**Atmospheric
Chemistry
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Discussions**

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Interactive comment on "**Long-term measurements of carbonaceous aerosols in the eastern Mediterranean: evidence of long-range transport of biomass burning**" *by* **J. Sciare et al.**

J. Sciare et al.

Received and published: 2 August 2008

Anonymous Referee #2 Received and published: 28 May 2008

We would like to thank the reviewer 2 for his/her comments that have helped us to prepare this final version. Most of the suggestions have been taken into account and all raised issues are answered one by one. Minor comments have been also taken into account. Below is a point by point answer to the reviewer 2 comments (by Italics).

Specific comments: 1. Page 6952, starting with line 24: It should be indicated how many trajectories per day (or per week) were calculated for arriving as Fig. 1.

One back trajectory was calculated every 12h and this information was added in the

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2. Page 6953, Aerosol sampling: Although it may be indicated in the references given, it is strongly suggested that the authors specify here at which temperature and especially at which relative humidity (RH) the Nuclepore filters were weighed. The aerosol contains a number of hygroscopic components, which may pick up water, particularly at RHs above 50%.

Most of the information required by the reviewer was already reported in the MS (section 3.5 Gravimetric analysis). The Nuclepore filters from the SFU samples were weighed at LSCE after 24 h equilibration at room temperature and RH below 10%. Information regarding the role of water uptake onto filters is added in the revised MS to explain the reason why analyses have been performed at low RH.

3. Page 6953, Aerosol sampling: A common problem with stacked filter unit (SFU) sampling on Nuclepore filters is filter clogging, particularly for the fine filter. Considering the levels of the fine (PM1.5) particulate mass (PM), it seems impossible to me to collect samples of 1 week at a flow rate of 1.5 m3/h without avoiding this problem. As a consequence of the clogging, the flow rate will decrease, the sample will not be representative anymore for the entire week, and the cut point between fine and coarse will increase. The increase in cut point may not be a major problem if most of the total PM is in the fine size fraction. I suggest that the authors provide a typical percentage for the ratio of fine to total PM.

The reviewer is absolutely right in his statement. For that reason, we have applied a sequential sampling (15 min sampling every 1 hour) to minimize the risk of filter clogging for SFUs (information reported in the manuscript). As mentioned by the reviewer, filter clogging would result in a rapid decrease of flow rate which was not observed in our samples (flowrate was checked at the beginning and at the end of each sampling). From one SFU sample to another, the difference in flowrate was on average 2.3+-2.5% indicating no major variability due to filter clogging. A possible clogging would also

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artificially change the composition of the 8μm pore size filters as this filter would collect chemical species that are usually found in the submicron mode (ammonium sulphate for instance, or BC that should darken the 8μm pore size filter). This was carefully scrutinized and did not reveal unusual chemical composition in these filters. The typical percentage for the ratio of fine to total PM is 0.4, which is in very good agreement with studies at the same location using SDI impactors (Koulouri et al., 2008, Atmos. Environ in press and Gerasopoulos et al., ACP, 2007)

4. Page 6954, Light absorption measurements: First, it should be indicated at which wavelength the aethalometer measurements were done. Secondly, the mass absorption efficiency (sometimes also called specific absorption coefficient) that was used to convert the babs data into BC data should be specified.

The following changes have been performed in the manuscript to answer the reviewers request. ...This Aethalometer is working with an incandescent light with a broad spectral distribution. The spectral response of a similar instrument (AE-10) is given by Weingartner et al. (2003). The aerosol absorption coefficient (bATN) determined in this way may differ significantly from the true aerosol absorption coefficient (babs) of airborne particles (Weingartner et al., 2003); Following the recommendation of the constructor a sigmaATN value of $19 m^2/g$ was used in this work. Weingartner et al. (2003) have proposed an R-value close to 1 for aged atmospheric aerosols at remote site, as the high amount of scattering aerosol material in the fibber matrix compensating the shadowing effect which is related to R. The same authors have also proposed a C value of 1.9 corresponding to aged atmospheric particles. Following these results, our absorption coefficients were calculated with R and C values of 1 and 1.9, respectively. Aethalometer BC measurements are reported in the following as BC(AETHALO);

5. Page 6956, Chemical Mass Closure: The authors state here that Ca was used to obtain mineral dust. I presume that nss-Ca was used for this. Furthermore, the occasional reader may wonder how ss-sulfate, ss-K, and ss-Ca were obtained. Actually, they were estimated from aerosol Na and from the sulphate/Na, K/Na, and Ca/Na ra-

tios in bulk seawater so that sea salt was actually calculated as 1.326 [Na] + [Cl] + [Mg]. I suggest that this is clarified, for example by providing this formula.

