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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.

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We thank the reviewer#2 for the useful comments on our paper.

Anonymous Referee #2 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 classiïňĄcation methods were used to identify air mass source regions via cluster analysis and 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 ïňĄnally they report on the seasonal variation of mass absorption coefïňĄcient (e.g., absorption/massEC).

1) The Andrews et al. (2011) paper which is cited throughout the manuscript appears to be the basis for a signiïňĄcant 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.

A sentence in the abstract was added. "The scattering measurements performed at MSC locate this site in the medium/upper range of values reported by Andrews et al. (2011) for other mountaintop sites in Europe mainly due to.....".

2a) 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 ïňĆows. Even if the MSC instruments 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.

The following sentences were modified accordingly to the reviewer's comment: a) Paragraph 3.1: "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 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."

Was replaced with:

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“At the alpine Jungfraujoch site, Fierz-Schmidhauser et al. (2010) reported mean B/S, SAE and SSA for dry aerosols (RH<20%) within the ranges 0.128-0.122, 1.787-1.671 and 0.91-0.93, respectively. The higher B/S measured at MSC was likely due to the lower size range measured at MSC (PM2.5) compared with Jungfraujoch (whole air) whereas the lower SAE at MSC, despite the different size cut-off, may be an indication of the prevalence of slightly coarser aerosols at MSC. This difference was likely due to the fact that more large particles had time to deposit out or be removed via wet scavenging since the altitude of Jungfraujoch is about twice the MSC altitude.”

b) Paragraph 3.2: “Thus, on average, thermally driven upslope winds and PBL height oscillations favour the transport of pollutants toward the MSC site during the warmest hours of the day.”

Was replaced with:

“Thus, on average, thermally driven upslope winds and PBL height variations favoured the transport of pollutants toward the MSC site during the warmest hours of the day. Similar diurnal variations at other mountain top sites have been observed for extensive aerosol optical properties (e.g. Andrews et al., 2011) and physical properties (e.g. Venzac et al., 2009; Marinoni et al., 2008).”

The following reference was added to the bibliography: Venzac, H., Sellegri, K., Villani, P., Picard, D., and Laj P.: Seasonal variation of aerosol size distributions in the free troposphere and residual layer at the puy de Dome station, France, Atmos. Chem. Phys., 9, 1465–1478, doi:10.5194/acp-9-1465-2009, 2009.

c) End of paragraph 3.3 “The observed reduced DC for scattering at MSC during spring and summer was due to the occurrence of NAF and SREG episodes and wildïňĄres in the WMB, together with the possible presence of intense polluted residual layers at night at the MSC altitude. These scenarios also linked with the high concentrations of PM and BC during the whole day as observed by Ripoll et al. (2014).”

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was replaced with:

"The observed reduced diurnal cycle for scattering at MSC during spring and summer was mainly due to the frequent occurrence of NAF and SREG episodes and to the possible presence of polluted residual layers at night at the MSC altitude. The NAF and SREG scenarios were also linked with high concentrations of PM and BC during the whole day by Ripoll et al. (2014). Thus, as already observed by Venzac et al. (2009) for the Puy de Dôme station, the seasonal change of FT data at MSC was in part due to the seasonal variability in air mass origin and transport routes."

d) Paragraph 3.1 (See also reviewer's comment #35): We improved the part related with the comparison with the regional background MSY station by using mean values calculated from data simultaneously collected at both stations. Consequently, the following sentence:

"At the regional background station of MSY mean B/S, SAE and SSA of 0.135, 1.33 and 0.90, respectively, were measured."

Was replaced with:

"At the MSY regional background station the mean B/S (525 nm), SAE (450-635 nm), SSA (635 nm) and g (525 nm) were different by around -16%, +5%, -4% and +6%, respectively, compared to MSC (only contemporary data were used). These differences were small suggesting, on average, similarity in microphysical aerosol properties measured at MSC and MSY stations. Recently, Ripoll et al. (2014) have shown that the mean chemical composition of particles at MSC and MSY is on average similar mainly due to the frequency of specific meteorological episodes affecting aerosol properties similarly at both sites. The main difference was observed for B/S which was higher at MSC likely because the lower size cut-off at MSC (PM2.5) compared to MSY (PM10). Moreover, the relatively lower SSA and higher SAE at MSY, despite the differences in the size cut-off, suggested the presence of relatively smaller and more absorbing particles at regional level likely because the proximity of MSY station to anthropogenic

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sources."

2b) 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.

The Delene and Ogren (2002) paper was removed from the Introduction.

3) Somewhere in this manuscript it should be noted that MSC is at signiïňĄcantly 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?

The following sentence: "The NAF and SREG episodes affecting the WMB have the potential to increase the aerosols mass and scattering measured in this area in summer (Fig.3 and JJA scattering in Fig.4a). Moreover, the high solar radiation in the Mediterranean Basin (especially in summer) favours the development of up-slope winds which, together with the possible presence of polluted residual layers at MSC altitude, contribute to the levels of scattering and absorption measured at MSC. Recently, Ripoll et al. (2014) have shown that the MSC site registers higher PM10 concentrations than those measured at other high-altitude central European sites and similar or lower BC concentrations."

was replaced with:

"As already noted, the NAF and SREG episodes affecting the WMB have the potential to increase the aerosol mass and scattering measured in this area in summer (Fig.2 and JJA scattering in Fig.4). At the same time, the lower altitude of MSC station compared with the stations reported in AND2011 (lowest altitude around 2.2 km) may have also contributed to the relatively higher scattering observed at MSC station."

