

Interactive comment on “Variability in regional background aerosols within the Mediterranean” by X. Querol et al.

Anonymous Referee #3

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This paper discusses aerosol properties across the Mediterranean basin from several multi-year records, namely their concentration, size ranges (PM₁, PM_{2.5}, PM₁₀), chemical composition, variability in time (daily and seasonal), and relationship with meteorological situations. The topic is very much within the scope of ACP. The paper is very interesting and well-written, and I learned a quite a bit by reading it. I was thankful to have the chance to review paper that compares data across multiple stations over a larger region, as opposed to the more common single-site papers. Also the Mediterranean tends to receive less attention in the scientific literature than Central and Northern Europe, so this focus is very welcome. I highly recommend the paper for publication in ACP after a few minor issues are addressed.

(I will use the notation 10154-15 for page 10154 line 15 etc.)

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Main issues

* 10159-22-24, here CO₂₃- was divided by 2 to account for possible coarse calcium nitrate and sulfate. This appears unnecessarily inaccurate. Why not do an ion balance on the Calcium? E.g. if 100 nmol/m³ of coarse Ca are measured along with 20 nmols/m³ of coarse NO₃ and 10 nmols/m³ of coarse SO₄, then the CO₃ necessary for ion balance is 85 nmols/m³. (Rather than 50 nmols/m³ as the authors's rule would estimate)

* 10159-25, I assume that the non-measured mass is mostly water, but this should be stated explicitly here for clarity

* 10161-16, why were trajectories not initiated at lower altitudes for the FKL (230 m amsl) and ERL (22 m asl) sites?

* 10162-14, can this trend only be explained by the gradual deposition of dust? It seems that just lower frequency of air transport arriving from the dust sources may be as or more important as a factor in creating the observed concentration gradient. Also dilution of dust-laden air with regional air can also create a gradient, without the need to invoke deposition. Probably all factors play a role and I suggest listing them all here.

* 10164-17, here photochemical activity is invoked to explain a peak at 4-6 am GMT. Has the photochemistry started at that time at this site? Perhaps transport may have more to do with this peak?

* 10168-3, the wording 'with a minor relevance' seems too strong here, as the acidity is likely an important factor on preventing the formation of NH₄NO₃, which makes the HNO₃ stay in the gas-phase longer and react predominantly with dust. Of course dust has to be present for this to happen, but in the absence of a modeling study to evaluate the importance of the submicron aerosol acidity in the formation of coarse nitrates, I suggest describing this in a more neutral language.

* 10168-11, 10171-28, 10172-5, 10175-20, these statements about the sources of

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SOA are too strong since no references are provided on studies of the relative importance of the different SOA sources for this region, and since this is a very controversial topic at present (see e.g. Hallquist et al., ACPD, 2009, <http://www.atmos-chem-phys-discuss.net/9/3555/2009/acpd-9-3555-2009.html>). I suggest that the authors list the possible sources of SOA and suggest that they may all play a role: biogenic VOCs, anthropogenic VOCs, and VOCs emitted by biomass burning. Note that the formation rates for all of them would be enhanced by the more intense summer photochemistry. Also the emission rates of anthropogenic precursors (e.g. gasoline evaporation) increase greatly with high temperatures due to the exponential increase of vapor pressure with temperature, so higher summer emissions are not limited to the biogenic precursors.

* Fig 4 for FKL shows significant coarse OM+EC which is not present at MSY, but this is not discussed in the paper. High concentrations of these species in the coarse mode are unusual in my experience, so the possible cause for their presence should be addressed.

* For the material in tables 1-4, in my opinion it would be much easier to communicate it with the readers if it was presented in graphical form. The authors are clearly skilled in creating complex figures such as Figure 4, so I strongly suggest that they invest the time to express most of this information in the form of graphics. Most readers are visual and tabular information is more often overlooked. The tables should then be moved to a Supp. Info. section, which is published together with the paper and is then available for readers who want to know the actual numbers (which would be a very small minority of the readers in my experience)

* The figures are very small, given the amount of detail in them, in the print ACPD version. The authors should make sure that in the ACP version they are reproduced at a large enough size to be able to comfortably read them.

