profiles helps in improving the agreement between MAX-DOAS and satellite retrievals. Profile validation is dearly needed, and this study explores new avenues on how to achieve this, even though the vertical resolution of the MAX-DOAS and model profiles differ substantially. One highlight is that~20% of the SO2 and HCHO underestimation can be explained by the IMAGES profile shapes insufficiently capturing the enhanced SO2 and HCHO concentrations in the Wuxi boundary layer.

Author reply: Many thanks for the positive assessment! We made four important modifications for the paper. Firstly we moved section 3.1 about the coincidence criteria into supplement as section 1. Secondly we moved the section 2.1.2 about the cloud effects on MAX-DOAS observations into the supplement as section 2. Thirdly we rewrote the discussion on the aerosol effects in section 3.5 of the revised version (see general comment b). Fourthly we rewrote the discussion about the influence of the eCF on the shape factor effects on the AMF (section 3.2 of the revised version). This modification is following the specific comment #6 from Reviewer 1.

b) Section 3.6 on aerosol effects on the AMFs is potentially also interesting, but I have serious concerns about the way it has been set up, and the current method does not allow drawing any firm conclusions. The section starts with an analysis of the NO2 discrepancies (satellite minus MAX-DOAS) as a function of AOD. This is relevant, but it does not become clear whether the discrepancies arise because of high AOD, because of residual clouds, or because of aerosols influencing the cloud fractions. Showing NO2 discrepancies only for cloud fraction < 0.1 is inconclusive since these cloud fractions. may represent real clouds, 'effective' clouds, or a combination of the two. To properly attribute the NO2 discrepancies to the effect of the aerosols, the authors should do what they did for Table 2: use MODIS to distinguish the cloud-free, aerosol loaded situations from the situations with residual clouds still present, and focus their analysis on that data cloud-cleared ensemble to rule out the contributions from clouds. The subsequent box AMF calculations are only just a brief sensitivity study for a limited set of situations that is not representative for the large and robust data ensemble collected by the authors over Wuxi. For instance, only one viewing geometry has been tested (P18, L1). Furthermore, how much box AMFs differ between implicit and explicit aerosol corrections depends strongly on the exact assumption of AOT (profile), particle type, NO2 profile, albedo (why always 0.1?), as shown in many previous studies (e.g. Leitao et al. [2011]). None of this becomes clear on page 18, yet the conclusion is drawn that "the implicit aerosol correction typically causes larger bias of the satellite TG VCDs than the clear-sky assumption". This conclusion is based on only a few calculations that do not represent the full range of situations encountered by the retrievals under evaluation. The authors should have been as rigorous as in section 3.3 and replace the implicit aerosol correction by an explicit aerosol correction for the full set of satellite pixels.

Author reply: Based on the comments of the reviewer, we rewrote the whole section

3.5 in the revised version about aerosol effects. One important point is that although the differences of clear sky AMF, implicit aerosol correction, and explicit aerosol correction have been systematically investigated in the previous studies (i.e. Leitão et al. (2010) and Chimot et al., 2016), here we characterize the aerosol effects for typical aerosol properties (profile, optical properties, and corresponding aerosol induced eCF and CTP) for a polluted region. Also, as mentioned by the reviewer the previous studies indicated that the aerosol effect "depends strongly on the exact assumption of AOT (profile), particle type, NO2 profile, and albedo". Thus we completely re-wrote the whole section, and we extended the RTM simulations to five different satellite observation geometries (listed in Table 2 of the manuscript) following the suggestions of the reviewer. The new results are shown in Fig. 17 of the revised manuscript. The new simulations indicate that the aerosol effects depends on the observation geometries, however the main conclusion on the effects of clear sky AMF and implicit aerosol corrections are consistent for different geometries. For the discussion on Fig. 14 in the revised version (Fig. 19 in the original version) about the analysis of the NO2 discrepancies as a function of AOD, we agree with the reviewer that for cloud fractions < 0.1 residual clouds can not certainly be excluded. Therefore we also used an additional criterium of CTP>900hPa, which can exclude residual cirrus clouds. Considering specific low altitude clouds (with either small OD and large geometric coverage or high OD and small geometric coverage) we performed additional simulation studies, which are described in the section 4 of the supplement. Our main conclusion is that for the selected cases the effect of residual clouds is negligible. We added this information to the main text of the manuscript. In addition we excluded the part about the six pure aerosol pollution days, because we can not draw any general conclusion from these cases. We added a new figure (Fig. 15) showing aerosol-induced eCF and CTP derived from the OMI cloud retrieval as a function of the corresponding AOD derived from MAX-DOAS. The reviewer asked the question "why always 0.1?". Here we updated the text as follows "The surface albedo is set to 0.1 for NO2 and 0.05 for SO2 and HCHO simulations based on the averaged value of the surface reflectivity data base derived from OMI by Kleipool et al. (2008) over Wuxi station." And we redo the RTM simulations with these surface albedo for more observation geometries using McArtim RTM.