4) How are the hourly averages of the intensive parameters calculated? Are they cal-

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culated from the hourly averaged extensive parameters with the hourly averaged values<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 ïňĄrst way is how it was done in Andrews et al (2011), with the constraint that BsG>1 Mm-1.

Here we applied the same methodology as in Andrews et al. (2011) but with different constraint. The following sentence in the paragraph 2.5 (Data processing):

"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." Was replaced with the following sentence (see also the reviewer's comment #27): "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 calculated from hourly-averaged scattering (635 nm), hemispheric backscattering (635 nm) and absorption above 0.4 Mm-1, 0.4 Mm-1 and 0.6 Mm-1, respectively."

5) Rearrange the order of section 3 so that all diurnal cycle discussions are talked about sequentially. Right now it goes from annual (ïňĄg 2) to diurnal (ïňĄg 3) to seasonal (ïňĄg4) to diurnal (ïňĄg 5).

Following the reviewer comments, the section 3 was changed and it is now organized as follows:

3.1 General features (Figure 2 was removed; see also reviewer comment #28); 3.2 Diurnal cycles and cluster analysis (Here we present the diurnal cycles and cluster analysis) 3.3.1 Identification of FT air (Here we report the diurnal cycles of normalized scattering and meteorological parameters; see also reviewer comments #6 and #7) 3.3.2 FT vs. all data: Comparison with mountaintop sites presented in AND2011 (Here we discuss the seasonal variation of aerosol optical extensive and intensive optical properties and compare MSC with AND2011).

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6) and 7) The Andrews et al (2011) paper used a simpliïňĄed 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 difiňĄcult to do detailed analysis for individual sites (b) not all sites provided additional meteorological data. It is ïňĄne 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 identiïňĄcation and something a little more speciïňĄc 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 conïňĄdent are you that MSC is in the FT during the summer? A plot like ïňĄgure 5 in Venzac et al 2009 (full citation in Andrews et al 2011) would be useful. The closest lidar station to MSC is located in Barcelona where radiosoundings are also available. We have a database with more than 2500 Mixing Layer Heights (MLH) calculated from radiosoundings at 12:00 UTC for the period 2003-2010 in Barcelona. The mean MLH over Barcelona at midday as a function of the seasons is reported in the Figure A below.

The lack of an annual cycle of MLH in Barcelona is due to the sea breeze which limits the convective growth of MLH. As reported in the Figure A, the MLH measured in Barcelona is always lower than the MSC altitude (a part from very sporadic episodes). We have shown (Pandolfi et al.: Effects of sources and meteorology on particulate matter in the Western Mediterranean Basin: An overview of the DAURE campaign, J. Geophys. Res. Atmos., 119, doi:10.1002/2013JD021079, 2014) that the variations of Barcelona MLH contribute to the levels of pollutants measured at regional level (MSY, which is at 720 m a.s.l. and around 50km from Barcelona). However, we cannot conclude that MSC station is in the free troposphere by looking at the MLH calculated at Barcelona, due to the distance between MSC and Barcelona and the complicated orography of the area. So this first method is not useful to show when MSC is in the free troposphere. Consequently, we used contemporary meteorological data collected at MSC station (1570 m a.s.l.), at the Montsec Observatory (800 m), at Os de Balaguer

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(576 m) and Vallfogona de Balaguer (238 m) to study the strength of the nocturnal and diurnal thermal inversions between mountain (MSC) and valley. Figure 1 (reported below) was changed in order to show the location of these meteorological stations.

The distance between MSC and Vallfogona de Balaguer is around 35 km. We used hourly meteorological data collected at these 4 stations in order to study the mean diurnal cycles of relative humidity, water vapour mixing ratio and potential temperature with the aim to estimate when the MSC station was in the free troposphere. Moreover, we also calculated the diurnal cycle of PBL height at MSC with HYSPLIT. The Figure 3 below (which was added to the manuscript as Figure #3) shows the results of this analysis:

Consequently, the following sentence was added to the new section 3.3.1 "In order to evaluate when the MSC station was in the FT, we used meteorological data collected at MSC and at three lower altitude meteorological stations (Fig. 1). Thus, contemporary meteorological data collected at MSC station (1570 m a.s.l.), Montsec Observatory (800 m), Os de Balaguer (576 m) and Vallfogona de Balaguer (238 m) were used to study the mean diurnal cycles of potential temperature (Fig.3a), relative humidity (Fig.3b) and water vapour mixing ratio (Fig.3c) as a function of altitude. The potential temperature and water vapour mixing ratio were calculated with the humidity conversion formulas provided by Vaisala (Vaisala Oyj, 2013). Moreover, the diurnal cycles of the gradients of potential temperature (Fig.3e) and actual temperature (Fig.3f) were also reported to study the strength of the nocturnal and diurnal thermal inversions between the four sites (i.e. between mountain and valley). This analysis may be affected be differences due to different instruments, calibration procedures or local features associated to a specific location (the MSC station and Vallfogona de Balaguer were around 35km apart). Consequently, we also simulated the mean seasonal PBL diurnal cycles at MSC (Fig.3d) by means of HYSPLIT model (http://www.ready.noaa.gov/READYamet.php). Grey and yellow rectangles in Fig.3 highlight hours when the MSC station was within the PBL and the hours of the

