Atmos. Chem. Phys. Discuss., 12, C4258–C4266, 2012
www.atmos-chem-phys-discuss.net/12/C4258/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimating marine aerosol particle volume and number from Maritime Aerosol Network data" *by* A. M. Sayer et al.

Anonymous Referee #1

Received and published: 4 July 2012

1. Summary and recommendation As the authors point out, the issue of satellite retrieval of aerosol volume and number concentration in the PBL is a very important one. Indeed, one can argue that the global coverage of aerosol concentration necessary to significantly improve assessments of changes in the global radiative forcing due to aerosols is predicated on the success of such retrievals – and they are not in hand. This study well illustrates this, as the authors rightly note. Nevertheless, it is quite useful to have a sort of interim progress report on the feasibility of such retrievals and in this sense the current manuscript would constitute a worthwhile publication. However, I find the presentation lacking in several regards. First, the authors must more clearly identify the value of the comparison of MODIS retrievals with the MAN data. These data are certainly not MBL aerosol concentrations and are really based on radiometers looking upwards rather than downwards (as in satellites) coupled with microphysical

models that do not differ too much from the models used in the MODIS look-up tables. The authors conclude from the comparison that the MODIS average solution may be better than the best solution. Is that it? It is worth noting in this regard that such average solutions usually DO give better results in chi-squared assessments since they strongly down-weight outliers. The price paid is, of course, less actual prognostic power for any give case. I think more needs to be done here to clearly define what is being tested by such a comparison and what is not. Second, the comparison of the MAN data with actual measurements of the MBL aerosol concentration is, in my view, flawed, and should be revised. Of course, the authors are careful to point out the difficulties in such a comparison (e.g., lack of the necessary data) but I feel that the in situ data used are not appropriate (more on this in the specific comments) and suggest a better agreement (poor though it is) than is actually the case. Finally, I do not feel that the confounding influences of such non-aerosol parameters as RH have been adequately addressed. Astonishingly, RH impacts on the aerosol have not been discussed at all. Associated with this, I think that the authors could do more to suggest approaches to deal with this admittedly difficult issue. Some of these shortcomings are quite serious. Nevertheless, the aerosol remote retrieval problem is a very difficult one and I think that the study could be made acceptable for publication if the issues I raised above can be successfully addressed. More support for this opinion is given below in my specific comments.

2. Specific comments 2.1 Page 14961, equation 1. The equation for optical depth is O.K. but, given the ensuing discussion, I think that the authors should show everything here. By everything I mean the actual equation which relates the AOD with the aerosol size distribution and thus Cv and Cn. Hence, I would use:

ãĂŰ_(τ_(λ=) $\int$ _⊗∞âŰŠãĂŰ($\int$ _⊗∞âŰŠãĂŰQ(m,λ,r)πr^(2) f(RH)N(r)dr)dzãĂŮãĂŮ)ãĂŮ

or some equivalent from. With this formulation one can more clearly see the problem that one is trying to address with the retrieval. Note that I have added a hygroscopic

growth factor to the equation, a topic that the authors do not really explicitly address at all (more on this later).

2.2 Page 14965 lines 1-27 (also bottom of P. 14967 and first lines of P. 14968) The authors here point out that they are essentially doing a fit with two free parameters, Cv,f and Cv,c. This would be an excellent place to point out, or perhaps reiterate, what the authors themselves have said elsewhere, that one can get a good fit with an incorrect model. My point here is that the authors have also incorporated other information into the model, for example the index of refraction of the two postulated modes (based on AERONET retrievals) and the modal diameters (or radii) of the two modes. The actual values of such variables may well be different from the assumed choices and, if so, the fitted Cv,f and Cv,c values will have partially compensated for this, i.e., they will not be values actually present in the MBL. Certainly the values, for example, of the indices of refraction used in the model are not in agreement with numerous in situ measurements (e.g., Sierau et al, J. Geophys. Res., doi: 10.1029/2006JD007568, 2006; Shiobara et al, Atmos. Environ., 41, 4638-4652, 2007). The authors are certainly aware of this issue (P. 14967 as indicated above) but I think that they must do more than simply mention it; rather they must at least briefly (and preferably quantitatively) assess the impact on their MAN retrievals.

2.3 Page 14968, lines 11-16. I do not see how the chi-squared value for the AOD fit, even assuming that it equals $\sigma\lambda$, MAN, is necessarily a good indicator of the uncertainty in Cv,f and Cv,c. As per Eq. 1 (that is, my equation 1), the relationship between AOD and Cv,f and Cv,c is complex. The uncertainty in various other parameters such as index of refraction and aerosol hydration will all contribute to the uncertainty in AOD. If there are covariances present between the various parameters in Eq 1, or if some uncertainties far exceed those in Cv,f and Cv,c , then the AOD and volume uncertainties may not even be linearly proportional. Let me give just one example. There is likely a negative covariance between f(RH) and m (the complex index of refraction), i.e., as the aerosols grow with RH, m decreases. Such covariances could easily destroy a simple

proportionality between uncertainties in AOD and the aerosol modal volumes. The authors must either more clearly demonstrate that the chi-squared values are reasonable proxies for the uncertainty in Cv,f and Cv,c or excise this claim and make some more reasonable assertion, perhaps with other, more credible, assumptions.

