Atmos. Meas. Tech. Discuss., 8, C2523–C2529, 2015
www.atmos-meas-tech-discuss.net/8/C2523/2015/

Atmospheric Measurement Techniques Discussions

Interactive comment on "Long-term variability of aerosol optical thickness in Eastern Europe over 2001–2014 according to the measurements at the Moscow MSU MO AERONET site with additional cloud and NO₂ correction" *by* N. Y. Chubarova et al.

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Received and published: 11 August 2015

Review for Atmospheric Measurement Techniques

Title: Long-term variability of aerosol optical thickness in Eastern Europe over 2001–2014 according to the measurements at the Moscow MSU MO AERONET site with additional cloud and NO2 correction

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General Comments:

This paper shows some interesting and useful analyses of the spectral AOT at the AERONET site located in Moscow, Russia (Moscow_MSU_MO site). Specifically, the analysis in this paper shows how the large NO2 amounts in this populous urban area have significant optical depths that must be accounted for properly in the estimation of aerosol optical depth. The local NO2 surface concentration monitoring done at the site combined with modeling and other less frequent NO2 measurements at higher altitudes were utilized to estimate total atmospheric column NO2. These estimated values are much higher than the total column NO2 from the SCIAMACHY satellite retrievals that are utilized in the monthly climatology in AERONET Version 2 processing. Therefore the AERONET Version 2 AOT at 340 through 500 nm (4 channels) are likely biased high at this site due to insufficient accounting of NO2 optical depth. This is an issue that may occur with AERONET data at other major urban region sites with high NO2 levels. Although it was not expected to be a part of this draft of the paper, it is worth mentioning here that the upcoming AERONET Version 3 database utilizes a new monthly mean climatology of total columnar NO2 from the OMI satellite sensor data and that these values are significantly higher than those of the SCIAMACHY database for the Moscow_MSU_MO site.

Another major topic of this paper is the application of additional cloud screening applied to the existing Version 2 Level 2 cloud screened data for the same site, Moscow_MSU_MO. The authors have additionally screened all AOT data where the cloud fraction exceeds nine tenths (i.e. overcast sky conditions) from March through October as determined by human visual observations. This threshold is dropped to greater than 6 tenths cloud cover for the other months (November through February). In my opinion this is a very general and non-rigorous method to screen clouds from data of direct sun observations, especially so for the November-February months. The human observation interval is hourly at this station, therefore measurements made au-

tomatically by AERONET sun-sky radiometers at 15 minute intervals (nominal) may be taken in gaps in cloud cover that occur between human observer estimates. Additionally, the relatively low cloud cover threshold selected for Nov – Feb of >60% does not account for the fact that at times the sun may be un-obscured by clouds when most of the rest of the sky is cloudy. The authors make a repeated point of suggesting that the elimination of a local AOT maximum in February in the AERONET monthly mean AOT climatology is evidence that their cloud screening is effective and valid. However, careful examination of the AOT data for the month of February for all years shows very high AOT values in 2005 and 2006 that are associated with large Angstrom exponents (AE 440-870 $\sim$1.4 in 2006), therefore suggesting that the authors additional cloud screening may have removed actual aerosol observations of fine mode aerosol in February.

Another aspect of the evidence that the authors present for the effectiveness of their cloud screening is the increase by $\sim$0.03 to 0.09 in monthly average Angstrom Exponents (AE) after the additional screening. Although this is undoubted true in many cases (since cloud contamination decreases AE), it is unknown how much of the lower AE in the additionally screened data may be due to cloud processing of particles and hygroscopic growth of particles in the presence of clouds. These well-known physical mechanisms are not mentioned at all in the manuscript, and some discussion of this should be added in the revision. Also missing in the current manuscript is some discussion of the fact that gas-to-particle conversion occurs much more rapidly in the aqueous phase in cloud droplets therefore some of the higher AOT observed during the cloudier time periods may be due to creation of new particles from oxidation of gases such as SO2 to sulfate in the cloud droplets (also formation of nitrates and SOA), in addition to hygroscopic growth (see Jeong et al. (2010; JGR), Su et al. (2008), Eck et al. (2014) for example). These physical mechanisms also need to be discussed in the current paper.

Finally regarding the cloud screening issue, the authors mention that the AERONET

cloud screening is effective for cumulus type clouds but not always for cirrus clouds. I agree with this assessment and this is well known, having been documented in other studies of cloud contamination of AERONET data. However, although the authors mention cirrus contamination they do not apply any checks specific to cirrus clouds, only to total cloud fraction. It is very puzzling that the authors did not reference the papers of Chew et al. (2011; in Atmospheric Environment) and Huang et al. (2012; in JGR), since these papers specifically investigated the cirrus contamination of AERONET AOT data by using lidars that can identify high altitude cirrus clouds. The authors should examine these two papers and include discussion of them in the revised manuscript.

I recommend that this paper be considered for publication after substantial revisions to address the issues I have raised above and also in response to the specific comments below.

Specific Comments:

Abstract, lines 6-8: Please mention here that these are monthly averages of AOT.

Abstract, lines 20-22: The way this sentence is written it suggests that Carbon Monoxide (CO) is an aerosol precursor gas, however it is not. Please rewrite this sentence to clarify.

Page 7845, line 14: There are about 400 AERONET sites, not 200 as stated here.

