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Comment

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.

M. Pandolfi et al.

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We thank the reviewer#1 for the useful comments on our paper.

Anonymous Referee #1

“General comments”.

1) The manuscript presents two-years in-situ measurements of aerosol optical properties at a European mountaintop observatory in the western Mediterranean Basin,

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run according to ACTRIS standards. This is a unique and novel dataset that is worth considering. Data are compared to a regional background station run according to ACTRIS standards. Extrapolated data of in-situ aerosol properties in the free troposphere are compared to other mountaintop sites. Measurements show the site to be in the medium/upper range of values measured at other similar European sites. African dust and regional recirculation during the warmer seasons are found to be the major sources of the relatively high values observed, indicating the potential impact of these two sources in the whole lower troposphere of the western Mediterranean. This is a substantial issue addressed by the manuscript. Despite that, I do believe that the manuscript still deserves a deeper analysis to fully reach the scope of the work, and substantially improve conclusions.

Abstract and conclusions have been improved. Both are reported below:

“ABSTRACT Aerosol light scattering (s_{sp}), backscattering (s_{bsp}) and absorption (s_{ap}) were measured at Montsec (MSC; 42°3’N, 0°44’E, 1570 m a.s.l.), a remote high-altitude site in the Western Mediterranean Basin. Mean (\pm sd) s_{sp} , s_{bsp} and s_{ap} were 18.9 ± 20.8 Mm⁻¹, 2.6 ± 2.8 Mm⁻¹ and 1.5 ± 1.4 Mm⁻¹, respectively at 635 nm during the period under study (06/2011–06/2013). Mean values of single scattering albedo (SSA, 635 nm), scattering Ångström exponent (SAE, 450–635 nm), backscatter-to-scatter ratio (B/S, 635 nm), asymmetry parameter (g, 635 nm), black carbon mass absorption cross section (MAC, 637 nm) and PM_{2.5} mass scattering cross section (MSCS, 635 nm) were 0.92 ± 0.03 , 1.56 ± 0.88 , 0.16 ± 0.09 , 0.53 ± 0.16 , 10.9 ± 3.5 m²/g and 2.5 ± 1.3 m²/g respectively. The scattering measurements performed at MSC were in the medium/upper range of values reported by Andrews et al. (2011) for other mountaintop sites in Europe due to the frequent regional recirculation scenarios (SREG) and Saharan dust episodes (NAF) occurring mostly in spring/summer and causing the presence of polluted layers at the MSC altitude. However, the development of up-slope winds and the possible presence of planetary boundary layer air at MSC altitude in summer may also have contributed to the high scattering observed.

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Under these summer conditions no clear diurnal cycles were observed for the measured extensive aerosol optical properties (s_{sp} , s_{bsp} and s_{ap}). Conversely, low s_{sp} and s_{ap} at MSC were measured during Atlantic advections (AA) and winter regional anticyclonic episodes (WREG) typically observed during the cold season in the Western Mediterranean. Therefore, a season-dependent decrease in the magnitude of aerosol extensive properties was observed when MSC was in the free troposphere with the highest free-troposphere vs all-data difference observed in winter and the lowest in spring/summer. The location of MSC station allowed a reliable characterization of aerosols as a function the main synoptic meteorological patterns. The SAE was the lowest during NAF and showed an inverse correlation with the outbreaks intensity indicating a progressive shift toward larger particles. Moreover, the strength of NAF episodes in the region led to a slope of the scattering vs absorption relationship among the lowest reported for other mountain top sites worldwide indicating that MSC was dominated by dust aerosols at high aerosol loading. As a consequence, SSA showed a nearly monotonic increase with increasing particle concentration and scattering. The SAE was the highest during SREG indicating the presence of polluted layers dominated by smaller particles. Correspondingly, the asymmetry parameter was lower under SREG compared with NAF. The MAC and MSCS were significantly higher during NAF and SREG compared to AA and WREG indicating an increase of absorption and scattering efficiencies associated with the summer polluted scenarios. 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.”

