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Interactive comment on "Saharan dust aerosol over the central Mediterranean Sea: optical columnar measurements vs. aerosol load, chemical composition and marker solubility at ground level" *by* M. Marconi et al.

F. Dulac

francois.dulac@cea.fr

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Congratulations for your interesting work on Saharan dust in Lampedusa. I find however that the solubility study is much weaker than the rest and could be omitted. Here are a few comments, which I hope will be useful for revision.

First, you might wish to remind that the Lampedusa station is a regional station of the WMO/GAW network.

Regarding the use of nssCa as a tracer for dust, I would like to emphasize that there is a rather high variability in crustal element ratio, as illustrated for instance by the early work of Bergametti et al. (1989) for Si/Al and Fe/Al ratios of dust aerosols in Corsica, depending on the source region of dust in Africa. This puts some uncertainty in using a single chemical tracer to identify desert dust events. In particular, loess deposits from southern Tunisia which are most probably an important source of dust over Lampedusa (see Fig. 5 in Prospero et al., 2002) seem particularly rich in calcite (e.g. Guieu et al., 2010). Avila et al. (2007) reports an average Al/Ca ratio of 0.65 and 0.53 for dust in red rains collected in NE Spain from source regions West and East of Greenwich meridian, respectively, which confirms this trend of higher Ca in long-range transported aerosols from the Great Eastern Erg region. Since you report having used back-trajectories to check that air masses with high nssCa events have an African origin, I wonder whether you could not use farther those air-mass trajectories to also check (i) a possible discrimination between the main dust source regions based on Si, Al, Fe and nssCa ratios, and (ii) the consistency of your single nssCa criteria depending on dust origins.

By the way, information on which trajectories were used is presently missing in the methodology. Information on the OPC fitting procedure is also presently missing in the methods. You might also wish to confirm that the Grimm OPC was used with a TSP sampling head in order to capture large particles despite its low sampling flow (otherwise there is no hope to capture large particles). Regarding cascade impactor samples, the information given in the material and methods section is very incomplete: how many samples, for what type of air mass...? For both the OPC and the cascade impactor, it would be worth reminding cut-off diameters. We also miss information on the daily filter sample set. I find that a table or a figure providing the number of available samples per month of the 6.5-yr sampling period would give a very useful idea of the data set. To finish regarding materials and methods, there is a need for further information regarding extraction methods: I note that the abstract only mentions extraction of the soluble fraction of dust at pH 1.5 in HNO3, whereas section 2 only

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describes a different method with $HNO_3+H_2O_2$ using a microwave oven, when figure 6 finally presents results of 3 different extractions using HNO_3 and a ultrasonic bath, HNO_3-H_2O_2 and a microwave oven, and Milli-Q water and a ultrasonic bath.

I feel that a better contrast in the manuscript should be made between data from long-term monitoring and data from much more limited sampling periods. The latter should be better presented as illustrations or case studies, not yielding general conclusions. In addition those limited data sets could probably be better exploited if they are to be included in the paper. Indeed, information from cascade impactors and the OPC comes from a very limited sampling period compared to the total monitoring period, i.e. less than one particular summer or spring season, respectively, compared to 6.5 yr for long-term monitored parameters. Such campaign results might well not be characteristic of the general situation unless you can argue the opposite. The use made of cascade impactors is the most questionable since it seems to me that you are deriving the size-segregated solubility of various elements using a single impactor sample. Since several samples are supposed to have been analysed from the summer sampling season, we would expect some synthetic information on the variability of the size distribution of the various elements. Regarding OPC data, it also seems that you are averaging data for all dust events of the 2-month sampling period to retrieve a single “average” dust size distribution, which seems rather questionable given the expected variability (for instance depending on the transport distance from source regions). Cannot you perform an inversion for every dust event during the campaign and check the variability? Can you compare the size distributions retrieved for those dust events to the size distribution of non-dust periods? To my knowledge, the Grimm OPC also derives PM1, PM2.5 and PM10 data, and I would expect at least some comparison to coincident PM10 data.

