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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.

Anonymous Referee #2

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General comments: Acknowledging the vast scientific effort that has been put into this manuscript, the referee has major objections to the general focus of the paper. The authors attempt to reconcile the concentrations of a few selected aerosol and gaseous species that they have measured in a campaign with the available source inventories for Europe. My specific concern here is the differences in time scales.

1) The authors produce large number of data within a relatively short period of their campaign (4 weeks in summer), at a location quite far from the continental Europe. Rather unusually, they use high time resolution (2 h) even for the sampling of aerosol species (e.g. sulfate), thus producing a large body of analytical data. But is such a sampling frequency meaningful? In my opinion, it is not, since the sampling time

is shorter by nearly two orders of magnitude than the mean atmospheric residence time of the main aerosol species and/or the characteristic time scale of major changes in meteorological conditions. Daily or diurnal sampling would have been just as appropriate. Therefore the large number of individual concentration data are by far not independent, consequently they should not serve as a basis for statistical treatment in the source-receptor model. In addition, short sampling times also imply that the concentrations are possibly loaded with higher analytical errors.

2) On the other hand, available source inventories refer to yearly averages, usually with a time lag of several years, but certainly not for a given period of the year. It is also well known that there are large variations in emissions during the course of a year (residential heating, transport, temporal shutdown of industrial facilities, etc.). Therefore a four-week period in summer is not expected to reflect annual averages in emissions of any species. Furthermore, here is another order-of-magnitude difference in the time scale.

Specific comments:

1) It seems that the authors expect correlation between gaseous SO2 and particulate nss-SO4, which should not exist due to differences in sources and sinks of these species as well as the nature and complexity of the S(IV) - S(VI) oxidation mechanism.

2) The referee is not particularly convinced about the clear diurnal ratio of the nss-SO4/(nss- SO4+SO2) mole ratio (from 0.45 to 0.5), since the nss-SO4 is a derived quantity (estimated amount sea-salt sulfate needs to be substracted), loaded with possible analytical errors and uncertainties, so this variation may be well within the error bars, though these are not given in the manuscript.

3) Unfortunately, the authors do not report the concentrations of particulate NH4+, which would have helped resolve the issue of photochemical nss-SO4 formation.

4) When trying to reconcile surface data with satellite observations, the authors cal-

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culate with the mineral dust concentrations, but they do not take into account sea salt aerosol which is probably more significant at the sampling site.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 1287, 2002.