“Conclusions The measurements of aerosol optical properties presented in this work and performed at Montsec remote site (MSC; 42°3'N, 0°44'E, 1570 m a.s.l.) add useful information on the limited amount of in-situ aerosol optical data obtained at high altitude/mountaintop sites worldwide. The aerosol scattering measurements performed at MSC located this site in the medium/upper range of values reported for other mountaintop sites in Europe (EU). The frequent African dust (NAF) outbreaks and regional

recirculation (SREG) scenarios, typical of the WMB in spring/summer, were mainly responsible for these relatively high values. Moreover, the lower altitude of MSC station compared with other mountaintop sites and the strong summer insolation in the Mediterranean regions, favoring the development of thermally-driven up-slope winds, may have also contributed to the relatively higher scattering observed at MSC. The mean scattering at MSC during the NAF and SREG scenarios were close to the values measured at a regional background station (Montseny; 720 m a.s.l.) thus demonstrating the potential of these two summer atmospheric scenarios in polluting the whole lower troposphere in the Western Mediterranean. As a consequence, in spring and summer no clear diurnal cycles were observed for the extensive aerosol optical properties due to the presence of polluted layer at the MSC altitude. Thus, the diurnal variation of scattering at MSC during spring and summer was subject to synoptic circulation which masked in part the mountain breezes and the dynamics transport at a more local scale. Conversely, during Atlantic advection (AA) and winter regional anticyclonic (WREG) episodes, mainly registered during the cold season, the extensive aerosol properties at MSC were considerably lower compared to Montseny and the highest diurnal cycle amplitudes were observed. The AA scenario in the WMB is typically characterized by high wind speed with air masses coming from the Atlantic Ocean thus favouring the dispersion of the accumulated pollution with consequent reduction of the concentrations of pollutants which is more effective at remote level. The WREG scenario is mainly characterized by weak synoptic winds leading to stagnation of air masses and to the accumulation and aging of pollutants over the region, whereas the PBL height mostly determines the dilution of pollutants around the emission sources and the degree of pollution at more elevated/regional areas in the WMB. Absorption at MSC was not as high as scattering compared with most of measurements in EU thus leading to relatively higher single scattering albedo (SSA) compared with EU data. Conversely, the scattering Angstrom exponent (SAE) and backscatter-to-scatter ratios (B/S) were in the middle of the corresponding EU ranges. All the extensive aerosol properties measured at MSC showed relatively lower medians when MSC was in the

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free troposphere (FT-data) compared with the whole database (all-data). These decreases were clearly seasonal at MSC site with the highest and statistically significant FT vs. all-data difference observed in winter and the lowest (and not statistically significant) in spring/summer. The frequent NAF and SREG scenarios in summer, and the less frequent FT conditions due to higher boundary layer, explained the lower FT vs. all-data differences observed in summer compared to winter. The aerosol optical measurements performed demonstrated that the MSC measurement site provides reliable information for a suitable characterization of the main synoptic meteorological patterns affecting the region. Clear differences were observed between NAF and SREG scenarios in terms of intensive aerosol optical properties. SAE during NAF was the lowest, indicating presence of larger particles, and was clearly anticorrelated with the intensity of Saharan dust outbreaks, whereas nearly constant and higher SAE was measured under SREG indicating an aerosol mode dominated by finer particles. Correspondingly, the asymmetry parameter was higher during NAF compared to SREG. The analysis of the relationships between scattering and other extensive/intensive aerosol properties measured at MSC showed a scattering-absorption slope in the lower range of slopes calculated worldwide indicating that the MSC site is dominated by dust aerosols at high aerosol loading. As a consequence, SSA increased nearly monotonically with increasing scattering. The MAC estimated at MSC showed a clear annual cycle with higher values in summer when the occurrence of NAF and SREG scenarios favoured the presence of polluted atmospheric layers containing aged BC particles likely mixed with other chemical components such as organics and sulfate. These summer conditions were also linked with higher scattering efficiency of PM.”

2) Additional relevant scientific questions should be addressed: in particular, an important point is that concerning absorption properties and Black Carbon sources that are merely presented, but barely discussed.