c) The paper is too long. The section on the coincidence criteria can be shortened considerably. Other studies have investigated these issues, and the findings are probably specific for the Wuxi circumstances anyway. I recommend to move much of section 3.1, including the figures, to the supplementary material and focus on the final criteria, and then refer the reader for justification of these criteria to the supplement. Also sections 3.3 can be shortened; I'm not sure if for each retrieval the discrepancies as a function of cloud fraction need to be discussed (and shown) at length.

Author reply: Many thanks for the suggestion! We followed your suggestion to move the entire section 3.1 into the supplement. And we added a new paragraph at the beginning of section 3 to describe the main conclusions about the coincidence criteria.

d) The systematic dependence of the HCHO spectral fitting uncertainty on the retrieved VCD for GOME-2 is intriguing, and deserves more attention. Why is this exactly? Why would this be different than for OMI? The authors should clarify these issues. Then their decision to only validate OMI HCHO retrievals with fitting uncertainties < 7 1015 molec.cm-2 is questionable, since setting this threshold basically excludes half the data, not just some outliers or misfits. The authors may report that validation results for this sub-set are better than for the full set, as long as those results are also reported, because users of OMI HCHO data typically use all data, not just the sub-set retrieved with SCD uncertainties < 7 1015 molec.cm-2.

Author reply: Unfortunately, at the moment we can't give any confirmed explanation on dependence of the HCHO spectral fitting uncertainty on the retrieved VCD for GOME-2 and the differences compared to OMI. We clarified this in the revised manuscript. Concerning the filter of the fit error, the Fig. 6b in the revised version (Fig. 11 in the previous version) shows the comparisons of the linear regression parameters for the data before and after the filtering. We also add a new Fig. S12 in the supplement to show the effect of the fit error on the daily averaged data. The two comparisons demonstrate that the filter only considerably improves the correlation coefficient, but hardly changes the slopes and y-intercepts. Thus we conclude that it will not impact the conclusion on the systematic bias of the OMI HCHO products. The point is clarified in the revised manuscript. Furthermore, as mentioned in the paper, data with large uncertainty need to be excluded for a further investigation on cloud and aerosol effects. Otherwise the effects will be overwhelmed by the large uncertainties.

Specific comments

1) P3, L17-20: here it should be stressed that methodological assumptions on how clouds and aerosols should be accounted for in the AMF calculation matter, e.g. Lin et al. [2015].

Author reply: We add this finding and the reference to the text.

2) P4, L1: studies investigating the shape factor are not "rare". There are many studies investigating the quality and effect of a priori profiles on retrievals and emission estimates; e.g. Boersma et al. [2004]; Hains et al., [2010]; Heckel et al. [2011]; Barkley et al. [2012]; Vinken et al. [2014]. Regardless, studying the impact of the shape factor remains relevant because profile measurements are indeed 'rare'.

Author reply: We corrected the sentence as "Here it is important to note that many studies already investigated the quality and effect of a-priori SFs on satellite retrievals (i.e. Boersma et al., 2004; Hains et al., 2010; Heckel et al., 2011) and demonstrated that the SF effect on the tropospheric AMFs can dominate the systematic errors of tropospheric satellite products especially in highly polluted (especially urban and industrial) regions (Boersma et al., 2011, Theys et al., 2015 and De Smedt et al., 2015), Nevertheless, because profile measurements are rare, the SF effect is still not well understood in many regions."

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3) P4, L35 and P5, L1-3: the argument in favour of the implicit aerosol correction in the Boersma-2011 paper is made for substantial AOD when particles are mostly scattering, i.e. not unlike cloud droplets. Castallanos et al. [2015] clearly showed that for absorbing particles and high AOD, the implicit aerosol correction breaks down. So the sentence that Castellanos demonstrated that for elevated biomass burning aerosols, the implicit correction does a good job is completely out of place. Their study showed that the implicit aerosol correction compares well with an explicit aerosol correction for low-modest AOD and SSA>0.95. For high AOD and lower SSA, the implicit aerosol correction breaks down, but these situations occur less frequently than the former.

