

Interactive comment on “Aerosol mass closure and reconstruction of the light scattering coefficient over the Eastern Mediterranean Sea during the MINOS campaign” by J. Sciare et al.

J. Sciare et al.

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First of all, the authors want to gratefully thank the reviewer 1 for his/her deep and very detailed review of the manuscript. Following his/her recommendations, corrections have been done in the revised version of the paper, significantly improving its scientific quality and conclusions.

I. General comments of referee 1

+ Major comment 1: “the English of the paper could be improved in many instances” Corrections have been done following the recommendations of the referee. Note also that this manuscript has been carefully checked by all the authors (H. Cachier, W.

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Maenhaut, M.O. Andreae) as well as by the editor. None of them have found any particular weakness in the English level of the paper.

+ Major comment 2: “There are some sections that are not really necessary and could be shortened or removed” Corrections have been done in the revised version following the recommendations of the referee.

+ Major Comment 3: “There should be a discussion on the implications of the fact that a simple method (...) can lead to proper reconstruction of the scattering properties” : The method proposed here to reconstruct the scattering coefficient is making many assumptions which are only valid for the time and the location of the campaign. For instance, our filter-based measurements will not determine in a quantitative way any of the semi-volatile species (ammonium sulphate, organics). As a matter of fact, and for locations with high levels of semi-volatile material, we will not be able to achieve the reconstruction of the scattering coefficient from our filter-based chemical analysis. Then, the statement done by the referee (“Measuring sulphate and OC is enough, and the hydrophilic nature of OC is not important”) can only true for the time and place of our campaign. This is the reason why we did not want to emphasize (as much as the referee would like) the potential role of our methodology in future studies dealing with the quantitative contribution of chemical species in the aerosol scattering properties.

II. More general comments of referee 1

+ Page 2432, line 8: Cut-off performed by the mean of an impaction plate. Information added in the revised manuscript.

+ Page 2433, line 7: Uncertainties are given for all the chemical species. Blank values have been added in the manuscript for BC and OC measurements.

+ Page 2434, line 22: the meaning of a correlation including all chemical species is that all these chemical species are summed in the chemical mass balance.

+ Page 2435: The purpose of an ionic balance is found in the title of the section; e.g.

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“Quality assurance”. Belongs to their own experience, the authors have noticed that an ionic balance which is not achieved rarely leads to good chemical mass closure. The results presented here are consistent. Then it cannot be argued later in the text that discrepancies in the mass closure are originating from errors done in the IC analysis.

+ Page 2435: The argument that justifies the choice of the EC obtained with the thermo-optical method for use in the mass closure is clearly written in the text (page 2435). Hence it is shown later in the text that the choice of EC (2-step thermal or thermo-optical) does not impact on the results of the mass closure. To get an answer to the question “what is changed if the other series is used”, the referee should carefully read the page 2441, section “Influence of the carbon analysis protocol”. All the details are given in this section.

+ Page 2437: “I do not understand equations 2 and 3: what are the percentages?”. This was better explained in the revised version.

+ “Also what is the reason to present equation 3 since you do not use it in the following?”. Conversely, the authors wonder why they should not present this equation since their discussion is based on [Fe]/[Al] ratio. (the ratio between equation 3 and 2 is used in this discussion; reference Guieu et al., 2002).

+ “if nss-K is mainly from biomass burning, there should be some associated Cl”. We do agree with the referee that nss-K should be associated with Cl. This is true for young smoke whereas more K₂SO₄ and KNO₃ particles should be present in aged smoke (see for instance Li et al., J. Geophys. Res., 2003). This is confirmed in our study by the levels of nss-K and Cl recorded in the fine mode during the campaign ([nss-K] = 11.1 nmol/m³, [Cl]=0.6nmol/m³).

+ “you should gather sections 4.3 and 4.5”. This has been done in the revised version

+ “sections 4.4 and 4.6 do not bring in much information. They could be combined and shortened”. Although the section 4.4 does not bring much information, it intro-

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duces many terms (ammonium sulphate, potassium) and cannot be shortened. The last section (4.6) emphasized the need to separate the fine and coarse modes. This is of particular importance and referee2 mentioned it as one of the most important result of the manuscript. For this reason, we decided to keep this section as it is.