Following the reviewer’s suggestion we have improved the section (section 3.5) related with BC and mass absorption cross section (MAC). We provided information on pos-

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sible origin of BC particles at Montsec and reasons for the observed annual cycle of MAC. Please, note also that a paper has been recently published (Ripoll et al., 2014) and another one has been recently submitted (Ripoll et al., submitted) describing in detail the PM chemical composition and possible sources at MSC. The new section 3.5 and new Figure 8 are reported below:

“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 ($R^2=0.82$). Given that s_{ap} 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; $R^2=0.77$) and WREG (9.4 ± 1.0 m²/g; $R^2=0.88$) scenarios compared to NAF (11.9 ± 0.7 m²/g; $R^2=0.61$) and SREG (12.6 ± 1.0 m²/g; $R^2=0.74$) 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 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

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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 scattering 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).

I therefore recommend publication of the manuscript after taking into consideration (at least) the following specific comments.

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3) Pag. 3790, line 24: Data measured were scaled to a wavelength of 550 nm by using 1 as AAE (Absorption Angstrom Exponent). The AAE=1 value represents an aerosol with optical properties of pure Black Carbon: that means that the absorption is totally dominated by Black Carbon, and that there are no additional absorbing compounds, such as Brown Carbon or dust. Both brown carbon and dust can increase AAE, indeed. AAE also varies with aerosol size. By using AAE=1, authors constrain results. In fact, AAE might be much larger than 1, the absorption coefficient at 550 nm resulting larger than the value calculated here. The comparison to other similar sites (Fig.4) may result in different conclusions. Authors should discuss the point.

We agree with the reviewer that an AAE of 1 could not represent properly the dependence of absorption with wavelengths at MSC. Only very recently (1 month ago) we have installed an Aethalometer (model AE33, Magee Scientific) at MSC. In the revised version of the manuscript we used the AAE calculated from the available 1-month 7-lambda absorption measurements. The experimental AAE was 1.4. This new AAE was used to calculate the absorption at 550 nm used in Figure 5 of the revised version of the manuscript.

The following sentence was added:

“MSC data were scaled to a wavelength of 550 nm (used in AND2011) by using 1.6 as SAE (median values for MSC; this work) and 1.4 as absorption Angstrom exponent (AAE). The AAE was calculated from 1 month absorption measurements performed at MSC with a 7 wavelengths Aethalometer (model AE33, Magee Scientific).”

The use of an AAE of 1.4 instead of 1.0 changed the calculated absorption, extinction and SSA. However, the differences were small and the conclusions related with the comparison with the other sites did not change substantially.

The new values of absorption, extinction and SSA obtained by using AAE=1.4 have been reported in the revised version of the manuscript.

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4) Pag. 3794, line 4-17: Authors observe conditions of low PM1 concentrations coupled to low Single Scattering Albedo and low g . They associate these conditions to small particles with higher absorption properties, similarly to AND2011 associating low SSA at very low aerosol loadings with an aerosol mixture in which large scattering particles are removed and relatively smaller and darker aerosol left. This is an interesting point, that may be carefully analyzed. It would be interesting to speculate on possible sources causing these aerosol properties at the mountaintop observatory examined here. These sources of Black Carbon may be discussed after due consideration of their coupling to African dust and summer regional recirculation (pag.3797, lines 4-6). The question of possible Brown Carbon sources (e.g., associated to summer regional recirculation) increasing AAE (see previous comment) may also be added. This has the potential to improve the understanding of polluting scenarios affecting the whole lower troposphere of the western Mediterranean.

Following the suggestions of the Reviewers we have modified Figure 6 in order to present the systematic relationship plots as a function of air mass type. As shown in the new Figure 6 (reported below), the conditions of low PM1 coupled to low SSA and g were always observed irrespective of the scenarios. At the same time, the new Figure 6 shows that the calculated parameters (and mainly SAE and g) are strongly a function of the different air mass types considered. Thus, the observed very low SSA and g associated with very low PM1 (irrespective of the scenarios) was likely due to processes which do not depend on air mass type or a specific source of BC. Thus, it seems probably due to processes, such as deposition or cloud scavenging, which result in the presence of smaller and darker aerosols. Concerning the possible BC sources refer to the reviewer's comment #2.

