

Review of Zheng et al., "Analysis of influential factors for the relationship between PM2.5 and AOD in Beijing" submitted for publication in ACP, May 2017.

### General comments

The authors investigate the relationship between AOD and PM2.5 using 4 years of data at a single site in Beijing. In particular, they investigate the influence of factors such as PBLH, RH, wind speed and direction and aerosol type. The rationale for this study is to explain the variability of the PM2.5/AOD relationship because of the possible application of satellite observations of AOD for PM2.5 monitoring.

In this study the authors use AOD from MODIS-Aqua (L2, C5.1 from the DB algorithm) and AERONET (L2 AOD, AE, FMF and SSA; these parameters were used to classify aerosol type) and aerosol profiles from CALIOP, together with hourly PM2.5 data from the U.S. Department of State which are freely available from a website. However, no information is provided on how these data were measured and what parameter is reported (dry/wet aerosol). Also, a disclaimer on this website ([http://www.stateair.net/web/assets/USDOS\\_AQDataFilesFactSheet.pdf](http://www.stateair.net/web/assets/USDOS_AQDataFilesFactSheet.pdf)) states that the data have not been validated or quality assured. Hence the authors should clarify what they have done to do this, and why these data were selected for their study over other possibly available PM2.5 data. The PM2.5/AOD ratio was analyzed using 3-hourly meteorological data provided by the CMA (CF, TP, RH, wspd and WD. PBLH was extracted from ECMWF interim re-analysis data.

This is a very comprehensive suite of experimental data. All of these are obtained at different times and with different spatial scales and, for ground-based measurements, at different locations. Hence the data sets need to be colocated in both space and time before any analysis can be undertaken. Although locations indicated in section 2 are within 1 degree, an inhomogeneous megacity environment, both as regards surface characteristics and aerosol sources, may result in substantial spatial variation of the aerosol characteristics. These points need to be addressed in the MS.

Overall, the manuscript shows the influence of various factor on the PM2.5/AOD relationship, but presentation lacks clarity and many things are left unexplained as detailed in my comments below. I miss clear conclusions (only a summary is provided at the end) and how the findings can be combined to provide a relationship between PM2.5 and AOD, if that would be possible even for a single site.

In general, the figures would be more interesting if they were easier to read, in some cases the text is too small. Furthermore, although a comparison is made between MODIS and AERONET data, in the main part of the MS only AERONET and CALIPSO data are used. Hence I do not understand why MODIS is included.

### Detailed comments

p4, lines 2-3 specify modifying: scattering and absorption ?

5, 3 usually it's not the extinction coefficient that is provided but the AOD, i.e. the integrated extinction over the whole column

5, 10 spelling van Donkelaar (also elsewhere in the MS)

5, 11-13 these statements need some references

6,18 what do you mean with 'and so on'? Either specify or remove

- 8, 11 Was AERONET version 2 or the new version 3 used?
- 9, 8 No, AOD is not extinction multiplied by layer depth, but the integral of the extinction over the whole layer. This is different when extinction is not constant with height, as is usually the case.
- 9, 20 why was MODIS C5.1 used and not the newer C6?
- 10, 2 why was Deep Blue selected and not dark target? Or the merged DB/DT AOD product from C6?
- 10, 14 is it the optical thickness per unit mass concentration, or inversely the mass concentration per unit AOD, as eq 1 says?
- 10, 16-19 it is not clear what extinction capability means: extinction per unit mass, or mass extinction coefficient? Or extinction efficiency? Or ....? Is it really true that for the same PM<sub>2.5</sub>, the extinction is weaker for the same AOD? Or is it more complicated and does size distribution and RH have an influence?
- 11, 3 discrepancies or differences?
- 11,5 FMF is the fraction of the AOD due to fine particles (smaller than 1 micrometer); AE is exponent for the power law describing the wavelength dependence of the AOD
- 11, 9 SSA is the ratio of the scattering coefficient to the extinction coefficient (=scattering + absorption)
- 11, 12 replace According to with Following Lee et al. (2010)
- 12, 2 what do you mean with 'since the high percentage of '?
- 12,6 replace ' the method' with 'the classification method'
- 13,6 what does it mean when MODIS correlated best, considering the bias, i.e. considering that MODIS AOD is too high?

Tables 1 and 2: what parameter is listed in the first 2 columns? How is bias calculated?

- 14, 1 suggest to change to: the hygroscopic growth factor  $f(RH)$  is defined as the ratio ... ; is it really the hygroscopic growth factor? This factor would relate to particle size, which indeed is one of the underlying factors, in addition to refractive index, influencing scattering; however, this article is about AOD, which is scattering and thus the scattering enhancement factor should be considered, see for instance Zieger et al., (2015) (Low hygroscopic scattering enhancement of boreal aerosol and the implications for a columnar optical closure study, Atmos. Chem. Phys., 15, 7247-7267, doi:10.5194/acp-15-7247-2015, 2015) and references therein for a discussion on the subject and the  $f(RH)$  for different aerosol types in Europe.
- 14,5 how was eq 2 derived? Has this eq been compared with measured hygroscopic growth curves, or better scattering enhancement?
- 14, 6-8 I don't understand this explanation since both the MODIS and AERONET AOD are derived in ambient conditions, and for the same air mass when well colocated. Hence hygroscopic growth cannot be the physical reason for larger MODIS AOD than AERONET AOD, since these should be the same. More likely these are retrieval errors. Since in this MS CALIPSO AOD is the main source for analysis, should a similar discussion be made for CALIPSO?

