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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Climatology of aerosol optical properties and black carbon mass absorption cross section at a remote high altitude site in the Western Mediterranean Basin" *by* M. Pandolfi et al.

Anonymous Referee #2

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Referee comment on Pandolfi et al., "Climatology of aerosol optical properties and black carbon mass absorption cross section at a remote high altitude site in the Western Mediterranean Basin"

This paper provides a climatology of the aerosol optical properties measured at an elevated site in Spain (Montsec, MSC). EC/OC measurements were also performed, although only the EC results are discussed in this manuscript. Several air mass classification methods were used to identify air mass source regions via cluster analysis and

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to explain differences in observed aerosol properties as a function of air mass type. Because the site is at altitude (1570 m asl), they also segregate the aerosol by time to look at statistical differences in optical properties between 'free troposphere' air and 'all data'. They explore how the different aerosol properties change as a function of aerosol loading and finally they report on the seasonal variation of mass absorption coefficient (e.g., absorption/massEC).

The Andrews et al. (2011) paper which is cited throughout the manuscript appears to be the basis for a significant amount of the analysis presented in the manuscript. Given how closely the discussion in this manuscript tracks the Andrews et al (2011) I think it's appropriate that the Andrews paper is mentioned in the abstract somewhere.

The Andrews et al. (2011) paper seems to be the primary source of references for the manuscript. I see two issues with this. First, why do the authors not cite other mountain sites in Europe in addition to those mentioned in the Andrews (2011) paper? Some of these (e.g., Puy de Dome, Hohenspeizenberg) are probably more similar to MSC than Jungfraujoch and Beo Moussala are in terms of aerosol sources and flows. Even if the MSC instrumentts aren't identical to what is made at some of the other sites (PUY, HPB) there are papers for those sites discussing FT aerosol and sources. Second, it is unclear whether the authors read some of the references they cite. For example, they cite Delene and Ogren (2002) as an example of 'high altitude aerosol optical measurements'. The highest site mentioned in that paper is at 315 m asl.

Somewhere in this manuscript it should be noted that MSC is at significantly lower altitude than any of the sites in Andrews et al (2011). The lowest site in Andrews et al 2011 is at 2.2 km, approximately 700 m asl higher than MSC, which is approximately the height difference between MSC and MSY. How might you expect that to affect your comparisons with the sites in the Andrews 2011 paper?

How are the hourly averages of the intensive parameters calculated? Are they calculated from the hourly averaged extensive parameters with the hourly averaged val-

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ues<detection limits not included? Or are they calculated from the high frequency data with the values<detection limits not included and then averaged to get hourly intensives? The first way is how it was done in Andrews et al (2011), with the constraint that BsG>1 Mm-1.

Rearrange the order of section 3 so that all diurnal cycle discussions are talked about sequentially. Right now it goes from annual (fig 2) to diurnal (fig 3) to seasonal (fig4) to diurnal (fig 5).

The Andrews et al (2011) paper used a simplified scheme based on time of day to identify the FT. They chose this approach because (a) they were dealing with 12 sites, some with more than a decade of measurements) making it difficult to do detailed analysis for individual sites (b) not all sites provided additional meteorological data. It is fine to use the same simple criteria for comparisons with the Andrews paper, but it would be more useful if the authors could also include and evaluate the differences in aerosol properties between the simple time-based FT identification and something a little more specific to MSC FT conditions.

Is there lidar (e.g., from EARLINET?) or sonde data that gives an indication of the height of the boundary layer near MSC and MSY as a function of season? How confident are you that MSC is in the FT during the summer? A plot like figure 5 in Venzac et al 2009 (full citation in Andrews et al 2011) would be useful.

I am not convinced that the differences between FT and all data presented in figure 4 are statistically significant. Is there a statistical test you can do to show that they are?

It would be interesting to see the systematic relationship plots as a function of airmass type in addition to looking at the overall pattern.

Can the authors also present mass scattering cross section (MSCS)? Could they subtract the EC mass from the PM10 mass to get an approximation of the scattering mass and then take the ratio of that with the measured scattering (appropriately averaged

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over the filter time)? It would be interesting to see if the MSCS differs as a function of air mass type.