The corresponding test is reported below: "However, under very low PM1 concentrations at MSC ($PM_{1<1.5} < 3 \mu g m^{-3}$) SSA and g reached very low values around 0.84 and 0.43 (ALL in Fig. 6), respectively, whereas the SAE increased. These low PM conditions at MSC were associated with the prevalence of small particles with rela-

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tively higher absorption properties irrespective of the scenario considered. Low values of SSA at very low aerosol loading have been observed in AND2011 and associated with an aerosol mixture in which large scattering aerosol particles have been preferentially removed (e.g., by cloud scavenging and/or deposition), leaving behind a relatively smaller and darker aerosol (e.g., AND2011; Berkowitz et al., 2011; Marcq et al., 2010; Targino et al., 2005; Sellegrì et al., 2003).”

5) The paragraph 3.1 merely presents data, with no or scarce discussion/interpretation. In particular, absorption properties should be discussed with more attention. In some parts of this paragraph, text can be reduced or eliminated (e.g., pag. 3787, lines 2-12).

The first part of the section 3.1 has been modified in order to eliminate unnecessary parts of the text:

“Table 2 reports the statistics for the measured parameters, including means, standard deviations, percentiles (1, 10, 25, 50, 75, 99 percentiles), minima and maxima values and skewness. All parameters showed skewness higher than 1 with the exception of SAE, MSCS and MAC, for which almost normal distributions (skewness close to zero) were observed, and SSA and g both showing a negative skewness. Similar SSA skewness was presented from Pandolfi et al. (2011) at the MSY regional background measurement site. Negative skewness for g is a consequence of the high positive skewness observed for B/S.”

6) A table could summarize comparison with other similar studies (pag. 3787, from line 23). It would be nice to include in a separate paragraph this table and all text comparing to previous related work, with a clearer indication of the original contribution of this work.

The comparison with measurements reported at other mountain top sites was presented and commented in the section comparing Montsec data with the data provided by Andrews et al. (2011) for 10 mountain top sites. Moreover, as suggested by the reviewer, abstract and conclusions have been improved in order to clearly indicate the

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original contribution of this work. Thus, if the reviewer also agrees, we would prefer not to duplicate this information already given in the manuscript.

7) Pag.3791, line 25: It is stated that there is a relative decrease in FT-data extensive properties, clearly being a function of seasons. However, either fig.4 can't show this decrease because box plots are too small, or this is not correct. Please, comment on that. Reasoning for seasonal difference of the FT vs all-data extensive properties (other than scattering seasonal DC in the fig.5) should also be given.

It is certain difficult to clearly appreciate the relative decrease in FT-data extensive properties only looking at the box plots of Figure 5. For this reason we added to the Figure the percentage values representing the relative differences between the medians calculated for all-data and FT-data. In order to further help the reader, we have coloured the percentage values with different colours depending on the statistical significance of the calculated differences. In the new Figure 5 (reported below), 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 following sentence has 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.”

In order to clarify the reasoning behind the seasonal difference of the FT vs all-data extensive properties, we have carefully studied when MSC station experiences FT air or PBL air. With this aim, we used contemporary meteorological data collected at MSC station (1570 m a.s.l.), at the Montsec Observatory (800 m), at Os de Balaguer (576 m) and Vallfogona de Balaguer (238 m) to study the strength of the nocturnal and diurnal

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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) shows the results of this analysis: Consequently, the following sentence was added to the new section 3.3.1 Identification of FT air

“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 by 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 time of the day approach (from 3:00 to 9:00 local time) for the identification of FT air

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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. Thus, the seasonal difference between FT and all-data medians was due to both the frequent NAF and SREG scenarios in summer, which led to aerosol layers at MSC altitude during the whole day, and to the possible presence of polluted residual layers at night during spring and summer."

Different parts of the manuscript were accordingly changed to take into account this result.

"Technical corrections".

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8) The abstract seems to be ineffective in explaining the quality of the manuscript. I thus suggest to improve it. Most importantly, specific conclusions should be added.

Please, refer to the reviewer's comment #1.

9) Correct WAE with WREG in tab.1.

Done

10) Add the site representativeness in tab.1 for MSC and MSY.