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time of the day approach (from 3:00 to 9:00 local time) for the identification of FT air proposed by Andrews et al. (2011), respectively. In this analysis we assumed that in a well mixed mixing layer the water vapour mixing ratio and potential temperature should be nearly constant with altitude within the PBL. In the free troposphere the water vapour content and potential temperature will decrease and increase, respectively, with altitude. Moreover, if the mixing layer has a uniform distribution of water vapour throughout, then the relative humidity has to increase with altitude. Fig.3 shows that when the relative humidity at MSC was higher compared to the other three stations, the potential temperature and water vapour content were fairly similar. We used these conditions to define the PBL air (grey rectangles). Conversely, at night/early morning (yellow rectangles) the relative humidity at MSC was the lowest and the differences in potential temperature and water vapour content among the four stations were the highest. Moreover, the gradients of potential temperature and actual temperature show that strong inversions were on average observed at night between Observatory and Os de Balaguer with MSC station above the inversion. Conversely, the gradients were lower and rather similar when MSC was within the PBL (grey rectangles). Fig. 3 also shows that our estimation of PBL conditions obtained using meteorological data agrees satisfactorily with the simulation performed with HYSPLIT. Thus, the MSC station was on average above the inversion at night-early morning and within the PBL during the warmest hours of the day in summer, spring and autumn. Thus, the presence of polluted PBL residual layers at MSC altitude at night cannot be excluded. Conversely, in winter the MSC was on average in the FT during the whole day."

Other small part of the text related with this comment were changed throughout the revised version of the manuscript.

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

The non-parametric Kruskal-Wallis test was applied to check whether the differences

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between all-data and FT-data medians were statistically significant.

The following sentence was added to the new section 3.3.2:

“The non-parametric Kruskal-Wallis test was applied to study whether the differences between all-data and FT-data medians were statistically significant. In Fig. 5 bold green numbers indicate statistically significant differences to a significance level of 5% (p-value < 0.05), blue bold numbers highlight marginally significant differences (p < 0.1) whereas black numbers report differences which were not statistically significant (p > 0.1).” Figure 4 (now Figure 5 reported below) and caption were accordingly modified.

Moreover, the following sentences have been accordingly modified:

“However, the relative decreases in FT-data extensive properties were clearly seasonal at the MSC site with the highest FT vs. all-data difference observed in winter (DJF in Figs.5a,b,c; 21-23%) and the lowest in spring/summer (MAM and JJA in Figs.5a,b,c; 0-8%). The differences between the FT-data and all-data medians were statistically significant for ALL, SON and DJF and marginally significant for JJA.” “Finally, the differences between the FT-data and all-data medians calculated for the intensive aerosol properties were statistically significant only for SAE in winter (DJF) with slightly higher SAE observed for FT air (1.47; Fig.5e DJF, FT-data).”

9) It would be interesting to see the systematic relationship plots as a function of air-mass type in addition to looking at the overall pattern We have replaced Figure 6 with a new Figure 6 (reported below) where the systematic relationships were reported for all data and for the different considered meteorological scenarios. Moreover, and taking into account the reviewer’s comment #67, the number of hourly values in each bin was added.

The paragraph 3.4 was accordingly modified. Below we report part of the paragraph 3.4.

“3.4 Relationships between s_sp and other extensive/intensive aerosol properties Fig-

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ure 6 shows the relationships between s_sp and some of the measured extensive backscattering, absorption, PM1 concentrations) and intensive (g, SAE, SSA) aerosol properties. Relationships are presented for the whole database (ALL; highlighted by the blue rectangle) and as a function of the main considered meteorological patterns (AA, NAF, SREG, WREG). Similar relationships were investigated e.g. by Delene and Ogren (2002), Pandolfi et al. (2011) and Andrews et al. (2011). However, to our knowledge this is the first time that these relationships are presented as a function of air mass type. These kind of relationships helps to constrain model parameterizations and to reduce uncertainties in the algorithms used for deriving intensive aerosol properties from remotely sensed data (Delene and Ogren, 2002). In Fig. 6 the frequency distribution of scattering coefficient at 635 nm was calculated for values between 0 Mm−1 and 90 Mm−1 with a bin of 5 Mm−1 for ALL and NAF scenario. For AA, SREG and WREG scenarios the occurrence for high s_sp was lower thus limiting the frequency distributions to 75 Mm-1, 75 Mm-1, and 55 Mm-1, respectively. As shown in Fig. 6 the frequency distribution of aerosol scattering was more right-skewed during AA (sk = 1.93) and WREG (sk = 1.45) compared with NAF (sk = 0.92) and SREG (sk = 0.83) when pollution levels at MSC were higher (Fig. 2). As reported in Fig. 6 the backscattering and PM1 concentrations increased with increasing s_sp. If the PM1 concentrations increase the intensity of light scattered, and also backscattered, increases almost monotonically. Also the absorption, which is roughly proportional to the concentration of absorbing aerosols in PM samples, increased with increasing scattering and PM1 concentration. Similar findings were reported by Pandolfi et al. (2011) for the MSY site and by AND2011 (Fig.6a in AND2011) for the 10 stations considered in their paper. AND2011 observed that the measurement sites which were dominated by dust aerosol at high aerosol loading tend to have a lower slope of the scattering-absorption relationship than the other sites. This is also the case at MSC which shows a scattering-absorption slope in the lower range of those reported by AND2011 (cf. with Fig.6a in AND2011). Fig. 6 shows that the increase of the extensive aerosol properties with increasing aerosol scattering was fairly similar during the

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different considered meteorological scenarios thus leading to very similar SSA as also reported in Fig. 2. Conversely, the most remarkable difference among the scenarios was observed for SAE which decreased with increasing scattering during NAF reaching values around 0.8 at s_sp =90 Mm-1. Thus, the SAE was clearly a function of NAF intensity. Conversely, during SREG SAE was higher and nearly constant (around 1.8 for 0<s_sp<90 Mm-1) indicating the prevalence of smaller particles with a relatively lower g (0.53 during SREG compared with 0.57 during NAF). No remarkable differences were observed for the intensive aerosol properties between the less polluted WREG and AA scenarios."