Page 3 Line 2 – '..performed at a regional background station. . .' make more clear that this is a low altitude site upstream of MSC?

Line 11 – '(IPCC, 2007, 2013)' incorrect reference format? Should be (IPCC, 2007; IPCC, 2013)?

Line 13-14 '..black carbon (BC) mass absorption cross section (MAC), among others, can be derived from these optical properties.' Derivation of MAC needs more than just optical properties.

Line 18-19 '..whereas soot particles (or BC) have strong absorbing properties over the entire visible spectrum. . .' particles also scattering strongly over the visible spectrum. I think the point you want to make is that BC particles affect the vertical heating profile in addition to TOA effects.

Line 23-24 '. . .atmospheric models convert the modelled/measured BC mass concentrations. . .' I agree that models convert modelled BC to absorption using a MAC value. Do the big climate models actually ingest measured BC values? My understanding (perhaps wrong) is that in-situ measurements tend to be too sparse for global models to be able to work with. So. . .I'd suggest removing 'measured' from the sentence unless there's a citation/clarification that can be made.

Page 4 Line 19-21 "However, lidars do not measure key climate variables such as aerosol absorption, SSA or MAC and the in-situ measurements (which provide these key variables) are often limited to the PBL." This is repetitive since the previous sentence says that in-situ measurements are primarily limited to the PBL.

Page 5 Line 2 'Delene and Ogren (2002)' does not include data from any high altitude sites

Line 3-4 'Andrews et al., 2004' is an aircraft in-situ measurements paper.

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Line 7 '. . .high altitude aerosol optical measurements. . .' is there a citation/webpage for this?

Page 6 Line 10 'Particles' CHANGE TO 'Particle.

Line 10 The Ecotech neph measures from scattering from 10-171o and backscattering from 90-171o (Table 1, Müller et al., 2011). Truncation corrections adjust for the instrument's angular non-idealities, allowing reporting of scattering for 0-360o and backscattering for 90-270 o.

Line 13 'All acronyms used in this work are reported in Table 1.' I would suggest moving this sentence to the end of the last paragraph of section 1 (page 5, line 16).

Page 7 Line 7:'. . .converted by the MAAP's software in BC concentration. . .' CHANGE TO '. . .converted by the MAAP's software to BC concentration. . .'

Page 8 Line 10 '. . .derived starting from the performed optical measurements. . .' CHANGE TO '. . .derived from the aerosol optical and mass concentration measurements. . .'

Line 16 '. . .the three nephelometer wavelengths as it follows. . .' CHANGE TO '. . .the three nephelometer wavelengths as follows. . .'

Line 24 'In this work SSA was estimated at 635 nm.' Add to this sentence: 'In this work SSA was estimated at 635 nm, as the difference between absorption at 637 nm (MAAP wavelength) and 635 nm was assumed to be negligible.

Page 9 'Given the position of MSC station often in the free troposphere, the B/S, g, SSA and SAE parameters were estimated by using only data (scattering, hemispheric backscattering and absorption) above detection limit (DL) of the instruments.' This sentence isn't really clear. Presumably you are implying that the FT is clean and so instruments often are measuring close to their detection limit. A better way to put it would be 'In order to eliminate issues with measurement noise during clean periods (e.g., when MSC was in the FT), the B/S, g, SSA and SAE parameters were only

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calculated when the hourly-averaged data (scattering, hemispheric backscattering and absorption) were above the detection limit (DL) of the instruments.'

P10 Line 24 I believe Figure 2 would be better replaced by a seasonal plot (e.g., fig 4) or a monthly cycle plot since much of the discussion that follows mentions seasonality before the seasonal plots are presented. The hourly time series is not so helpful to the reader and the statistics for the time series are provided in the table so it seems fairly extraneous.

P11 Line 17 'The Hourly PM1 levels. . .' Hourly does not need to be capitalized.

Line 27-28 'At Izaña (Canari Island, 2400 kma.s.l.) . . .' CHANGE TO 'At Izaña (Canary Islands, 2400 m a.s.l.)

