

We thank MJ Jeong for his insightful comments and thorough reading of our manuscript.

Comments related to cloud screening/contamination from MJ Jeong:

For the analysis on the cases with enhanced (fine-mode) AOD in the vicinity of clouds, are cloud-contaminated measurements (i.e., thin clouds/cloud edge placed in the line of sight between the sun and Cimel) allowed?

Significant parts of this study rely on SDA, but it is not clear how authors could make sure cloud-contaminated portion of radiance contributes only to coarse-mode AODs, leaving fine-mode AOD intact from cloud-contamination. While it was briefly mentioned in the Instrumentation and methodology section, more details would help readers greatly.

Also, when thin parts of clouds interfere the sun-photometric measurements, stronger (than that for aerosols) forward scattering can be expected due to presence of large cloud particles. One would wonder how it can influence the measurements and the analysis in this study. I would like to suggest authors to include discussion about this matter in the text.

In Fig 5, for such circumstances, clouds should have obscured (or interfered) the measurements of (SDA) AOD for several AERONET sites, but it is not clear when and where it happened. I do not see many of spikes or peaks of coarse mode AOD for many sites. It would be great if authors can mention about this in the manuscript.

In response to MJ's comments above the following paragraph will be added at the end of Section 3.1:

It is noted that there is substantially less cloud contamination from cumulus clouds versus cirrus clouds in the AERONET version 2 Level 1.0 AOD data. This is due to the significantly larger temporal variance of cloud optical depth (COD) in cumulus and higher optical depths of most cumulus clouds compared to most cirrus clouds. Thus the AOD triplet variability (the principal cloud screening check) typically shows significantly more temporal variance due to cumulus cloud presence than for cirrus clouds. Of course for all clouds the COD is underestimated relative to true cloud optical depth due to strong forward scattering by large (relative to aerosol size) cloud droplets or ice crystals into the Cimel instrument field of view. However this effect is less for typical water droplets in cumulus clouds (~20-40% underestimate) than for larger ice crystals in cirrus (~ up to 100% underestimate) as shown by Kinne et al. (1997) for simulation of the effects of cloud effects on sunphotometer measurements. Additionally, although the Level 1.0 AOD data do not have the AERONET cloud-screening algorithm of Smirnov et al. (2000) applied, there is still a basic filter of large temporal variance of the signal applied to all Level 1.0 data. The direct sun measurement data are not included in the AERONET Level 1.0 data set if the variance of the raw signal is very high within the triplet sequence. The variance threshold applied is based on the root-mean-square (RMS) differences of the three direct sun triplet measurements relative to the mean of these three values. If the $(\text{RMS}/\text{mean}) \times 100\%$ of the triplet values is greater than 16% then the data are not used for computation of AOD and the data do not appear in the Level 1.0 data set. This temporal variance threshold primarily removes data that are affected by clouds with large spatial/temporal variance in COD. This effectively removes much of the cumulus cloud contaminated data, although some of the thinner edges with lower COD do remain in the data, see the decreases in Angstrom Exponent in Figure 6b and the increases in coarse mode AOD in Figure 8b for examples.

Additionally, as noted by MJ in his review, the following sentence in section 2.1 addresses how SDA can detect cloud contamination:

“The AERONET data in Figure 2 were not screened for clouds (Level 1; see section 2.1 below), since O’Neill et al. (2003) have shown that SDA identifies cloud optical depth as the coarse mode AOD component.”

In addition to this sentence we have added the following to section 2.1: Analysis by Chew et al. (2011) of AERONET measured spectral AOD in conjunction with lidar data in Singapore has shown that the SDA technique effectively separated the coarse mode (cirrus cloud contamination, as identified by lidar) from the total optical depth without affecting the fine mode component. Additionally, Kaku et al. (2014) have verified that the SDA technique is also effective in separating the fine and coarse modes from in situ spectral optical measurements.

