Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-735-RC2, 2016

Atmospheric Chemistry and Physics Discussions

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.

Anonymous Referee #2

Received and published: 11 October 2016

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 column measurements obtained with the MAX-DOAS technique. This technique is sensitive 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 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

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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 percents 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 retrievals on: the a priori trace gas vertical distribution in the retrievals, the cloud corrections 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 $\sim$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.

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.

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.

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.

Specific comments

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].

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’.

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.

P5, L31: it should be ‘heavy fog’.

P6, L4-6: it should be clarified if the difference between the geometrical approxima-

tion and profile integration is systematic, or that the discrepancies are variable in both directions.

P6, L12: Capital S missing in ‘sky’.

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

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.

P8, L13: suggest to state ‘similar data assimilation procedures’.

P9, L26: what is meant with the ‘statistical uncertainty of the satellite data’?

P11, LL27-28:

P12, L11-14: with underestimations of $\pm$50%, 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.

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

P13, L1: dependencies.

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

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typically been done in successful applications of the OMI HCHO data.

P14, L11: 'latitude range' should be altitude range, and 'larges' should be 'largest'.

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.

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.

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?

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.

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?

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?

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

P20, L21-22: the sentence "NO2 satellite products systematically overestimate the magnitude of NO2 diurnal variation" is misleading. The diurnal variation is overes-

timated because the GOME-2 retrievals are too high, but OMI is in agreement with MAX-DOAS. Suggest to rephrase accordingly.

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.

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