Interactive comment on “Tropospheric aerosol scattering and absorption over Central Europe: a closure study for the dry particle state” by N. Ma et al.

Anonymous Referee #1

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Comments on: Tropospheric aerosol scattering and absorption over Central Europe: a closure study for the dry particle state N. Ma, W. Birmili, T. Müller, T. Tuch, Y. F. Cheng, W. Y. Xu, C. S. Zhao, and A. Wiedensohler

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This paper is an analysis of a 4 year data set of aerosol parameters (scattering, backscattering, and absorption coefficients, SMPS) measured at Melpitz. First a closure study is performed to compare scattering and backscattering coefficient measurements with the results of a Mie code calculation based on SMPS measurements. Second,
the Mie code was used to calculate “true values” of the scattering and backscattering coefficients without the truncation and non-Lambertian error. These results allow to make assumption on the validity of the Anderson and Ogren (1998) algorithm that correct for these two systematic errors. Third, statistical values and seasonal cycles of the aerosol parameters are presented. Finally the variations of the aerosol parameters as a function of air masses origins are analyzed.

The paper presents a worthy 4-years dataset and some interesting analysis. There is however different points that have to be revised:

- Air mass classification: it is first quite difficult to obtain an idea of the classification scheme presented, since the publication about it is planned in a forthcoming paper. Second, there is already several synoptic weather classification schemes over Germany and Europe (see for ex the final report of the Cost 733 action: http://cost733.met.no/FinalEvent.html), which were tested and used in several studies. It would be worth to use an already well-known and accepted classification scheme instead of developing a new one. The resulting 13 air mass types are not completely self-explaining. Several questions remain open: what are the cold/warm season? How is the evolution of the mixed layer defined? What is the effect of the normalization of the vertical profiles (which ones, P, T, humidity, wind?)?

- The authors have used SMPS measurements associated with Mie codes to obtain the closure study. Other assumptions given later in the paper could be tested/explained by SMPS results and Mie codes, but the authors prefers to cite other papers or rely on global explanations. Here are several examples: 1) each time where primary or secondary aerosol production are taken as an argument, SMPS could help to show the relation between, for example, the lower single scattering albedo in winter and the secondary aerosol production(p. 27827). 2) the relation between $b$ and the decrease in aerosol mean size (p. 27827) 3) the difference of the seasonal cycles of the scattering Angström exponents calculated with different pairs of wavelengths (Fig. 4)
Generally the choice of the figures is not appropriate: some assumption could be validated by making correlation between different parameters (for ex. between b and the effective radius of sub-micron aerosol or their volume fraction), whereas several figures does not bring a lot of informations (Fig. 1 and 3)

Minor comments:

- p. 27812 line 6: specify that you are speaking of the scattering Angström exponent
- p. 27813: update your results with the IPCC 2013 report
- p. 27813 and 27825: I do not agree at all that only seldom studies have been published on at least one year of aerosol optical measurements. See first the ebas data base listing the data regularly submitted or the recent papers on long-term trend analysis of aerosol optical properties and number concentration (Asmi et al., ACP 2013, Collaud Coen et al., ACP 2013) that obtain more than 20 datasets with more than 10 years of measurements. There is certainly a lot of publications involving all these datasets.

- §2.2: it is not clear is the truncation and non-lambertian error correction were applied to the scattering and backscattering coefficients before the closure or not

- p. 27821: the uncertainties were set according to Ma et al., (2011) dealing with aerosol measurements in China: are the aerosol types and the instruments used in both studies sufficiently comparable to have the same uncertainties?

- p. 27822 §3.1.3: please use the same structure of sentences to explain how are estimated the boundary of the scattering and the backscattering.

- p. 27822: in my opinion, the stability of the measurement cannot be estimated by the presented closure study. The stability has to be tested as a function of time and not only as a percentage of measured data explained by the closure study. For me also, the aim of a closure study does not seems to be the validation of the measurements.
- p. 27823: the difference between the regular and modified Mie model are not explained

- §3.2: this study seems to me the most interesting point of the paper. It should by however compared to the results at other sites. The measurements were done with a PM10 inlet. What is the influence of the size cut on the results of Fig. 2?

- §3.3: Fig. 3, 4 and tables 7 and 8 are somewhat redundant. The comparison with mean values found at other stations does not bring a lot of informations since it is quite difficult to estimate if these aerosol measured at these station are comparable. For example, what is the meaning of Finokalia (marine station) scattering coefficient being 10% higher than at Melpitz? Moreover, there is a lot of papers published of the time series of Puy du Dôme, Hohenpeissenberg, Jungfraujoch, Cabauw, Mace Head, Sonnblick, Ispra, K-Puzta,… The comparison of the single scattering albedo would have been more valuable.

- p. 27825: the wavelength difference between 2 measurement sites does not impede the comparison of the data. An absorption Angström exponent of -1 is usually applied to obtain the data at the right wavelength.

- p. 27826, line 12: the single scattering albedo does not have a clear annual cycle!

- p. 27826, lines 14-18: the high pollution events during winter are directly visible in Fig. 3 and not only in the statistics.

- p. 27826: the single scattering albedo cycle seem to have higher values from March to June, which does completely correspond to “spring-summer”! Is the secondary aerosol production visible in SMPS measurements?

- p. 27827: in winter, is there also a relation between the low single scattering albedo and greater contribution of carbon due to larger combustion processes (heating, fires,…)?

- p. 27827: the discussion about the scattering Angström exponent should differentiate
the cases calculated with the various wavelength pairs. The SMPS associated with Mie code could also explain the different annual cycles as a function of wavelength pairs.

-§3.5: the discussion about the influence of the different air masses is not straightforward and to some extend has the same conclusion than the annual cycle: in winter there is higher concentration due to the lower mixing layer high than in summer. P. 27830: a figure could explain explicitly that “the fine mode effective radius seems to be more important than the fine mode volume fraction in explaining the variation of the Angström exponent” ? the same correlation could be done with b.

- p. 27830 last §: the first 2 sentence compare the b as a function of season. The third one gives a reference comparing b and the particle size. Could you please better explain where is the inversion in the relationship? The Mie code could also reproduce all these relationships?

- p. 27830 last §: a figure could present the inverse correlation between b and the effective radius of sub-micron particles?

-Table 2 reports log(b) and not b according to the text p. 27820

- Fig. 4: the annual cycle of N=sum of all SMPS could be also presented?

- Fig. 5 should be given with §2.3

- fig. 6: air masses cannot be easily read.

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