Author reply: Many thanks for this hint! We modified the sentence as "For mostly scattering aerosols at high altitudes the implicit aerosol correction can largely account for the aerosol effect on the TG products (Boersma et al., 2011). However in some important cases (for low altitude aerosols with high AOD and small SSA) the implicit correction might even increase the errors of the AMF Castellanos et al. (2015).".

4) P5, L31: it should be 'heavy fog'.

Author reply: corrected

5) P6, L4-6: it should be clarified if the difference between the geometrical approximation and profile integration is systematic, or that the discrepancies are variable in both directions.

Author reply: We clarified it as "Our previous study (Wang et al., 2016) demonstrated that the tropospheric trace gas VCDs from the full profile inversion are in general much more accurate than those from the geometric approximation. The discrepancy of VCDs between the two methods is systematic and can be mainly attributed to the errors of the geometric approximation, for which the errors can be up to 30% depending on the observation geometry, and the properties of aerosols and TGs. ".

6) P6, L12: Capital S missing in 'sky'.

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Author reply: Corrected.

7) P6, L30: what is the source of information for the 68 x 14 km2 pixel size at OMI swath edges?

Author reply: Many thanks for this hint! We changed the values to 150 x 13 km2, see: Levelt, P. F., van den Oord, G. H. J., Dobber, M. R., Malkki, A., Visser, H., de Vries, J., Stammes, P., Lundell, J., and Saari, H.: The Ozone Monitoring Instrument, IEEE Trans. Geosci. Remote Sens., 44, 1093–1101, 2006b.

8) P7, L5-7: it would be appropriate to refer to Dirksen et al. [2011] here when discussing the data assimilation procedure to estimate the stratospheric background NO2. Similar to OMI SO2 from BIRA, DOMINO v2 can be regarded as the 'proxy' algorithm for the upcoming TROPOMI mission.

Author reply: We added the reference to Dirksen et al. [2011] and also clarified "The retrieval algorithm for DOMINO v2 forms the basis of NO2 retrievals for the upcoming TROPOspheric Monitoring Instrument (TROPOMI) aboard the Sentinel-5 Precursor mission (Veefkind et al., 2012)."

9) P8, L13: suggest to state 'similar data assimilation procedures'.

Author reply: Corrected.

10) P9, L26: what is meant with the 'statistical uncertainty of the satellite data'?

Author reply: We delete "statistical".

11) P11, LL27-28

Author reply:

12) P12, L11-14: with underestimations of50%, it is rather odd to conclude that GOME-2A products are "most accurate" for cloud fractions below 30%. Also the 'recommendation' to use SO2 observations with cloud fractions below 10% is far fetched. One

might as well recommend to not use any SO2 data over the Yangtze area at all in view of the large, systematic biases shown in this study.

Author reply: We modified the description as “Thus we conclude that the cloud effects on both GOME-2A products are appreciable for eCF > 30%. For the GOME-2B BIRA data, an obvious decrease of R2 and slope is found for eCF > 10%, while for eCF>30% largely variable MRDs are found. Thus clouds can considerably impact the GOME-2B BIRA product for eCF > 10%, and more significantly for eCF > 30%. ”.

13) P12, L29: ‘because of the weaker degradation’ than OMI or GOME-2A? Please clarify.

Author reply: We changed the text to “because of the weaker degradation of GOME-2B during the short time after launch compared to OMI and GOME-2A.”.

14) P13, L1: dependencies.

Author reply: Corrected.

15) P13, L4: when suggesting that HCHO products should be used for cloud fractions < 0.3, the authors should be more aware that their recommendation is based on the situation for Wuxi, which is not necessarily representative for situations with enhanced HCHO concentrations elsewhere (just think about the high aerosol loadings). Also, if they make such a recommendation, they should discuss it in the context of what the algorithm providers actually recommend for appropriate use of their data, and what has typically been done in successful applications of the OMI HCHO data.

Author reply: We modified the sentence as “In general cloud effects on the HCHO products become substantial for eCF > 30% for the three satellite instruments. However it needs to be noted that our findings are derived for one location (Wuxi) and might not be fully representative for other locations. The use of the HCHO products with eCF < 40% is recommended by the retrieval algorithm developer (De Smedt et al., 2015).”.

16) P14, L11: ‘latitude range’ should be altitude range, and ‘larges’ should be ‘largest’.

Author reply: Corrected.

17) P14, L11-14: it would be fair to clearly conclude here that the TM4 a priori profile shapes agree well with the MAX-DOAS NO2 profiles in an average sense.

Author reply: We added this finding.