Other small parts of section 3.4 related with this comment were changed in the revised version of the manuscript.

10) 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 over the ïňĄlter time)? It would be interesting to see if the MSCS differs as a function of air mass type. Following the reviewer comment, the MSCS was calculated and studied as a function of the different atmospheric scenarios. The Paragraph 3.5 and Figure 8, reported below, have been accordingly changed.

"3.5 MAC and MSCS climatology Mean MAC, at Montsec determined as the error-weighted slope of the absorption-EC scatterplot, was 11.1±0.3 m2/g (R2=0.82). Given that ïĄşap and EC concentrations measurements were available since the end of 2009, the mean MAC presented here was calculated over the period November 2009 – June 2013 (384 sample pairs on 24h base). Mean MSCS at 635 nm (228 sample pairs) was 2.5±1.3 m2/g. MSCS at 525 nm and 450 nm are reported in Table 2. On average, lower MAC values were observed during AA (9.7±0.7 m2/g; R2=0.77) and WREG (9.4±1.0 m2/g; R2=0.88) scenarios compared to NAF (11.9±0.7 m2/g; R2=0.61) and SREG (12.6±1.0 m2/g; R2=0.74) scenarios. Similarly, low MSCS was on average observed during AA and WREG (2.0±1.1 m2/g and 1.5±0.6 m2/g, respectively at 635

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nm) whereas MSCS was higher during NAF and SREG (3.7±1.4 m2/g and 3.5±0.7 m2/g, respectively). The non-parametric Kruskal-Wallis test was used for testing the equality of medians among the four selected categories (scenarios). The difference between the NAF and SREG medians was not statistically significant ($p>0.5$) for both MAC and MSCS. The same was observed for the AA and WREG medians ($p>0.3$). Conversely, statistically significant differences ($p<0.001$) were observed between the medians calculated for WREG and AA and those calculated for NAF and SREG. The higher MAC and MSCS under NAF and SREG compared to AA and WREG were likely due to differences in particles origin and particle properties during these scenarios. The SREG scenario is a summer scenario (cf. Fig. 8) which favours the recirculation and aging of pollutants in the WMB. Several publications have shown higher sulphate (e.g. Pey et al., 2009; Querol et al., 1999) and organic matter concentrations (e.g. Querol et al., 2013; Pandolfi et al., 2014) in summer compared to winter in the WMB at regional and remote levels. The summer sulphate and organic matter maxima were due to higher temperatures and increased photochemistry in the atmosphere enhancing the SO2 oxidation and the formation of secondary organic aerosols from biogenic emissions from vegetation (Seco et al., 2011). Moreover, Ripoll et al. (2014) have shown higher concentrations of BC particles in the warmer months at MSC attributed to the impact of the SREG episodes and to the higher occurrence of wildfires in North Africa and/or in the WMB (Cristofanelli et al., 2009). Once formed these particles can recirculate and age under SREG scenario in the WMB. On the other hand the NAF scenarios, which are more frequent in summer in the WMB (Pey et al., 2013), increase the concentration of mineral dust in the atmosphere. Moreover, Rodríguez et al. (2011) and Ripoll et al. (2014) have shown that pollutants such as sulphate and BC may be transported together with dust across the WMB during NAF episodes. The mixing of BC particles with other chemical components, such as sulphate and organics, have the potential to increase the absorption properties of BC particles (e.g. Bond et al., 2013) and could explain the higher MAC observed at MSC during NAF and SREG. At the same time also the MSCS was higher during NAF and SREG indicating higher scat-

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tering efficiency of PM. Similar dependence of the MAC with atmospheric scenarios was reported by Pandolfi et al. (2011) for Montseny station. Exception was observed for the MAC calculated at Montseny during WREG which was the highest compared to AA, SREG and NAF. The likely reason for the different MAC at MSC and MSY under WREG was the lower altitude of MSY station which was often within the polluted PBL under WREG winter scenarios (i.e. Pandolfi et al., 2014) whereas the MSC was above. As a consequence of the observed variations of MAC and MSCS as a function of the four considered season-dependent scenarios, the MAC and MSCS at MSC showed a clear annual cycle with the lowest values observed in winter and the highest in summer (Figure 8). Similar seasonal dependence of the MAC with higher values in summer was observed at the Jungfraujoch high alpine site (Cozic et al., 2008)."

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

The sentence was has been modified as follows:

"The optical measurements performed at the MSC remote site were compared with those simultaneously performed at a regional background station in the Western Mediterranean Basin located at around 700 m a.s.l. upstream the MSC station."

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

The format of the references has been changed.

13) 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.

The sentence has been modified as follows:

"Specific aerosol optical parameters such as single scattering albedo (SSA), scattering Ångström exponent (SAE), backscatter-to-scatter ratio (B/S) and asymmetry param-

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eter (g), among others, can be derived from these optical properties. Moreover, the mass scattering cross section (MSCS) and the mass absorption cross section (MAC) of sampled aerosols can be obtained from independent measurements of particulate matter (PM) and elemental carbon (EC) concentrations, respectively, and the measured aerosol scattering and absorption coefficients."