- 14, 13 what is the consequence of this assumption? Uniform mixing would imply that dry aerosol particles, water vapour and potential temperature are well-mixed, but RH would in that case increase with height and thus all aerosol parameters that vary with RH. In addition, often a scale height is used to account for an aerosol gradient.
- 14, 19 this assumption implies that there are no disconnected aerosol layers and thus may introduce errors in experimental conditions where these may occur, as revealed by lidar.
- 15, 2-3 I would agree that an increase of PBLH could result in the decrease of surface PM2.5, in the absence of sources and sinks for PM2.5, but that does not follow from eqs 3 and 4, unless PM2.5 column would be assumed constant.
- 15, 7 opposite trend, suggest to replace with anti-correlated; what are the colour bands in Fig 4a?
- 15, 11 I assume that this is a typo and fig 4a is meant, but still don't understand the sentence
- 16, top para: here AOD dry is plotted where AODdry was obtained using the correction factor given by eq 2, which has to be explained. However, as mentioned above, RH is not constant with height, and hence  $f(\text{RH})$  also varies with height. How was this accounted for? It seems that the good correlation between AODdry and PM2.5 is somewhat fortuitous. Does this good agreement lead to the conclusion that PM2.5 was measured dry (at low RH)? See also the discussions on vertical variation of aerosol profiles later in this MS.

Figs 5 and 6 both show the diurnal variation of PBLH and RH, for different seasons averaged over many years, for the whole day in Fig. 5, and for 5-20 in Figure 6 (a-d). I would expect that the curves for each season are exactly the same for the overlapping time periods. Could the authors explain the differences?

- 17,4-8 why is a linear relationship expected between AOD and PM2.5, while the authors showed already in the above that there is not a good correlation, as also shown in Figs 6a2-d2? Why are these plots shown, why not only the corrected AODdry, since the necessity of that correction was discussed above? Furthermore, Figs 6a3-d3 do not show the R2, they show time series which show that part of the time AODdry and PM2.5 trace each other, but at other times not; furthermore, these plots show the offset, i.e. AODdry much larger than PM2.5, which is in contrast to what was shown in Figure 4c, where the time series are much closer. Could the authors explain the differences? The PM2.5 scales in Figs 4c and Fig 6 a3-d3 are much different but plotting on the same scales would make the discrepancy even larger.

- 17, 10-12 why only this narrow time frame (11-17 LT)? why does this limit the diurnal and seasonal variation on PM2.5/AOD?

- 17, 13 Is the classification based on the scheme described on p. 11? Please refer to that scheme, and if not provide a reference. In Figure 7 are 8 aerosol types are shown whereas on p. 11 there are only 6, so I suspect that the some other classification than that on p. 11 is used

- 18, 5 what is 'heavily non-absorbing aerosols'? This type neither occurs in Fig 7 nor on p. 11.

Figures 7 and 8 present the same aerosol types, but with a different colour scheme. It would be much easier to read when the colour schemes are the same. Please change.

- 18, 14 extinction capacity? On p. 10 extinction capability was used. Is that different? I don't understand either of these terms, see my comment on p. 10.

- 18, Fig 8 and Table 3 discussion: rather than only providing correlation coefficients, the authors should discuss the reasons for the observed differences. There are two fundamental reasons for the variation of the

PM2.5 / AOD ratio: PM2.5 does not include coarse particles while do they do contribute to the extinction at wavelengths in the visible, and thus to AOD. In addition, the extinction efficiency peaks for a particle size which depends on the ratio between particle size and the wavelength.

19, 1-5 Figure 9 is too small to read the numbers, but it is clear that there are large differences in the slopes from one aerosol type to another, as also stated on line 9. Hence I don't understand where the numbers given on line 3 come from.

19, 6 why does high RH increase the extinction efficiency? Since the refractive index of water is lower than that of most other materials, would the condensation of water vapour lower the extinction efficiency?

19, 10 Similar as on line 3, I do not understand where these numbers come from. I cannot find them in Fig 9

19, 12 Figure 4 does not show that the slope decreases with particle size.

19, 15 this conclusion is clear, if only looking at Figure 9. However, I do not understand how the discussion in this section supports this conclusion.

20, 13-15 this is a very surprising conclusion, regarding the location of Beijing with near-by mountains blocking the circulation, pollution advected from the south and clean air from the north, as well as the transport of desert dust aerosol in the spring. What leads to this conclusion? I don't see it in Figure 10. And also the first 5 lines on top of p. 2 contradict this conclusion.

20, Fig 11 does not show haze occurrence but AOD and PM2.5.

22, 2 and fig 13: what is actually plotted: the axis annotation reads AOD, but AOD thus far was the column-integrated extinction, i.e. independent of height. May be extinction is plotted? Should these profiles reflect that the PBLH varies with season, and thus also the aerosol vertical distribution vary with season? The strong decrease with height contradicts the assumption of a uniform mixing (p. 14).

22, 9 as for Fig 13, I don't understand what is plotted here

22, 18-21: same comments as for the top of the page. I don't understand what is shown here, and hence it is hard to comment. I also don't understand why in summer, with peak AOD at a height of 516 m, is concluded that most aerosol is below that height. Integration of the profiles below and above 500 m would give a number supporting this conclusion.

23, eq 5 This eq needs some more explanation. Apparently the authors assume here that CALIPSO AOD underestimates the true value (fig 3)? And therefore they scale to AERONET AOD?

23, 15 how is top of atmosphere defined?

Figure 15 would be easier to understand when the AOD was plotted on the same scale. Note that, PM2.5 is plotted vs AOD, not AOD vs PM2.5 as the caption says.

24, 1-3 this conclusion is clear and follows from Figure 13, and hence Fig 15 is obsolete, including the explanation on p.23; it also shows that the uniform-mixing assumption on p 14. is not valid for aerosol.