The corresponding part of Table 1 was modified as follows: MSC: Montsec (NE Spain; mountaintop measurement site) MSY: Montseny (NE Spain; regional background measurement site)

11) Pag.3793, line 16: correct from -2 to 6 with from -2 to 4.

Done

12) Titles of the subparagraphs 3.2 (Diurnal cycles and cluster analysis) and 3.3 (FT vs. all data) might be changed to describe contents of the paragraph in a clearer way.

The section 3 is now organized as follows:

3 RESULTS AND DISCUSSION 3.1 General features 3.2 Diurnal cycles and cluster analysis 3.3 FT conditions at MSC station 3.3.1 Identification of FT air 3.3.2 FT vs. all data: Comparison with mountaintop sites presented in AND2011

FIGURE CAPTIONS:

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

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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. (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 ($p < 0.05$); 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 (\ddot{a}_{ssp}) at 635 nm and backscattering coefficient (\ddot{a}_{bsp} at 635 nm), absorption coefficient (\ddot{a}_{sap} 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).

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

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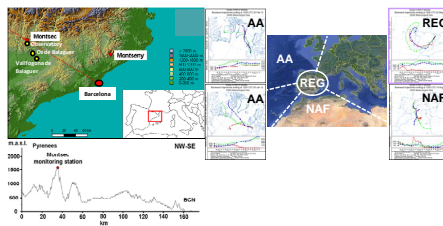


Figure 1

Fig. 1. Figure 1

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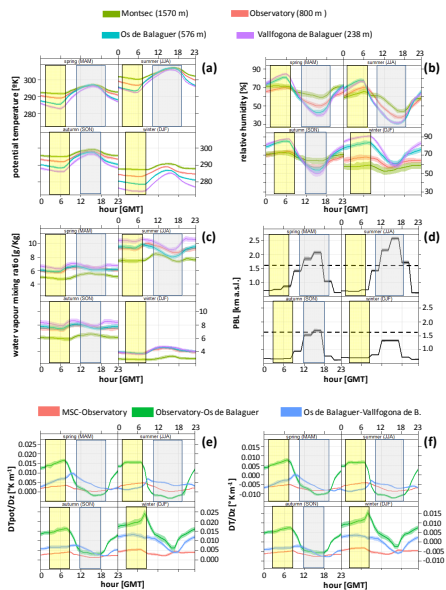


Figure 3

Fig. 2. Figure 3

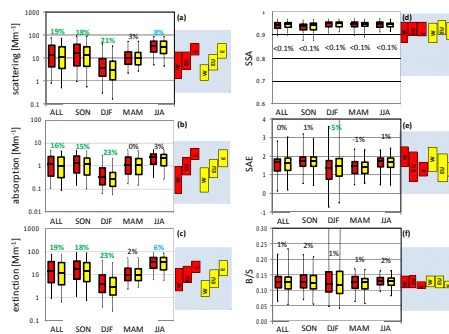
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Figure 5

Fig. 3. Figure 5

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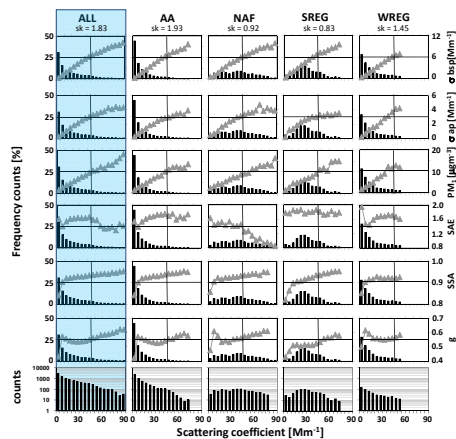
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Figure 6

Fig. 4. Figure 6

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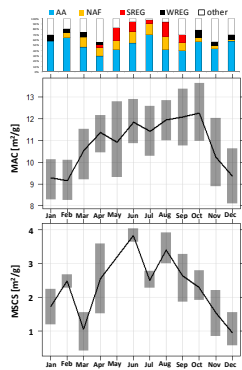


Figure 8

Fig. 5. Figure 8

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