14) 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.

The sentence has been modified as follows:

"Most particles scatter the sun light causing a net cooling at the top of the atmosphere, whereas black carbon (BC) particles (or EC) absorb radiation over the entire visible spectrum thus affecting the vertical heating profile and causing a net warming of the Earth-atmosphere system (IPCC, 2007; IPCC, 2013)."

15) 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.

We agree with the reviewer and "measured" has been removed from the sentence.

16) 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.

The sentence was modified as follows:

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“However, lidars do not measure key climate variables such as aerosol absorption, SSA, MSCS or MAC.”

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

The Delene and Ogren (2002) paper was removed from the Introduction.

18) Line 3-4 ‘Andrews et al., 2004’ is an aircraft in-situ measurements paper.

The reference to the paper “Andrews et al., 2004” was removed from the Introduction.

19) Line 7 ‘ high altitude aerosol optical measurements’ is there a citation/webpage for this? To our knowledge no citation/webpage is available.

20) Page 6 Line 10 ‘Particles’ CHANGE TO ‘Particle Done

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

The following sentence was modified accordingly to the reviewer comment:

“Particle scattering (s_sp; 10–171°) and hemispheric backscattering (s_bsp: 90–171°) coefficients at three wavelengths (450nm, 525nm, 635nm) were measured with a LED-based integrating nephelometer (model Aurora 3000, ECOTECH Pty, Ltd, Knoxfield, Australia). The ïĄşsp and ïĄşbsp data were corrected for truncation errors, allowing reporting scattering for 0-360° and backscattering for 90-270°, and for non-ideal (non-Lambertian) illumination function of the light source as described by Müller et al. (2011a).”

22) 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).

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Done

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

Done

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

Done

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

Done

26) 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.

The sentence has been added

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

The sentence has been modified accordingly to the reviewer's suggestion as follows:

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"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 calculated from hourly-averaged scattering (635 nm), hemispheric backscattering (635 nm) and absorption above 0.4 Mm-1, 0.4 Mm-1 and 0.6 Mm-1, respectively."

28) 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.

Given that the seasonal plots are presented in Figure 4, we prefer removing Figure 2 from the text. Moreover, the section 3 has been re-arranged as follows

3.1 General features (Figure 2 was removed; see also reviewer comment #5); 3.2 Diurnal cycles and cluster analysis (Here we present the diurnal cycles and cluster analysis) 3.3.1 Identification of FT air (Here we report the diurnal cycles of normalized scattering and meteorological parameters; see also reviewer comments #6 and #7) 3.3.2 FT vs. all data: Comparison with mountaintop sites presented in AND2011 (Here we discuss the seasonal variation of aerosol optical extensive and intensive optical properties and compare MSC with AND2011).

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

Done

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

Done

31) P12 Line 1-3 'In Italy (Monte Cimone, 2200ma.s.l.) Andrews et al. (2011) reported mean s_sp and s_ap of around 11Mm-1and 3Mm-1, respectively.' There are detailed papers for Monte Cimone discussing dust and other sources. For example: Cristo-

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fanelli 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.

The following sentence was added: “It should be taken into account that in some cases the comparison may be affected by the different size cut-off of the particles sampled at the different sites.”

The sentence has been modified as follows: “Monte Cimone (Italy, ∼2200 m a.s.l.; whole air sampled) registered mean s_sp and s_ap of around 11 Mm-1 and 3 Mm-1, respectively, and both parameters increased highly during Saharan dust outbreaks and wildïňĄre emissions over North Africa (Andrews et al., 2011; Cristofanelli et al., 2004).”

The following sentence (Section 3.2) has been modified: “Similar diurnal variations at other mountain top sites have been observed for extensive aerosol optical properties (e.g. Andrews et al., 2011) and physical properties (e.g. Venzac et al., 2009; Marinoni et al., 2008).”

32) 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’

Done

33) 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).’

Done

34) 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 preva-

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lence 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?

We agree with the reviewer that the differences in frequency and intensity of Saharan dust outbreaks between MSC and JFJ are probably rather small. Recently, Pey et al. (2013; Pey, J., Querol, X., Alastuey, A., Forastiere, F., and Stafoggia, M.: African dust outbreaks over the Mediterranean Basin during 2001–2011: PM10 concentrations, phenomenology and trends, and its relation with synoptic and mesoscale meteorology, Atmos. Chem. Phys., 13, 1395-1410, doi:10.5194/acp-13-1395-2013, 2013) showed that the mean annual contribution of Saharan dust at MSY station was around 1.5 ugm-3 in PM10 which is only slightly higher compared to the mean dust contribution reported by Collaud Coen et al. (2004; M. Collaud Coen, E. Weingartner, D. Schaub, C. Hueglin, C. Corrigan, S. Henning, M. Schwikowski, and U. Baltensperger.: Saharan dust events at the Jungfraujoch: detection by wavelength dependence of the single scattering albedo and ïňĄrst climatology analysis, Atmos. Chem. Phys., 4, 2465–2480, 2004) for JFJ around 0.8 ugm-3 in TSP. Consequently, the sentence was modified as follows:

"Fierz-Schmidhauser et al. (2010) reported mean B/S, SAE and SSA for dry aerosols (RH<20%) within the ranges 0.128-0.122, 1.787-1.671 and 0.91-0.93, respectively. The higher B/S measured at MSC was likely due to the lower size range measured at MSC (PM2.5) compared with Jungfraujoch (whole air) whereas the lower SAE at MSC, despite the different size cut-off, may be an indication of the prevalence of slightly coarser aerosols at MSC. This difference was likely due to the fact that more large par-

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ticles had time to deposit out or be removed via wet scavenging since the altitude of Jungfraujoch is about twice the MSC altitude."