As mentioned in the MS; All the hypotheses using here to perform the CMC were taken from the mass closure study reported in Sciare et al. (2005) for the MINOS campaign and are briefly reported here;. Following reviewer suggestion we have additionally reported in the MS the information related to the calculation of sea salt atmospheric concentration.

6. Page 6957, from line 14 on, and Table 1: Based on the data given in that Table, thereby using an OC-to-OM conversion factor of 1.8, I arrive at other numbers for PM-CMC than those given in Table 1. For example for the month of January, I obtain 8.22 instead of 9.66 and for the Average, I arrive at 9.37 instead of 8.88.

As mentioned in the MS; Monthly mean concentrations of the major aerosol species, PM and PMCMC are given for the fine mode in Table 1;. This means that PMCMC reported in Table 1 (like PM and other species) is monthly mean of PMCMC calculated for each individual filter sample. In other words, PMCMC values reported in this table are not obtained from the sum of the monthly means of each chemical species. That is the reason why the reviewer could not recalculate by himself PMCMC. That is also the reason why PMCMC is presented with standard deviation in the table. We have made this clearer in the manuscript. Furthermore, on line 4 of page 6958, a positive number is given for the difference between PM and PMCMC. This is contrast with the data given in Table 1. Incidentally, the positive number also does not agree with the difference between PM and the PMCMC that I obtain. As explained in the previous comment, the value reported as the difference between PM and PMCMC stands for the mean standard deviation calculated from the monthly mean PM and PMCMC values reported in Table 1. We have made this clearer in the manuscript.

7. Page 6960, lines 18-20: It is far from sure that "this" fraction should be classified as "real" BC. The increase in light transmission could also be due to desorption of pyrolitic

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black carbon, i.e., to desorption of OC that was converted into BC by charring. Actually, it does not matter that the light transmission already increases during the part of the analysis in a pure He atmosphere. The distinction between "real" BC and "real" OC in the thermo-optical analysis is based on the setting of the OC/EC split point. It does not matter whether this split point already occurs at the end of the phase in pure He, as long as all "real" OC has been desorbed before the split point.

We have removed the sentence; The increase in the light transmission during this temperature step and observed in almost all our samples indicates that this fraction should be classified as BC rather than OC;. This sentence is obviously incorrect and we would like to thank the reviewer for notify us. We also agree with the referee that....It does not matter whether this split point already occurs at the end of the phase in pure He, as long as all "real" OC has been desorbed before the split point. On the other hand, when we have both OC and EC evolving at the last He plateau, how can we be sure that OC has been evolved well before the split point? Actually it is impossible to answer to this question. The best way is probably to avoid this release of O2 at the last He plateau, and thus to better choose a last He plateau that does not induce release of O2 from metal oxides (like the IMPROVE protocol). See also answers given to the first reviewer on this matter.

8. Page 6961, lines 5-6: I am confused by the fact that the authors state here that BC in the 2-STEP method is more sensitive to fossil fuel combustion aerosols than to biomass burning aerosols. It is possible that I do not catch what the authors want to say, but I thought that the percentage difference between BC data obtained by the 2-STEP method and those obtained by IMPROVE for fossil fuel combustion aerosols is smaller than the percentage difference between the two methods for biomass burning aerosols. When comparing the T2S (i.e., 2-STEP) and TOR (i.e., IMPROVE) data for the urban aerosols samples (which likely contained mostly fossil fuel combustion BC) in Figures 2 and 3 of the carbon shoot-out paper of Schmid et al. (2001) I see little difference [H. Schmid et al., Atmos. Environ. 35 (2001) 2111-2121].