18) P15, L15: please provide more detail on the months in the x-axis of Figure 17; we now only have tick marks for month 5 and 11. Some more specific indication for the bi-monthly averages would be useful.

Author reply: We modified the figure accordingly. Note that the Fig. 17 in the previous version is Fig. 12 in the revised version.

19) P15, L23-24: please clarify why the TM4 NO2 columns are so much lower than those from the measurements. Later on page 16, same for SO2 modelled by IMAGES; why is HCHO from IMAGES doing a good job whereas SO2 is not?

Author reply: We added that "The significant underestimation of the TM4 NO2 VCDs could be due to many factors, most importantly the limited spatial model resolution, which is especially relevant for species with strong horizontal gradients such as NO2 and SO2 (see Figure 1), but also possible errors in the emissions, transport and/or chemical mechanism. The determination of the specific contributions of the different error sources should be the subject of future studies." in the revised version. We also mention that the results of the IMAGES model for SO2 and HCHO need further investigations in the future.

20) P16, L22-24: it would be appropriate to refer to Boersma et al., JGR, 2008 here. That study was the first to investigate the diurnal cycle of NO2 with satellite measurements. Also some more explanation on what causes the diurnal changes in NO2, SO2, and HCHO columns is needed here.

Author reply: We added the reference and now mention that "The diurnal variations can be attributed to the complex interaction of the primary and secondary emission sources, depositions, atmospheric chemical reactions, and transport processes.”.

21) P17, L18: some more information is needed on the ‘clear-sky AMF’ that is applied in SO2 and HCHO retrievals for cloud fractions < 0.1. How is such an AMF calculated – in an atmosphere with Rayleigh scattering only? Or is there some aerosol background assumed in the radiative transfer calculations?

Author reply: ‘clear-sky AMF’ means in an atmosphere with Rayleigh scattering only. We clarified this in the revised version.

22) P19, L24: please clarify what is meant with “cloud effects become significant”. Do you mean that the discrepancies between MAX-DOAS and satellite columns are larger when cloud fractions are larger?

Author reply: Yes. We already clarified it in the sentence before that sentence as “The consistency (correlations and systematic bias) of satellite data with MAX-DOAS results deteriorates with increasing eCF.”.

23) P19, L33-34: suggest to be more specific here and state that IMAGES profiles and TM4 profiles have been compared against MAX-DOAS profiles.

Author reply: We added this information in the revised version.

24) P20, L21-22: the sentence “NO2 satellite products systematically overestimate the magnitude of NO2 diurnal variation” is misleading. The diurnal variation is overestimated because the GOME-2 retrievals are too high, but OMI is in agreement with MAX-DOAS. Suggest to rephrase accordingly.

Author reply: We changed the text to “The systematic difference of RatioSat and RatioM-D can be attributed to the known overestimation of the GOME-2 A/B tropospheric VCD compared to the MAX-DOAS results (see Fig. 12a). This finding also indicates that using GOME-2 and OMI data can lead to wrong conclusions about the diurnal cycles of NO2.”.

25) P20, L30-35: this part is too strong-worded and should be rephrased after the authors have addressed my concerns about section 3.6. The current sensitivity study provides too little ground to base these conclusions on.

Author reply: We modified the section 3.5 in the revised version (section 3.6 in the previous version). Thus the relevant conclusion part is re-written as: Finally we studied aerosol effects on the OMI products over Wuxi station based on the MAX-DOAS observations. We find that the underestimation of the TG VCDs derived from satellite observations for mainly cloud-free observations compared to the MAX-DOAS observations systematically increases with AOD. We also investigate the aerosol effect based on RTM simulations. Here it is also possible to separate the aerosol effect into two contributions: a) the effect of using a clear sky AMF instead of an AMF taking explicitly into account the aerosol effects, and b) the effect of aerosols on the cloud retrievals, which are used in the satellite TG retrievals (implicit aerosol correction). We find that for the measurements affected by high aerosol loads in Wuxi, in general the effect of the implicit cloud correction on the retrieved TG VCDs is much stronger than the difference of a clear sky AMF compared to an AMF taking explicitly into account the aerosol extinction. We also showed that for eCF <10% and CTP >900hPa the effect of residual clouds can be neglected if aerosol extinction is explicitly taken into account. Moreover, the observed underestimation of the OMI NO_2 VCD for large AOD can be well explained by the error caused by the implicit aerosol correction. Therefore it could be reasonable to apply the clear-sky AMFs in the satellite retrievals of TG tropospheric VCDs in case of CTP > 900hPa and eCF<10% if explicit aerosol information is not available.

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