35) 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?

The mean B/S, SAE and SSA measured at MSY and reported in this manuscript were taken from Pandolfi et al. (2011) where they reported data collected during the year 2010. To avoid different measuring periods affecting the comparison between MSC and MSY we report in this revised version of the manuscript the mean values for MSY averaged over the same sampling period of MSC station. On the base of the new values calculated for MSY station we conclude that SAE at MSY and MSC is similar (difference of 5% with SAE at MSY slightly higher compared with MSC). The sentence was modified as follows.

"At the MSY regional background station the mean B/S (525 nm), SAE (450-635 nm), SSA (635 nm) and g (525 nm) were different by around -16%, +5%, -4% and +6%, respectively, compared to MSC (only contemporary data were used). These differences were small suggesting, on average, similarity in microphysical aerosol properties measured at MSC and MSY stations. Recently, Ripoll et al. (2014) have shown that the mean chemical composition of particles at MSC and MSY is on average similar mainly due to the frequency of specific meteorological episodes affecting aerosol properties similarly at both sites. The main difference was observed for B/S which was higher at MSC likely because the lower size cut-off at MSC (PM2.5) compared to MSY (PM10). Moreover, the relatively lower SSA and higher SAE at MSY, despite the differences in the size cut-off, suggested the presence of relatively smaller and more absorbing particles at regional level likely because the proximity of MSY station to anthropogenic sources."

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

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3.5)'

Done

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

Done

38) Line 7-11 rewrite for better grammar

The sentence was modified as follows:

"The absolute values of extensive optical properties and PM mass concentrations and their diurnal cycle amplitudes were higher at MSY site compared to MSC (Fig.2) mainly because the proximity of MSY to the highly urbanized/industrialized coastline (Fig.1) and its lower altitude compared to MSC. As a consequence, the SSA and SAE at MSY station also showed marked diurnal cycles compared to MSC due to the effectiveness of thermally-driven up-slope winds in transporting fine highly absorbing particles of anthropogenic origin to MSY (Fig.2; cf. Pandolfi et al., 2014)."

39) 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:'

The sentence has been accordingly modified as follows:

"As a consequence, the SSA and SAE at MSY station also showed marked diurnal cycles compared to MSC due to the effectiveness of thermally-driven up-slope winds in transporting fine highly absorbing particles of anthropogenic origin to MSY (Fig.2; cf. Pandolfi et al., 2014)."

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

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synoptic scenarios'

Done

41) Line 19 'Similar pattern' CHANGE TO 'A similar pattern'

Done

42) Line 27 spelling antyciclonic' CHANGE TO 'anticyclonic'

Done

43) 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'

Done

44) Line 16-17 'are related with the location' CHANGE TO 'are related to the location"

Done

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

Done

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

Done

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

Done

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

Done

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

Done

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

Done

51) 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. . ..."

Done

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

The sentence was modified as follows:

"The observed decrease of SAE was due to the prevalence of dust (i.e. large, primarily scattering) particles at high aerosol load at MSC."

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

Done

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

The SAE values were calculated when the hourly averages of scattering at 635 nm were above 0.4 Mm-1. The mean (±st.dev.) scattering at 635 nm during AA was

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11.7±13.9 Mm-1 (This average value has been added to the text).

We present below the scatter plots (Figure B) with SAE calculated when scattering@635nm > 0.4 Mm-1 (Left; as presented in the manuscript) and with SAE calculated when scattering@525 nm > 1 Mm-1 (right; as suggested by the reviewer).

As reported in the Figure B the mean (+/- st.dev.) SAE calculated when Scat525nm > 1 Mm-1 (150 values were higher than 4) is very similar to the mean SAE reported in the manuscript when Scat635nm > 0.4 Mm-1 (135 values were higher than 4).

Given that the two criteria lead to similar results, we would like to keep our constraint (i.e. Scat635nm > 0.4 Mm-1) without modifying related text, Figures and Table.

55) a) Page 18 Line 8-10 'However, under very low PM1 concentrations at MSC (PM1 < 1.5 ugm-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).?

The Figure C below shows the systematic relationship plots for SSA and g (by using all data) calculated with the constraint that Scat635nm > 0.4 Mm-1 (above; as in the manuscript) and with the constraint that Scat525nm > 1 Mm-1 (below; as suggested by the reviewer). As already pointed out in the previous reviewer's point (#54) the two constraints lead to very similar results, as shown in the Figure C. Thus, we would like to keep our comment on SSA and g under low PM1 concentrations (PM1 < 1.5 ugm-3).

b) Also where is the plot of PM1 vs SSA and g?

The relationship plots were presented taking the scattering as reference for the calculation of the frequency counts. Thus, the increasing scattering is compared with PM1, SSA and g. Given that PM1 increased fairly monotonically with increasing scattering we would like to keep the scattering as reference for this analysis, without adding other plots We report below (Figure D) the analysis requested by the reviewer.

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c) What are the PM10 values that go along with these very low PM1 values?

As reported in the Figure E below the PM10 concentrations were the lowest when scattering and PM1 were the lowest.

Given that the optical properties reported in this manuscript refer to PM2.5 fraction we would like to avoid reference to the PM10 fraction in Figure 6.