P12 Line 1-3 'In Italy (Monte Cimone, 2200ma.s.l.) Andrews et al. (2011) reported mean _sp and _ap of around 11Mm−1and 3Mm−1, respectively.' There are detailed papers for Monte Cimone discussing dust and other sources. For example: Cristofanelli et al., Atmos. Chem. Phys., 9, 4603–4619, 2009 Marenco, F., et al. (2006), Characterization of atmospheric aerosols at Monte Cimone, Italy, during summer 2004: Source apportionment and transport mechanisms, J. Geophys. Res., 111, D24202, doi:10.1029/2006JD007145. Marinoni et al, (2008) Science of the Total Environment. 391, pp 241-251.

Line 3 'The values measured at MSC were lower compared with the . . .' CHANGE TO 'The mean scattering and absorption values measured at MSC were lower than. . .'

Line 5-7 '. . .due to the higher influence of anthropogenic emissions at regional level (MSY) compared with remote level (MSC).' CHANGE TO '. . .due to the higher influence of anthropogenic emissions in the lower boundary layer (MSY) compared with at higher altitude (MSC).'

Line 12-17 'The main difference was observed for SAE which value was slightly higher at Jungfraujoch (1.671–1.787) compared with MSC (1.56) indicating the prevalence

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of slightly coarser aerosols at MSC, probably due to the difference in intensity and frequency of NAF episodes between the two sites, with MSC site more affected.' CHANGE TO: 'The main difference was observed for SAE which was slightly higher at Jungfraujoch. . .' Also, what is the uncertainty in the Angstrom exponent? Are these numbers really different? What is the prevalence of dust storms at JFJ? There are several "dust at JFJ" papers – does one of them provide stats for that? Could the higher SAE at JFJ be because more large particles have time to deposit out or be removed via wet scavenging since JFJ is 2x higher than MSC?

Line 16-17 'At the regional background station of MSY mean B/S, SAE and SSA of 0.135, 1.33 and 0.90, respectively, were measured.' SAE at MSY is lower than at MSC. Is this because it experiences more NAF events?

Line 19 'As shown later (paragraph 3.5). . .' CHANGE TO 'As shown later (section 3.5). . .'

Page 13 Line 2 '. . .at MSC showed diurnal. . .' CHANGE TO '. . .at MSC exhibited diurnal. . .'

Line 7-11 rewrite for better grammar

Line 12-13 '. . .the effectiveness of the transport of fine highly absorbing particles of anthropogenic origin at RB sites. . .' this is a generalization which may not be true of all regional background sites (for example it is not true at the two continental surface sites in Delene and Ogren, 2002, which are also regional background sites). It should just say '. . .the effectiveness of the transport of fine highly absorbing particles of anthropogenic origin at MSY. . .'

Line 14-15 '. . .of the considered synoptic scenarios. . .' CHANGE TO '..of the different synoptic scenarios. . .'

Line 19 'Similar pattern. . .' CHANGE TO 'A similar pattern. . .'

Line 27 spelling antyciclonic' CHANGE TO 'anticyclonic'

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Page 14 Line 2-4 '. . .as a consequence the proximity of MSY station to important BC anthropogenic sources (cities and industrialized/urbanized coastline). . .' CHANGE TO 'due to the proximity of the MSY station to anthropogenic sources of BC. . .'

Line 16-17 '. . .are related with the location. . .' CHANGE TO '. . .are related to the location. . .'

Line 27 '. . .colours in Fig. 4 stay. . .' CHANGE TO '. . .colours in Fig. 4 represent. . .'

Page 15 Line 6 '. . .represent the ALL median values. . .' CHANGE TO '. . .represent the median values. . .'

Line 11 '. . .the aerosols mass. . .' CHANGE TO '. . .the aerosol mass. . .'

Line 25-26 '. . .a function of seasons at MSC site. . .' CHANGE TO '. . .seasonal at the MSC site, . . .'

Page 16 '. . .to the concentrations. . .' CHANGE TO '. . .to the concentration. . .' Line 11 'These scenarios also. . .' CHANGE TO 'These scenarios are also. . .'