Page 7846, line 1: "...where the aerosol network is rare" should be modified to "...where the aerosol network is sparse"

Page 7846, lines 16-17: "...full field CIMEL..." should be "...full field of view CIMEL..."

Page 7846, line 23: 670 nm should be 675 nm

Page 7847, line 23: Please mention here the altitude of the NO2 measurements made with the APNA-360 and the data sampling time interval at the Moscow MSU MO site. NO2 has significant diurnal variability since it is a short-lived gas so it would also be

useful to know the data sampling interval.

Page 7848, lines 8-15: Please be very clear that the observer indicates total cloud cover in increments of tenths and that when you say N>9 you really mean that only when N=10 (which is overcast total cloud cover) are the AOT observations screened as cloud contaminated. Also clarify that for November-February when you change the total cloud cover threshold to N>6 you are saying that for N=7, 8, 9, and 10 visual cloud fractions that you then eliminate all AOT measurements for the entire hour interval.

Page 7848, lines 25-27: Please add 'monthly mean' so that this sentence reads 'In some years the monthly mean difference can even exceed 0.1. . .' Also you should give some details about the two months that you mention here, October 2012 and February 2005. It should be noted that the data you eliminated for October 2012 had relatively high Angstrom Exponent, therefore suggesting that you may have removed good AOT observations dominated by fine mode particles, and also that there were only 3 days of AERONET Level 2 AOT data for Oct 2012, even before you eliminated any data.

Page 7849, lines 7-10: You say that water vapor is overestimated in the high cloud fraction observations, but I think this is likely a misinterpretation. It is expected that for high cloud fraction observations that total column water vapor would be higher than average. This is well known since clouds are liquid water droplets or ice crystals that have a relatively short lifetimes since they are continually evaporating or forming in a high relative humidity environment. Clouds also often form in converging air that contains higher moisture content.

Page 7850, line 8: Please give the distance in kilometers between the Moscow and Zvenigorod sites in this sentence.

Page 7851, lines 6-7: Please add some information in this sentence on why the NO2 lifetime is longer in winter (i.e. photochemistry, etc.).

Page 7852, lines 10-11: Please elaborate what second derivative you are referring to

in this sentence, the 2nd derivative of ln AOT versus ln WL or something else, and also provide a reference to clarify.

Page 7853, lines 25-28: In Figure 4 you have plotted the AOT versus wavelength in linear scales but the standard and most common analyses typically plot these in logarithmic coordinates (both x- and y-axes). You say that the revised AOT spectra are smoother (in linear coordinates), but are they also smoother when plotted in logarithmic space?

Page 7855, line 21 and Table 2: You mention December AOT is this sentence, but is the data sampling sufficient in that month to make any definitive conclusions? In Table 2 you show only 4 days of data for the month of December (total for all years!), while all other months have much more data, ranging from 33 days to 290 days. If the month of December truly has only 4 days of AOT data (revised data set) then I would suggest that you remove any discussion of differences in the December monthly means since a 4-day data sample is not statistically robust enough to make any useful conclusions.

Page 7857, line 20: Similar to the abstract, the way this paragraph is written it suggests that carbon monoxide (CO) is an aerosol precursor gas, however it is not. Please revise to clarify.

Page 7858, lines 7-10: Similar to above, in page 7857, you suggest in this sentence that CO plays a role in aerosol generation. Please explain or clarify here.

Page 7859, lines 4-7: You suggest here that the standard AERONET data set overestimates AOT at 500 nm by up to 0.03 due to cirrus contamination at the Moscow_MSU_MO site. However your additional data screening is done only based on total cloud fraction, so you cannot specify that cirrus clouds were the only reason. Of course your statement also does not consider that AOT may actually be higher in some high cloud fraction observations due to aerosol humidification growth, cloud processing, or new particle formation in clouds. Large-scale convergence that results in cloud formation may also in some cases result in higher aerosol concentrations and

higher AOT in the presence of clouds (see Quass et al. (2010; ACP).

Page 7859, lines 7-10: Please also add that your much more strict filter of human observed cloud fraction >60% in winter also guarantees that more cloud filtering is done in winter and therefore highly likely to have larger differences with the standard AERONET values.

Page 7859, lines 11-18: For the record, the AERONET project would never apply such a general and non-rigorous cloud screening as suggested in this sentence, based on human visual observations of total cloud cover. Additionally, cirrus clouds (and other types of clouds as well) are often discontinuous in space therefore a station as far as 60 km from an AERONET sun-sky radiometer may often have a significantly different cloud fraction. Additionally in this paragraph you seem to ignore the fact that your cloud-screening test is based on only total cloud fraction, not cirrus amount.

Page 7859, lines 17-18: The citation in this sentence given as (Radiation in cloudy atmosphere, 1984) should be (Feigelson, 1984).

Page 7860, lines 13-15: Please be careful about generalizing here, since the 675 nm channel is in the visible and does not have any NO2 absorption, so the only changes in that wavelength are due to your revised cloud screening, therefore similar to the near infrared spectral range.

Figure 8 caption: Again you list carbon monoxide as one of the “main aerosol precursors”, therefore this caption needs to be corrected or CO (Carbon Monoxide) removed from the graph.

Interactive comment on Atmos. Meas. Tech. Discuss., 8, 7843, 2015.