56) 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'

Done

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

Done

58) Line 28 '. . .. . .NAF and SREG,. . .. . .' CHANGE TO '. . .. . .NAF and SREG. ' and Page 19 Line 1 ". . .. . ..compared with those obtained for AA and WREG. Conversely, no. . .. . . :' CHANGE TO 'Conversely, no . . ..'

The sentence was modified as follows (see also reviewer comment #10):

"The difference between the NAF and SREG medians was not statistically significant ($p>0.5$) for both MAC and MSCS. The same was observed for the AA and WREG medians ($p>0.3$). Conversely, statistically significant differences ($p<0.001$) were observed between the medians calculated for WREG and AA and those calculated for NAF and SREG. The higher MAC and MSCS under NAF and SREG compared to AA and WREG were likely due to differences in particles origin and particle properties during these scenarios."

59) 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

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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).

The sentence was removed.

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

Done

61) 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.

Please, see reply to reviewer comments #6 and #7 above.

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

Done

63) 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.

The sentence was modified as follows:

"Under Atlantic Advection (AA) scenarios the lowest scattering and absorption were measured at MSC. The AA scenario in the WMB is typically characterized by high wind speed with air masses coming from the Atlantic Ocean (Pandolfi et al., 2014) thus favouring the dispersion of the accumulated pollution with consequent reduction of the concentrations of pollutants."

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64) 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?

The sentence was modified as follows:

"However, the relative decreases in FT-data extensive properties were clearly seasonal at the MSC site with the highest FT vs. all-data difference observed in winter (DJF in Figs.5a,b,c; 21-23%) and the lowest in spring/summer (MAM and JJA in Figs.5a,b,c; 0-8%). The differences between the FT-data and all-data medians were statistically significant for ALL, SON and DJF and marginally significant for JJA. As a consequence of the time of day segregation used for the identification of FT air (3:00–09:00 am vs. 24h) the remote sites showing stronger diurnal cycles tend to have larger decreases in FT aerosol loading compared to all-data (24h) aerosol loading (Andrews et al., 2011). In our case, the high FT vs. all-data difference observed in winter is consistent with the strong DJF diurnal cycle of normalized scattering presented in Fig.4. The lower FT vs. all-data difference in spring/summer was due to the occurrence of NAF and SREG episodes and to the fact that FT conditions were less frequent in spring/summer due to higher boundary layer."

65) 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.

The Table 1 caption has been modified. The acronyms RB and DC were removed from the Table 1 and from the text. The acronym WMB is extensively used throughout the text and we prefer do not remove this acronym.

66) Figure 2 – delete? Other than showing the time series, I don't think it's discussed

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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 2 was deleted. Please, see the reply to the reviewer comment #28.

67) 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.

Figure 6 was modified to show the systematic relationships as a function of airmass type. The number of points in each scattering bin was added (see also reviewer comment #9).

FIGURE CAPTIONS:

Figure A: Mixing Layer Height (MLH) calculated in Barcelona at 12:00 UTC from radiosoundings for the period 2003-2010. Median, 25th and 75th percentiles and minimum and maximum values are reported. Dashed line indicates the Montsec altitude.

Figure 1: Location of the Montsec (MSC; remote-mountaintop site) and Montseny (MSY; regional background) measurement sites. Barcelona is also shown. Yellow dots are meteorological stations (Observatory (800 m a.s.l.), Os de Balaguer (576 m) and Vallfogona de Balaguer (238 m)). Air mass backtrajectories from Atlantic Ocean (AA), regional (REG) and North Africa (NAF).

Figure 3: Seasonal diurnal cycles of relative humidity, potential temperature and water vapour mixing ratio measured at Montsec (1570 m a.s.l.), Montsec Observatory (800 m), Os de Balaguer (576 m) and Vallfogona de Balaguer (238 m). Also shown are the diurnal cycles of the planetary boundary layer height (PBL) from HYSPLIT and of the potential temperature and actual temperature gradients. Yellow rectangles highlight the time of the day approach for the identification of FT air proposed in Andrews et al.

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(2011) and used in this work (from 3:00 to 9:00 local time). Grey rectangles highlight hours when the MSC station was within the planetary boundary layer. Meteorological data at the 4 stations were available from 1th Jan 2011 to 31 Dec 2012.

Figure 5: Aerosol optical properties for all-data and FT-data data. Data are reported at 550 nm. Red=all-data, Yellow=FT-data. Horizontal lines within the boxes are the medians (50th percentile), edges of box are 25th and 75th percentiles, and whiskers are 5th and 95th percentiles. Ångström exponent is calculated for 450/635 nm pair. For MSC values are calculated for the whole period considered here (ALL), and for fall (SON), winter (DJF), spring (MAM) and summer (JJA). The percentage values represent the relative difference between the medians calculated for all-data and FT-data. Green bold numbers indicate statistically significant differences (p-value<0.05); blue bold numbers highlight marginally significant differences (p<0.1); black numbers indicate differences which were not statistically significant (p>0.1). The red and yellow rectangles within the blue areas on the right of each figure represent the range of variability of the medians presented by Andrews et al. (2011) calculated for sites in the western hemisphere (W), Europe (EU) and eastern hemisphere (E).

Figure 6: Correlation between the frequency distribution of aerosol scattering coefficients (ïĄşsp) at 635 nm and backscattering coefficient (ïĄşbsp at 635 nm), absorption coefficient (ïĄşap at 637 nm), PM1 concentrations (PM1), asymmetry parameter (g at 635 nm), scattering Ångström exponent (SAE; 450-635 nm), single scattering albedo (SSA at 635 nm). Correlations are presented for all data (ALL) and for the different atmospheric scenarios (Atlantic Advection, AA; Saharan dust outbreaks, NAF; summer regional recirculation scenario, SREG; and winter anticyclonic scenario, WREG). Frequency count (%) and the absolute number of hourly values (counts) in each been are reported.