Minor items, typos etc.

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* 10154-15, the wording of this sentence is slightly ambiguous / confusing. I think the authors mean to say that both V and Ni are high in the WMB and only V in the EMB, but a reader could think that both elements are only high on the WMB. I suggest re-wording

* 10155-2, object -> subject

* 1016-8, giving -> given

* 10157-26: not clear to me what you mean with the 'contrast throughout instead of along the year', suggest re-wording

* 10158-23, does the Gerasopoulos article describe only the prevailing local winds, or also the transport paths / back trajectories to FKL?

* 10159-4, corrected with -> scaled to

* 10160-2: the Turpin et al. reference listed is not the one in which OM/OC values are reported. Rather the following paper should be cited:

Turpin, B. J.; Lim, H. J. Species contributions to PM_{2.5} mass concentrations: Revisiting common assumptions for estimating organic mass. *Aerosol Sci. Technol.* 2001, 35 (1), 602–610. <http://www.informaworld.com/smpp/content~content=a713834675~db=all~order=page>

* 10160-2, a recent paper that also reports measurements of OM/OC ratios with a different technique from that of Turpin and Lim but that arrives at almost the same values is Aiken et al. (*ES&T* 2008, <http://pubs.acs.org/doi/abs/10.1021/es703009q>). I suggest citing this reference as well so that there is broader support for the values chosen.

* 10160-7, the greek letter beta should be used here

* 10162-4, I suggest referring to Figure 1 at this point, otherwise the reader may miss the connection with this discussion

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- * 10162-7, not sure what is meant by '2-day in average samplings'
- * 10162-16, please provide the concentrations measured at the Spanish EMEP stations
- * 10164-4, orphan period at the end of the sentence
- * 10166-15, show -> shows
- * 10166-20, pls give both PM10 and PM2.5 when removing the 6 major dust events, otherwise this sentence is confusing
- * 10167, as later discussed -> as discussed later
- * 10168-21, shore-line -> shoreline
- * 10168-24, suggest inserting 'in the Mediterranean' after 'coastal areas' for clarity, since Atlantic sites are mentioned next
- * 10169-26 and also 10171-3, remove 'the' before 'ammonium nitrate'
- * 10170-4, not clear why there is a reference to Fig 5 here
- * 10170-9, in the coarse mode -> to the coarse mode
- * 10172-4, rate -> ratio
- * 10172-8, suggest inserting 'markedly' after 'change', since the correlation coefficient does change some between the seasons
- * 10172-14, the wording 'agricultural waste, and burning' is unclear. I assume the authors mean 'agricultural waste burning'?
- * Some references that support the lack of significant of OC content in Saharan dust, and which could be cited here are:

Formenti, P., M. O. Andreae, L. Lange, G. Roberts, J. Cafmeyer, I. Rajta, W. Maenhaut, B. N. Holben, P. Artaxo, and J. Lelieveld, Saharan dust in Brazil and Suriname during the Large-Scale Biosphere-Atmosphere Experiment in Amazonia (LBA) - Cooperative

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LBA Regional Experiment (CLAIRE) in March 1998, J. Geophys. Res., 106, 14919-14934, 2001.

Mayol-Bracero, O. L., Santos-Figueroa, G, Morales, F, Colon, L, Carbonaceous Aerosols in African Dust Over the Caribbean, EOS Trans. AGU, Fall Meeting Supp., Abstract A11C-0122, 2008.

- * 10173-3, associated to -> associated with
- * 10173-25, pls define WAE in the text
- * 10175-12-18, this explanation about the depletion of NH3 is clearer and more detailed than the one earlier in the paper, I suggest using the same wording in both locations.
- * 10176-20, it is not clear to me how this study shows that the Mediterranean 'is vulnerable to climate destabilization'

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