+ “Section 4.6: ...”. We do not agree with the referee. Normalized concentrations as it could be reported in figure 2 will bring less information than atmospheric concentrations expressed in $\mu\text{g}/\text{m}^3$

+ Figure 2: “there are 2 categories in very close blue colours”. It is true (if your printer is not a good one). These colours are consistent with those reported in Figure 5.

+ “Table1: ...”. Ok. These errors appeared when ACP converted the table 1 in 2 parts. Also we let the digit as they are since they have all the same format in the table.

+ “Page 2440, line 19: you should present the equations of the regressions between the measured and reconstructed masses ...”. These equations are already reported in Figures 3a and 3b and are discussed in the text. The authors don’t think that these equations need to be put again in the text.

+ “section 5.1: you need to give the complete equations of the regressions”. Since these equations are of the type “ $y=ax$ ” and since the slope (a) is given in the text, there is no need to give more details about these equations.

+ “It is correction affecting more the samples that were previously out of the 10 percent limit”. The authors do not understand very well this question. Correction from bound water is done on all samples. All the samples are affected by this correction

+ “Section 5.2: there is a need for a figure or table comparing the results using the 2 series of OC measurements”. This would be interesting if the results using these 2 series were significantly different, which is not the case. Although there is a serious difference between the BC data from the two methods (on average 33 ± 22 percent), the difference between OC measurements is much lower (on average 11 ± 8 percent).

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Assuming that POM represents 1/3 of the reconstructed mass in the fine mode, this 11 + Page 2442, line 7-11: “how do you reconcile the fact that bound water can have some influence and that a CF of 2.1 is the best one to fit the data (without taking into account the influence of bounded water)?”. The authors have decided to report their chemical mass balance without taking into account bounded water influence, as it is the case for all the chemical balance which are reported in the literature. The conversion factor of 2.1 was also taken from the literature. It is interesting to note here that the missing mass (Figure 4) is calculated with a constant CF and without taking into account bound water influence. As shown in this Figure 4, the missing mass is not connected with a change in inorganic salts (which will increase/decrease bound water influence) but is connected with a change in the BC/TC ratio. This result would suggest that the choice of CF is more important than bound water influence in our chemical mass balance. This sentence has been added in the revised version of the manuscript.

+ Page 2442, line 23-25: “it should be possible to calculate the most appropriate CF deduced from these values” A new paragraph has been dedicated to the CF deduced from the missing mass.

+ Page 2443: “the second paragraph of section 5.3 is rather descriptive and is leading to strong conclusions”. We absolutely agree with the referee. It is leading to strong conclusions.

+ Figure 5: “there is no caption to tell what is the line with circles”. Changes have been made in the revised manuscript.

+ Section 5.4: “all this section is not bringing useful information. The only interesting point (...) is not discussed in depth”. First, we do believe that this section should remain as it is since it brings to our attention that Factor analysis and source identification (as widely used in literature) can lead to erroneous conclusions. We do agree with the referee that the point dealing with carbonaceous aerosols is rather vague (it is also one

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of the comments of referee2). For this reason, we have brought some changes on this point in the revised version.

+ Page 2446, section 6.1: “What is the impact of the changing RH during the course of each 12h period of sampling ? Since the relation is not linear between RH and scattering, what is the bias introduced by using 12h averaged RH ? Could you show the range of variation of RH ?”. We absolutely do agree with the referee that 12h averaged RH will produce some bias in the calculation of the scattering coefficient (equation 8 of the paper). A short paragraph has been added in the revised version with sensitivity tests which have shown that errors done in 12h averaged RH are not significant and do not control the errors done between measured and reconstructed scattering.

+ End of section 6.1 and Figure 6: “you should have a short discussion on the episodes with significant deviation between measured and reconstructed scattering coefficients (around 10/8 and 20/8)”. These deviations do not correspond to any particular changes in chemical composition, weather conditions, or air masses origin. However, it is interesting to note that when 2 different CF are applied (respectively 2.7 and 1.8, before and after 12 August as previously calculated), mass scattering efficiencies are slightly changed (2.53 and 4.19m²/g for ammonium sulphate and POM respectively). On the other hand, the correlation coefficient is significantly improved (from 0.83 to 0.90). That is to say that the reconstruction of the scattering efficiency can bring further constraints on our chemical mass closure (and OC to POM conversion factor). A new paragraph has been added in the revised manuscript.

+ Section 6.1 (and conclusion): “there should be a discussion on the implication of this work ...”. See our answer of the major comment no3

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