2.4 Page 14974, lines 14-21. It is here where the authors appear to make their assessment of the value of their comparison of MAN and MODIS retrievals (of course these are echoed in the conclusions). They conclude that the average MODIS solution is in better agreement with the MAN data than the “best” solution. This is something, I suppose, but not really a surprise. Unless one has strong variance in the underlying data (and restriction to marine cases severely attenuates this), an average solution will nearly invariably produce the least total residual. However, I think that the key question for any such comparison as that done here – which the authors themselves are clear to note is not a definitive evaluation against the ultimate target parameter – is what do we gain by it? Reiterating my remarks in the recommendation section, the comparison is between integral measurements looking up with fairly similar integral measurements looking down, followed by the use of, essentially, look-up table fits with rather similar parameters to retrieve the target column aerosol values. What is different between the two approaches and what does the agreement (modest, but I agree that it is there) tell us? For example, looking down, the satellite detector must deal with such issues as surface glint that the MAN photometers are not subject to. Agreement suggests that the glint issue has been successfully addressed. What else? This is the question that the author should be going all out to explicitly addressing here and I do not really see it. There are some piecemeal, partial attempts to deal with the issue at various points in the text but no coherent and definitive assessment is offered. It is needed. 2.5 Page 14973, lines 22-25. There is a misstatement here, likely a typo but it is confusing. The authors state that the fine-mode per particle extinction is tow orders of magnitude greater than that for the coarse mode. It is of course the opposite that is true, as can be seen in Table 1.

2.6 Page 14974, line23 to p. 14975, line 12. The authors commence here their comparison of MAN-derived surface aerosol concentrations with actual in situ measurements of aerosol number concentrations in the MBL. The first issue that arises in this attempt is to somehow relate the column concentrations, or burdens, with the surface concentrations. To do this, the authors adopt the assumption of exponential decreases in aerosol concentration with altitude, leading to the simple and well-known relationship between surface concentration, column burden, and scale height. For this purpose, a reasonable assortment of studies from which scale heights can be derived for marine aerosols is given in Table 7. The authors also note that one does not always have exponential aerosol profiles in the marine atmosphere. I would agree; aerosol layers aloft, for example, are quite common in the marine atmosphere (e.g., Clarke and Kapustin, J. Atmos. Sci., 59, 363-382, 2002; Kuzmanoski et al, Aerosol Sci. and Technol., 41, 231-243, 2007). Indeed, they commonly occur near one of the key sites (Lanai) used by the authors for the derivation of their aerosol model (Sharma et al, J. Atmos. Oceanic Technol., 28, 418-425, 2010). So, what does this signify? The authors do not pursue the issue, which is understandable since there is really no way to actually retrieve the surface concentrations without a simplifying assumption such as the exponential distribution. I think the authors should explicitly acknowledge this and also offer some assessment – admittedly it will be rough – of how common the exponential profiles actually are. Furthermore, there are also substantial vertical variations in other key parameters such as composition (and thus index of refraction and hygroscopicity) and, of course, RH. One compositional example would be organic aerosols, largely secondary , that lead to much of the free tropospheric aerosol mass over the oceans being organic (cf. Murphy et al, J. Geophys., 111, doi: 10.1029 /2006 JD007340, 2006; Thomberry et al, J. Geophys. Res., 111, doi: 10.1029/2009JD012721). These issues need to be explicitly acknowledged and at least briefly discussed.

2.7 Page 14975. There is really no specific place to cite the inadequate discussion of RH effects on the aerosol retrieval since the issue is not discussed at all. However, this issue will become most acute when one is trying to compare retrievals with actual MBL

aerosol concentrations so I choose to raise it at this point in the text. The first point to remember is that RH has a marked effect on aerosol size and thus on aerosol volume but only a limited impact on particle number (essentially by moving small particles into a detectable size range). This is so well established that I hardly think I need support it. Nevertheless, let me cite at least one study (see also the numerous citations in this reference) that shows the strong impact of RH on light-scattering, namely, Carrico et al (Tellus, 52B,694-715, 2000), for various aerosols, including marine. Given this, the number-to-volume ratio will be a strong function of RH and, of course, modal radii and, to a somewhat lesser extent, the modal standard deviations of the aerosol size distribution will be impacted. This is simply physics and you cannot get around it. So how do the MODIS and MAN retrievals deal with this issue? For MODIS, it is fairly clear that the issue is simply not really dealt with (Remer et al, 2005; 2008, as cited by the authors). The MODIS LUT simply includes a number of aerosol size distributions associated with water soluble or “wet” aerosols. If these are selected in the fit, then one could conclude either that the aerosols were more hydrated than otherwise or they had differing dry sizes. Clearly there is a mixing of actual differing dry size parameters with varying degrees of hydration. For AOD retrievals one can more or less get away with this. As the authors themselves point out, RH is a function of altitude. Hygroscopicity is also a function of altitude in the marine atmosphere, and elsewhere (cf. Hegg et al, Geophys. Res. Lett., 29, doi: 10.1029/2001GL014495, 2002). And, of course, aerosol sizes change with altitude in the marine atmosphere (Clarke and Kapustin, J. Atmos. Sci., 59, 363-382, 2002). All of these things are mixed together in the AOD as per Eq. 1 (mine) so it is not too surprising, given expected covariances, that one can get at least reasonable AOD’s with no explicit RH dependence – but microphysical aerosol properties recovered from such a LUT are not likely to be accurate. The MAN retrieval, as given in Sayer et al (2012, as cited by the authors), at least discusses the issue somewhat but again the authors do not seem to appreciate how difficult it is to take partial derivatives of compound variables in the real atmosphere. They examine the dependence of AERONET retrieved size distributions on RH as derived from either