Chew, B. N., J. R. Campbell, J. S. Reid, D. M. Giles, E. J. Welton, S. V. Salinas, and S. C. Liew, Tropical cirrus cloud contamination in sun photometer data, *Atmospheric Environment* 45 (2011), 6724-6731, doi:10.1016/j.atmosenv.2011.08.017

Kaku, K. C., J. S. Reid, N. T. O’Neill, P. K. Quinn, D. J. Coffman, and T. F. Eck, Verification and application of the extended Spectral Deconvolution Algorithm (SDA+) methodology to estimate aerosol fine and coarse mode extinction coefficients in the marine boundary layer, *Atmos. Meas. Tech. Discuss.*, 7, 2545–2584, 2014, www.atmos-meas-tech-discuss.net/7/2545/2014/, doi:10.5194/amtd-7-2545-2014

Also in Section 3.1 (in reference to Figure 3a), in the existing text we mention gaps in AOD data (level 1.0) due to the presence of cumulus clouds.

“However, the afternoon AOD data at the site show large gaps, for example most of hour 1800 and also from ~2040-2220 UTC, and satellite data (both GOES and MODIS Aqua (see below)), indicate significant cumulus cloud cover.”

Another part of the existing text where the ability of SDA to discriminate clouds from aerosol is in section 3.3:

“The SDA retrieval of fine and coarse mode AOD (Figure 9b) shows that fine mode dominates the temporal variance except for the first observation point that is cloud contaminated. For this first point the cloud is cirrus (altitude >8 km; Fig 9c) and therefore at much higher altitude than the principal aerosol layer. Note that the fine mode AOD for this one mixed aerosol-cloud observation is consistent with the subsequent non-cloud contaminated observations.”

Fig. 8c. Does “Range” stand for altitude? Then, it would be better to interpret the figure if the axes of the plot can be switched.

Yes, range does stand for altitude. The authors prefer to maintain the current plot axes.

The MPLNet image in Fig. 9c shows very interesting feature – enhanced backscattering within the boundary layer (esp., 0.5 – 1.7 km a.g.l.) several hours before the vigorous cumulus genesis in the late afternoon. The feature may not be associated

with cloud-processing as it seems that there was no cloud at that time span. It could be associated with newly generated aerosols or aerosols from transport/convergence. It will be nicer if more discussions about the feature can be provided in the manuscript.

Yes the MPL image shows enhanced backscattering from 12 through 18 UTC, before clouds formed. We do not have the information to identify the source of these aerosol dynamic changes, although convection without cloud formation and also sea breeze circulations may have contributed. It is noted that the AOD changes (Figure 9a) associated with these vertical profile changes are much smaller than those that occur during cumulus cloud occurrence.

Fig. 12. I wonder if the effect of the enhanced light scattering near bright clouds (so called 3D cloud effects; e.g., Varnai T., and A. Marshak (2009). MODIS observations of enhanced clear sky reflectance near clouds Geophys. Res. Lett., 36(L06807). doi:10.1029/2008GL037089) is considered in MAIAC algorithm. Satellite aerosol retrievals for the pixels near clouds could be positively biased due to this effect. A caution should be exercised for interpreting satellite-based AOD data near clouds.

In response to this comment (above), the following paragraphs have been added to the manuscript at the end of Section 3.6:

Presently, 3D effects are not considered directly in MAIAC retrievals. Indeed, the side-scattering from clouds (usually in the backscattering direction) creates an additional source of radiation which is further scattered into the sensor's field of view mostly by Rayleigh scattering. This effect is noticeable at short wavelengths (e.g., in the Blue band) and can increase retrieved AOD. While MAIAC does not directly account for these 3D effects, it still filters the suspicious cases. The filter is based on the Angstrom Exponent computed from AOD independently retrieved at $0.47\mu\text{m}$ and $0.66\mu\text{m}$ over dark surfaces. The 3D effects would give unrealistically high Angstrom Exponent values (e.g., above 3.5), which are filtered.

A different type of 3D effects is frequently observed over brighter surfaces in conditions of broken cloudiness. It is caused by multiple scattering between the surface and clouds, with photons eventually re-directed into the gap between the clouds. In these cases, the Red-band enhancement is significantly larger than the Blue-band enhancement (in proportion to the surface reflectance), and therefore, such cases produce both enhanced AOD and very low Angstrom Exponent typical of dust. Such cases are challenging and we are still working to resolve these in MAIAC retrievals.