Figure 8: Monthly mean mass absorption cross section (MAC) and mass scattering cross section (MSCS) at MSC station and occurrence (%) of the main atmospheric scenarios (AA: Atlantic advections; NAF: Saharan dust outbreaks; SREG: summer

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regional recirculation scenarios; WREG: winter anticyclonic scenarios). Bars represent 95 % conïňĄdence intervals.

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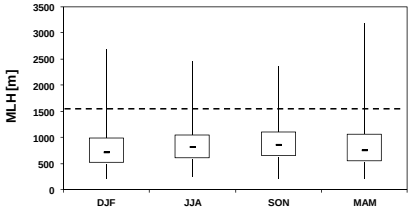

Figure A

Fig. 1. FIGURE A

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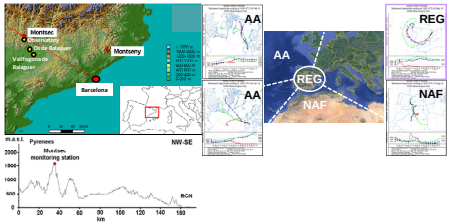

Figure 1

Fig. 2. FIGURE 1

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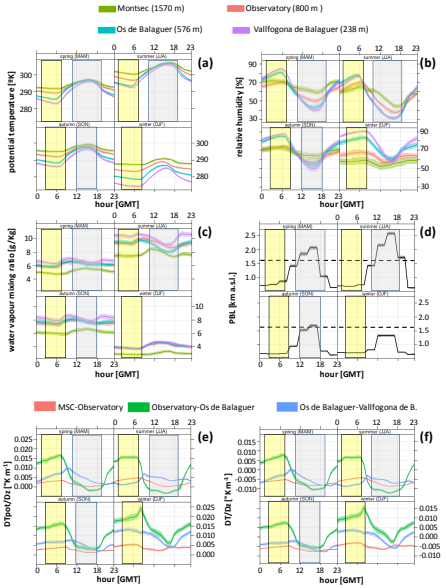

Figure 3

Fig. 3. FIGURE 3

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Interactive Discussion
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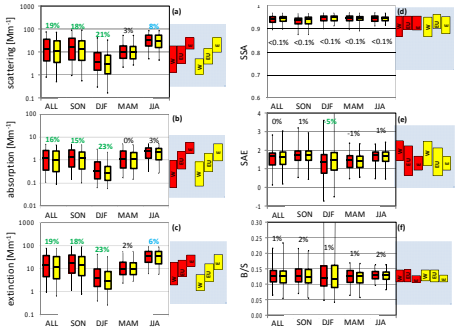

Figure 5

Fig. 4. FIGURE 5

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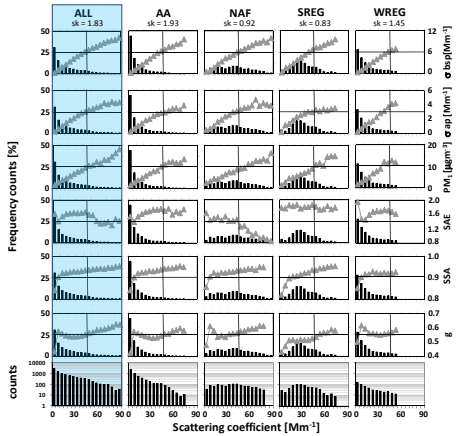

Figure 6

Fig. 5. FIGURE 6

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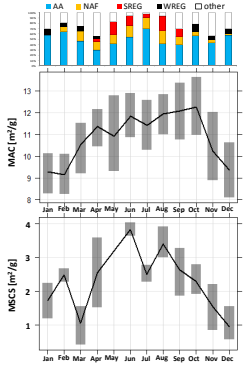

Figure 8

Fig. 6. FIGURE 8

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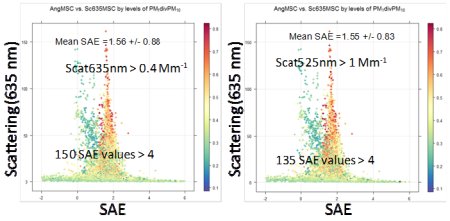

Figure B

Fig. 7. FIGURE B

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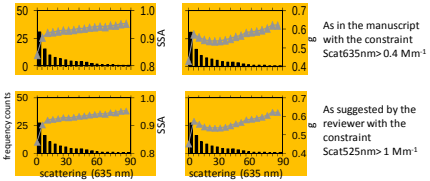

Figure C

Fig. 8. FIGURE C

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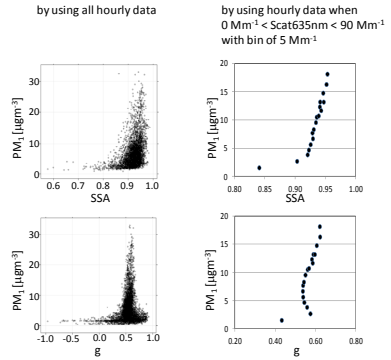

Figure D

Fig. 9. FIGURE D

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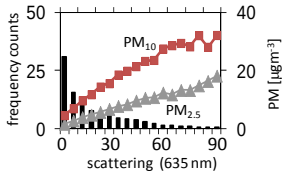

Figure E

Fig. 10. FIGURE E

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