Line 18 'This kind of relationships. . .' CHANGE TO 'These kind of relationships. . .'

Page 17 Line 2-3 'This is the case of MSC site showing a scattering–absorption slope. . .' CHANGE TO 'This is also the case at MSC which shows a scattering-absorption slope. . .

Line 9 '. . .with larger scattering particles. . .' CHANGE TO '. . .dominated by dust (i.e., large, primarily scattering) particles. . .'

Line 12 'As shown in Fig. 7a . . .' CHANGE TO ' 'As shown in Fig. 7a, . . .'

Line 16 '. . .SAE values (from -2 to 6). . .' are these values calculated using the system detection limits and hourly averages? What is the mean and stdev of the scattering for these values? Perhaps you should use higher values than the detection limits of the data when calculating the intensive properties. For example, Andrews et al 2011 only

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calculated intensive properties when green scattering at STP > 1 Mm-1.

Page 18 Line 8-10 'However, under very low PM1 concentrations at MSC (PM1 < 1.5 μgm$-$3) SSA and g reached very low values around 0.84 and 0.43, respectively, whereas the SAE increased (reaching around 1.6).' how would these findings change if higher constraint was chosen (e.g., only calculate SSA and g when scattering > 1 Mm-1) Also where is the plot of PM1 vs SSA and g? What are the PM10 values that go along with these very low PM1 values?

Line 19-20 'Mean MAC at MSC determined as the error-weighted slope of the absorption-EC concentration scatterplot was. . .' CHANGE TO 'Mean MAC at MSC, determined as the error-weighted slope of the absorption-EC concentration scatterplot, was. . .'

Line 23 'On average lower. . .' CHANGE TO 'On average, lower. . .'

Line 28 '. . .NAF and SREG,. . .' CHANGE TO '. . .NAF and SREG. '

Page 19 Line 1 '. . .compared with those obtained for AA and WREG. Conversely, no. . .' CHANGE TO 'Conversely, no . . .'

Line 17-19 'On average, also the pollutants transported toward the MSC station by the up-slope winds and PBL oscillations, which are expected to be more intense in summer, contributed to the observed MAC annual cycle.' This sentence doesn't really make sense (or it's restating what you've already described above).

Line 25 '. . .rather little. . .' CHANGE TO '. . .limited. . .'

Page 20 Line 7-10 'As a consequence, the strongest diurnal cycle (DC) for scattering at MSC was observed in winter, whereas in spring and summer no clear DC was observed due to the presence of polluted layer at the MSC altitude.' The authors did not convince me that they were observing polluted layers as opposed to the boundary layer height > height of MSC.

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Line 15 '. . . (B/S) were on the. . .' CHANGE TO '. . .(B/S) were in the. . .'

Line 16-19 'Under Atlantic advection (AA) scenarios the lowest scattering and absorption were measured at MSC due to renovation of accumulated pollution in the aged air masses typically associated with AA episodes in the WMB.' I thought the AA scenario was typically representative of cleaner marine air. Also the phrase 'renovation of accumulated pollution' doesn't make sense.

Line 22-24 'These decreases were clearly a function of seasons at MSC site with the highest FT vs. all-data difference observed in winter and the lowest in spring/summer.' Possibly because FT conditions are less frequent in the spring/summer due to higher boundary layer?

Table 1 – retitle 'Acronyms used in this work' The table is helpful, but I think in general the authors overuse acronyms. For example daily cycle (DC) in section 3.2, which is not included in the acronym table, doesn't need to be acronymized. Western Mediterranean Basin (WMB) and Regional Background (RB) are other acronyms that seem unnecessary.

Figure 2 – delete? Other than showing the time series, I don't think it's discussed at all. The only information it appears to provide is data coverage for the different. Replacing the time series with monthly plots might be more useful to the reader? Check out figure 1a in Venzac et al., 2009 (full citation in Andrews 2011) as an example of a plot that might be more useful to the reader.

Figure 6 is nice – I think it is helpful to include the number of points in each scattering bin as was done. It would be interesting to see the systematic variability as a function of airmass type – that would be new and unique I think.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 3777, 2014.