We thank Referee #2 for his/her useful and constructive comments and careful reading of the manuscript.

Minor comments:

p.18809. l.25-50, presents some interesting results on chemical effects (organics, inorganics). Could you please elaborate?

Yes, in response to your comment further elaboration is provided as follows:

The previous sentence was:

The analysis of the aerosol chemical composition show a strong increase of the water-soluble organic carbon (WSOC) by a factor of 2 (up to $6 \mu\text{gm}^{-3}$ after the cloud formation) within the layer from 1 to 2.5 km, while the sulfate and nitrate concentration do not show any significant evolution.

The new sentences replacing this sentence is:

The analysis of the aerosol chemical composition shows a strong increase of the water-soluble organic carbon (WSOC) by a factor of 2 (up to $6 \mu\text{gm}^{-3}$ after the cloud formation) within the layer from 1 to 2.5 km. According to previous studies (Blando and Turpin, 2000, Ervens et al., 2011), the formation of SOA through cloud processing is highly plausible. Hennigan et al. (2008) found that the fraction of WSOC in the particle phase increases sharply with RH. The positive correlation with liquid water rather than with organic matter (Hennigan et al., 2009) suggests that aqueous reactions was the dominant SOA formation process rather than gas-phase reactions. Moreover, the inorganic analysis, integrated over a 3-5 minutes period (limited to few data point per profiles), show an increase by a factor of 1.5 of the sulfate concentration (from $1.6 \mu\text{gm}^{-3}$ before the cloud formation up to $2.5 \mu\text{gm}^{-3}$ after the cloud formation), while the nitrate concentration do not show any significant evolution. This study case could be used to test the new schemes described by Lim et al. (2010), which take into account the wet processes for SOA formation, and improve numerical models to better take into account the cloud processing products in this particular area.

Blando, J.D. and B.J. Turpin, Secondary organic aerosol formation in cloud and fog droplets: a literature evaluation of plausibility, *Atmos. Environ.*, 34 (10), 1623-1632, doi: 10.1016/S1352-2310(99)00392-1, 2000.

Hennigan, Christopher J., Bergin, Michael H., Dibb, Jack E., and Weber, Rodney J., Enhanced secondary organic aerosol formation due to water uptake by fine particles, *Geophys. Res. Lett.*, 35 (18), L18801, DOI: 10.1029/2008GL035046, 2008.

Hennigan, C. J., Bergin, M. H., Russell, A. G., Nenes, A., Weber, R. J., Gas/particle partitioning of water-soluble organic aerosol in Atlanta, *Atmospheric Chemistry And Physics*, 9 (11), 3613-3628, 2009.

Lim, Y. B., Tan, Y., Perri, M. J., Seitzinger, S. P., Turpin, B. J., Aqueous chemistry and its role in secondary organic aerosol (SOA) formation, *Atmospheric Chemistry And Physics*, 10 (21), 10521-10539, doi: 10.5194/acp-10-10521-2010, 2010

It could be mentioned in the conclusions that this data set would be ideally suited to test models in case studies.

This is an excellent suggestion and we have now done so at the end of the Conclusions section.

Point 6 of the conclusions could mention that this may have led to underestimation of AOD in AERONET and satellite retrievals.

We also agree with this suggestion and have added this to Point 6 of the conclusions.

Some of the figures are too small (should be improved in ACP publication).

We will consider this in the final figure preparation.

The text is full of abbreviations, which is not a problem, but untrained readership could be helped by an appendix listing them.

We agree that this is not a problem, and choose not to include an appendix.

Please check for typos:

We have found and corrected all of the typos/mistakes that the Reviewer #2 has identified below:

p.18788, l.23, meteorological

p.18789, l.4, Lelieveld; l.12, within minutes

p.18797, l.6, as a consequence

p.18800. l.9, near solar noon

p.18801, l.3, retrievals of the

p.18809, l.25, shows

p.18815, l.22, slight

p.16616, l.2, in -> from

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