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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol sources and their contribution to the chemical composition of aerosols in the Eastern Mediterranean Sea during summertime" *by* J. Sciare et al.

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Interactive comments on "Aerosol sources and their contribution to the chemical composition of aerosols in the Eastern Mediterranean Sea during summer time" by J. Sciare et al.

Reply of the authors to the comments given by the anonymous referee #2.

First of all we wish to thank the reviewer for the attention that he/she brought to our work. Below are the answers to his/her comments.

The aim of this paper is NOT to attempt to achieve reconciliation of our measurements with EMEP inventories as it is claimed by the anonymous referee (line 3, page S449). The aim of the paper is to identify, by the use of various approaches, the contribution

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of different sources to the levels and chemical composition of aerosols and relevant gases in the Eastern Mediterranean during summer. Among these methods a source-oriented model was applied to better localize the sources of aerosols on Crete Island. Despite the limitation in time of our campaign, it is quite interesting to compare the output results of this model with data available in the literature (EMEP inventories) focusing on the location of the potential sources. Although this comparison is by no mean quantitative and definitely not the main focus of the paper, the authors do believe that the agreement between our model results and the EMEP inventories should be mentioned somehow in this publication. We believe for instance that our results on BC, BC/nss-SO4, comparison of dust aerosols with AOT, have also at least the same importance with, if not higher than, the comparison with the EMEP sources.

Major Comments:

Below is the reply to the comments given by the referee following the same annotation:

1: The authors believe that providing quality-controlled fine resolution measurements of SO2 and nss-SO4 should be a positive rather than a weak point of the paper as mentioned by the referee.

2: We do not agree with the referee that our sampling period of 2 hours is "shorter by two orders of magnitude with the characteristic time scale of major changes in meteorological conditions". For receptor sites far from the sources, the typical duration of "continental events" is short in time and normally does not exceed few hours. See for instance the numerous studies performed at Amsterdam Island in the southern Indian Ocean, a receptor site located 3500 km far from Africa (Sciare et al., 2001; Gros et al., 1998, and references therein). In addition, the variability observed in the Rn222 activities during the campaign is indicative of important short term changes in transport conditions: Some changes of one order of magnitude (from 100 to 10 pCi/m3) could occur within 4 hours. (Figures are not possible on the ACP interactive website but are available upon request).

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3: For the source-receptor model, the number of data points is a critical parameter (refer to Charron et al. 2000, for more information on the equations used in this model). Each data point is associated with an air mass back trajectory. More data points you use in the source-receptor model more accurate will be the definition of the sources, especially far from the receptor site. Daily or diurnal sampling (as suggested by the referee) would simply be not sufficient for use as input parameter in this source-receptor model.

4: We do not believe that analytical errors are higher due to the short sampling period. For instance for SO2 analysis: Based on the 2-h sampling (cofer-mist), the average SO2 concentration analysed by IC was 350 ppb. As mentioned in page 6 (line 18), blanks collected during the campaign were below the detection limit (5 ppb). Thus, blanks would represent at maximum 1.4% of the SO2 concentration. The only analytical error related to a short time sampling is the blank correction, which lies within the 8.5% error derived by comparing the two SO2 techniques described in page 6, line 12. Similar errors are obtained for sulphate or sodium (used to calculate nss-SO4) in aerosol phase. Note also, that sea salt sulfate are in very low concentrations most of the time during the campaign (page 7, line 4). Thus the errors associated with the calculation of the nss-SO4 levels are also very low and not influenced by the short sampling time since such bias are independent on the sampling time (2h, 12h, 1 day, etc)

Few sentences will be added in the revised version of the manuscript on the errors associated to measurements.

2) As mentioned above, comparison with EMEP inventories is of course not quantitative (how quantitative comparison would be achieve between concentrations of compounds in Crete for a month period and S emissions in kT/year ??). The only aim of this comparison is to check if the location of our sources correspond or not to the location of the main sources given by EMEP. The authors acknowledge that such explanation should be added in the manuscript in order to prevent remarks on the use of the yearly aver-

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aged emissions from EMEP inventory. The authors do believe that the good agreement between their model results and the EMEP inventories should be mentioned somehow in this publication. Here again, the authors do believe that this comparison is NOT one of the main foci of the paper.

Specific comments:

1) The correlation coefficient between SO2 and nss-SO4 brings further evidence on different source locations (for instance); source locations which are within the main focus of the manuscript. We agree with the referee that the sentence (lines 7-9, page 9) would suggest that an agreement is expected between SO2 and nss-SO4. The sentence will be rephrased.

2) We agree with the referee that error bars should be given on the measurements (probably within the paragraph 2.3 "Sampling and analysis" and also in page 9).

To give to the referee a better idea on the observed diurnal variation of the ratio nss-SO4 / (SO2 + nss-SO4), we have reported in the table below the diurnal variation of this ratio. Normalised ratios were obtained by dividing the given ratio to the daily averaged ratio. Error bars (standard deviation) have been reported in the table and show that the diurnal variation is NOT within these error bars. (Days July 13th and July 15-17th have been removed following the criteria given in page 9, line 16).

Hour(LT) Ratio SD

00:00 0.99 0.13

06:00 0.92 0.12

12:00 1.04 0.12

18:00 1.05 0.13

Note that such diurnal tendency could not be obtained if the sampling period was longer.

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3) The only reason for not reporting the NH4+ levels is to lighten this manuscript since they don't bring more information than nss-SO4 on the origin of aerosols in Crete Island. However, if needed, we can add few words on their levels and variability, especially on the change of the ratio NH4/nss-SO4 as a function of air mass origin.

4) We agree with the referee that sea-salt aerosols would affect significantly the AOT especially in marine environment with important wind speed such as Crete Island during summer. However, one must keep in mind the very high nss-Ca levels recorded during the campaign (figure 2) highlighting the major role of dust aerosols.

A calculation can be done to estimate the mass loading of sea-salt and dust aerosols during the campaign, assuming the following:

Sea-salt = Na + Cl + ss-SO4 + ss-Ca + Mg + ss-K

Nss-Ca = Dust x 0.1259 (1)

(1): Equation obtained from PIXE analysis of Al, Fe, Ca at Finokalia and based on Al/dust = 7.7% and Fe/dust = 5% ; Wedepolh, 1995). Note that these ratios were validated during the MINOS campaign at Finokalia in summer 2001 (Sciare et al., posters presented for IGAC Crete, 2002).

On average, dust aerosols are in mass concentration (μg/m3) 4 times higher compared to sea salt during the ELCID campaign. Considering that measurements were performed at sea level (ie where the sea salt concentration is the higher) and that dust loading is higher in altitude (see below), this factor of 4 can be considered as a lower limit. Note also that during the "dust events" associated with important AOT (figure 6 in the Manuscript), the ratio dust/sea salt at sea level reached values of the order of 10. Kouvarakis et al. (2002) as well as the results from the MINOS experiment during summer 2001 (Sciare et al., posters presented for IGAC Crete, 2002), show that even higher dust loadings / lower sea salt aerosols are observed in the lower Free troposphere compared to the Marine Boundary Layer (MBL). Thus, it is legitimate to

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assume that the column integrated concentration of dust is at least an order of magnitude higher compared to sea salt aerosols during this campaign in summer 2000.

To make it more clear, few sentences will be added to the manuscript to point out the dominant role of dust compared to sea-salt aerosols.

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