NCEP reanalysis (sfc, i.e., 10m) or shipboard data, in other words, surface data. These are clearly not the RH's the column aerosol sees. (Incidentally, perhaps in future work, the authors should consider the use of GCM's or CTM's to get vertically resolved RH data as per, for example, van Donkelaar et al, Environ. Health Perspect 118, 847-855, 2010.) Furthermore, the size distributions with which the RH comparison is made are AERONET retrievals and, as with MODIS, a number of variables that plausibly vary with altitude – and RH – are incorporated into the retrieved AOD's. Because the retrieved size distributions inferred from the AOD's show little dependence on RH in the 60-80% range characteristic of the lower MBL, the authors essentially neglect any RH dependence. Once again, I think that you can get away with this lack of dependence, or at least weak dependence, for AOD's but definitely not for the surface size distributions and thus Cv and Cc values derived from them – and certainly not for MBL aerosol number concentrations derived from them. I feel that a much more extensive discussion of the issue is necessary. The authors can perhaps make a case for their approach based on current necessity and the extensive averaging that they do, but it must be made plain (in my view) that this is a very problematic approach for estimating MBL aerosol number and volume concentrations.

2.8 Pages 14976-14977 (Discussion of Figure 10) The last, and most challenging, analysis offered by the authors is the comparison of the MAN derived MBL aerosol concentrations with direct measurements in the MBL. The authors derive actual aerosol concentrations in the MBL from various sources to compare with zonally averaged Man retrievals. There is a great deal of spatial and temporal averaging involved in this comparison but I do not find it unreasonable for a rough estimate of bias, though perhaps this could be more clearly stated. My main concern here, in addition to the RH issue just discussed in the previous comment (which will be most acute here), is that it is not clear to me that the authors are comparing the right quantities. First, it must be remembered that MODIS – and MAN - are retrieving first the fine mode scattering signal and, based on this, a volume fraction that is associated, essentially, with a given number-to-volume ratio. Hence the interest of the authors in pointing out that these ratios (as per

Table 1) are similar to those observed from in situ measurements (p. 14970, lines 7-11). However, the fine mode scattering is dominated by the accumulation mode, down only to particle diameters of perhaps 100 nm, or even larger. The smaller particles in the lower Aitken mode or nucleation mode scarcely impact the scattering signal at all in the marine atmosphere. In accord with this, the number –to-volume ratio used in the MAN model – and the observed ratios with which it is (sort-of) validated – are for the accumulation mode. Given this, when particle concentrations are derived from the MAN retrieval, they should be compared to in situ measurements of the accumulation mode. This does not appear to be the case in Figure 10. The data from the GAW network are CN data. Similarly, the data from both the Heintzenberg et al study and the Bates et al study are essentially CN concentrations (sum of all DMA channels and thus down to the detection limit of the actual detectors – which are CN counters). These counters (typically TSI 310's or 3020's) measure below 20 nm. In a way, the authors are making things difficult for themselves in trying to recover concentrations of particles that their instrument does not "see." The more immediate issue, however, is that they are looking at much higher concentrations from the in situ measurements than they should be. Both the Bates et al study and that of Heintzenberg et al report accumulation mode concentrations in addition to total number concentration and I think that the authors should do a comparison with these concentrations. In principle, I would expect to see higher bias but perhaps better correlation between the in situ and MAN values. In any case, the extant comparison is not, in my view, valid. Another issue worth pointing out, though it is not so critical since the authors did not choose to do the relevant comparison, at least explicitly, is that I would not expect retrieved Cv,f values to agree with those from the in situ measurements. Such a comparison might have been made, for example, to try to isolate where discrepancies are arising in the retrieval. If they had been made, it would be well to remember that both the measurements reported by Heintzenberg et al and those by Bates et al are at reduced RH ($\sim$40% or less and 55% or less respectively) whereas the retrievals are implicitly for RH's in the 60-80% range. The authors' own analysis suggests there will be decided differences in the volumes

between such disparate RH's.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 14959, 2012.