Over the past 5 years we had the opportunity to compare in many locations T2S and TOR methods. In accordance with Schmid et al. (2001), we had very good agreement (and little difference) when comparing the two methods at urban sites with fresh fossil fuel carbonaceous aerosols. At remote sites (e.g. aged fossil fuel + possibly biomass burning) we have seen sometimes significant differences between these 2 methods. From our point of view, this is not really surprising since T2S method is mainly based on thermal properties of EC (which might evolve with ageing), whereas the TOR method relies both on thermal AND optical properties of EC (which both optical and thermal properties might also evolve differently with ageing). Our statement in this work is mainly based on the results reported at the same location by Sciare et al. (2003) during the MINOS campaign. In this paper, the authors have shown that the TOR method correlated with nss-K (tracer of biomass burning) whereas T2S was not following the variations of nss-K but it was better correlated with nss-SO4 (tracer of fossil fuel). Different thermal and optical properties of biomass burning and aged fossil fuel EC may clearly explain these two features. We are aware that what has been found in Crete may not be valid in other locations with a mix of fossil fuel and biomass burning.

9. Page 6961, lines 8-12: Some motivation should be given why weighed averages were calculated and why equation (4) was used for this. I understand that the data of all years were used for obtaining these weighed averages. This should be indicated and it is suggested that the periods used for obtaining them are indicated, as these periods are not the same for all species.

This is correct. The reason why we have decided to smooth the atmospheric signals is to better observe seasonal tendencies. A more detailed explanation can be found in the answer given to the first reviewer and in the MS.

10. Page 6962, line 17: Acronyms and abbreviations should be defined (written full-out) when first used. Here for AOD.

Definition was given for AOD.

11. Page 6964, line 8: I read from Fig. 3 a nss-K concentration of around 55 ng/m3 for May and June instead of the 50 ng/m3 given here in the text.

Correction was done in the MS by reporting that 50 ng/m3 is a round average using May and June months

12. Page 6964, from line 12 on: Apparently, the BC* and OC* data are weighed monthly averages. It is unclear, though, whether bb-BC, BC(IMPROVE), bb-OC, and OC(IMPROVE) are also weighed monthly averages.

Information was given in the MS

13. Page 6966, from line 1 on: The authors should not forget that OC and BC data are method-dependent and that one should be careful when comparing OC/BC ratios of various authors. A proper comparison is only possible when the data were obtained with the same method.

The reviewer is absolutely right and explanation was added in the revised MS.

14. Page 6967, l. 15-17: The explanation that the authors give here is unclear for me. It seems to be in contradiction with what they wrote earlier in this paragraph.

The reviewer is absolutely right. A word...not..is missing were. The corresponding sentence has also been re-written in a less definite way: In other words, if this OC* increase is due to SOA formation, it has then the implication that this SOA comprise almost equally water insoluble and water soluble organic material, which is in contra-diction with the previous studies suggesting that SOA is mainly composed of WSOC;

15. Cases where wording and/or grammar should be improved and technical correc-tions: All the suggested corrections have been taken into consideration.

p. 6950, l. 6: replace "have shown" by "were shown". p. 6950, l. 11: replace "has shown" by "was shown". p. 6950, l. 12: there is something missing here; perhaps

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"expected a high" can be replaced by "expected to exhibit a high". p. 6951, l. 17: replace "light properties" by "optical properties". p. 6951, l. 24: replace "IMROVE" by "IMPROVE". p. 6952, l. 24: replace "interferes in" by "interfere in". p. 6953, l. 17: replace "made of" by "using". p. 6956, l. 2, and also on other occasions later in the paper (e.g., p. 6959, l. 11): SFUs stands for Stacked Filter Units, thus for the samplers. One does not analyse SFUs, but instead SFU samples or SFU filters. p. 6956, l. 22: replace "using" by "used". p. 6957, l. 6: replace "were found" by "are present". p. 6957, l. 25: replace "of the time from the condensation" by "of the time condensation". p. 6959, l. 21: replace "air masses origin" by "air mass origin". p. 6960, l. 1: replace "protocol" by "protocols". p. 6960, l. 7: replace "were also" by "are also". p. 6960, l. 10: I would not call a period of more than 3 years short. p. 6963, l. 22-23: There is something wrong with this sentence. p. 6965, l. 22-25: There is something wrong with this sentence. p. 6966, l. 18: There is something wrong with this line and with the entire sentence. p. 6968, l. 27: replace "Although, SO2" by "Although SO2". p. 6973, l. 30-31: There is no reference to Zerefos et al. (2000) within the text, Tables or Figures.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 6949, 2008.

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