Regarding elemental solubility, I have the feeling that the section is presently too limited to the presentation of results. Added to the above remarks on methodology (description missing, size segregated solubility based on a single impactor sample), I find that this

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part of the paper is particularly weak. The relevant discussion is not complete and hardly accounts for an abundant literature on the topic: there are only 2 references cited (Fairlie et al., 2010, and Becagli et al., 2012) whereas many additional references are expected to be used. For instance, just taking some papers I have in mind, the important role on iron solubility of organic substances (in particular oxalate, see e.g. Saydam and Senyuva, 2002; Paris and Desboeufs, 2013) or of iron mineralogy (e.g. Journet et al. 2007) should be mentioned. The fact that Fe solubility increases with decreasing particle size (top of p.21279) has already been reported (e.g. Baker and Jickells, 2006) but does not seem to be a constant (Buck et al., 2010) and it has also probably much to do with the anthropogenic fraction of iron which is known to be more soluble than the crustal fraction (e.g. Baker et al., 2006) and possibly a forest fire fraction (Guieu et al., 2005). Avila et al. (2007) also addressed the problem of high variability in calcium solubility. Aluminium solubility is deeply discussed by Losno et al. (1993) and the role of acidic species by Losno et al. (1991). Etc. I would therefore recommend to skip the solubility part from the present paper and to submit it in another dedicated manuscript with additional work on cascade impactor samples, and serious consideration of the existing literature.

Finally, a few technical corrections:

-p. 21265, line 27: you refer to the present section 2.1. By the way this sentence on extraction should rather be in section 2.2 where extraction is described.

-you have some misspelling in the references of Spanish authors: Rodriguez et al citations in the text omit the accentuated í that is used in the Ref. list; Gomes Amo in the text (p.21265) is Gómez-Amo in the references.

-you might plot on figure 3 the limits used for dust events selection.

References:

Avila, A., Alarcon, M., Castillo, S., Escudero, M., García Orellana, J., Masque, P., and Querol, X.: Variation of soluble and insoluble calcium in red rains related to dust sources and transport patterns from North Africa to northeastern Spain, J. Geophys. Res., 112, D05210, doi:10.1029/2006JD007153, 2007.

Baker, A.R., and Jickells, T.D.: Mineral particle size as a control on aerosol iron solubility, Geophys. Res. Lett., 33, L17608, DOI: 10.1029/2006GL026557, 2006.

Baker, A.R., Jickells, T.D., Witt, M., and Linge, K.L.: Trends in the solubility of iron, aluminium, manganese and phosphorus in aerosol collected over the Atlantic Ocean, Mar. Chem., 98, 43-58, 2006.

Bergametti, G., Gomes, L., Remoudaki, E., Desbois, M., Martin, D. and Buat-Ménard, P.: Present transport and deposition patterns of African dusts to the north-western Mediterranean, iIn Paleoclimatology and Paleometeorology: Modern and Past Patterns of Global Atmospheric Transport, M. Leinen and M. Sarnthein, Kluwer Academic Publishers, 227-251, 1989.

Buck, C.S., Landing, W.M., and Resing, J.A.: Particle size and aerosol iron solubility: A high-resolution analysis of Atlantic aerosols, Mar. Chem., 120, 14–24, 2010.

Guieu, C., Bonnet, S., Wagener, T., and Loÿe-Pilot, M.-D.: Biomass burning as a source of dissolved iron to the open ocean?, Geophys. Res. Lett., 32, L19608, doi:10.1029/2005GL022962, 2005.

Guieu, C., Dulac, F., Desboeufs, K., Wagener, T., Pulido-Villena, E., Grisoni, J.-M., Louis, F., Ridame, C., Blain, S. Brunet, S., Bon Nguyen, E., Tran, S., Labiadh, M, and Dominici, J.-M.: Large clean mesocosms and simulated dust deposition: a new methodology to investigate responses of marine oligotrophic ecosystems to atmospheric inputs, Biogeosci., 7, 2765-2784, doi:10.5194/bg-7-2765-2010, 2010.

Journet, E., Desboeufs, K., Caquineau, S., and Colin, J.-L.: Mineralogy as a critical factor of dust iron solubility, Geophys. Res. Lett., 35, L07805, 2008.

Losno, R., Bergametti, G., Carlier, P., and Mouvier, G.: Major ions in marine rainwater with attention to sources of alkaline and acidic species, Atmos. Environ., 25A, 763-770, 1991.

Losno, R., Colin, J.L., Le Bris, N., Bergametti, G., Jickells, T., and Lim, B. : Aluminium solubility in rainwater and molten snow, J. Atmos. Chem., 17: 29-43, 1993.

Paris and Desboeufs, K: Effect of atmospheric organic complexation on iron-bearing dust solubility, Atmos. Chem. Phys., 13, 4895-4905, 2013.

Prospero, J.M., Ginoux, P., Torres, O., Nicholson, S.E., and Gill, T.E.: Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product, Rev. Geophys., 40, 2-1 to 2-31, 2002.

Saydam, A. C. and Senyuva, H. Z.: Deserts: Can they be the potential suppliers of bioavailable iron?, Geophys. Res. Lett., 29, 1524, 2002.

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