Anonymous Referee #2

1) How SSA and Angstrom exponents were estimated should not be described in the abstract.

The following sentence in the Abstract was deleted.

"Single Scattering Albedo (SSA) at 635 nm was estimated starting from aerosol scattering and absorption measurements, while Ångström exponents were calculated by means of the three wavelengths (450 nm, 525 nm, 635 nm) aerosol light scattering measurements from Nephelometer."

2) MAAP instrument measures the absorption coefficient but no BC.

In order to clarify this point the following sentence (Pag. 14092, lines 1-8):

"Aerosol light scattering, black carbon (BC) and particulate matter (PM) concentrations were measured at Montseny, a regional background site in the Western Mediterranean Basin (WMB) which is part of the European Supersite for Atmospheric Aerosol Research (EUSAAR). Off line analyses of 24h PM filters collected with Hi-Vol instruments were performed for the determination of the main chemical components of PM. Measurements of BC were used to calculate the light absorption properties of atmospheric particles."

Was replaced with:

"Aerosol light scattering, absorption and particulate matter (PM) concentrations were measured at Montseny, a regional background site in the Western Mediterranean Basin (WMB) which is part of the European Supersite for Atmospheric Aerosol Research (EUSAAR). Off line analyses of 24h PM filters collected with Hi-Vol instruments were performed for the determination of the main chemical components of PM."

Thus, "black carbon (BC)" was replaced with "absorption" and the sentence "Measurements of BC were used to calculate the light absorption properties of atmospheric particles." was removed.

3) Page 14095, Line 24 – some references about the correction of the truncation errors should be included.

References have been added and the following sentence (Pag. 14095, Line 24):

"Scattering measurements from nephelometers need to be corrected for truncation errors due to non-ideal detection of scattered radiation."

Was replaced with:

"Scattering measurements from nephelometers need to be corrected for truncation errors due to non-ideal detection of scattered radiation (Anderson and Ogren, 1998; Müller et al., 2011).".

4) From Page 14095, Line 25 to Page 14096, Line 22 – These sentences are very confused to understand the process to the data correction. The presentation should be improved and consider shortening it a bit.

We need to clarify this point for the following reasons:

As we wrote in the manuscript, the correction of scattering data as a consequence of the non-ideal detection of scattered radiation is done by nephelometers users by means of parameterized λ-dependent correction factors ($C_{sp,\lambda}$) provided in dedicated technical papers. Thus, the corrected total scattering data ($\sigma_{sp,\lambda}$) are obtained by applying the following formula: $\sigma_{sp,\lambda} = C_{sp,\lambda} \times \sigma_{uncorr\ sp,\lambda}$, where $\sigma_{uncorr\ sp,\lambda}$ is the uncorrected total scattering measured by the nephelometer.

The users of TSI nephelometer for example can refer to the paper from Anderson and Ogren (1998) where the authors provide correction factors ($C_{sp,\lambda}$) as parameterized functions of the Angstrom exponents calculated starting from the uncorrected total scattering data ($C_{sp,\lambda} = a + b \times Å$; where a and b are constant and Å are the Angstrom exponents).
This Angstrom-based correction scheme is very useful given that the Angstrom exponents can be easily obtained from the uncorrected scattering measurements from nephelometer and than used to calculate the correction factors $C_{sp,\lambda}$.

The technical paper for the Ecotech nephelometer is Muller et al. (2011) [T. Müller, M. Laborde, G. Kassell, and A. Wiedensohler. Design and performance of a three-wavelength LED-based total scatter and backscatter integrating nephelometer. Atmos. Meas. Tech., 4, 1291-1303, 2011] **published in AMT on 29 June 2011 (**about two months after the submission of our paper to ACPD) and providing two correction schemes: one based on the Angstrom exponents (as Anderson and Ogren, 1998) and one based on the volume median diameter of the collected particles.

However, when we submitted our manuscript the paper from Müller et al. was still under discussion. At that time Müller et al. (2010 AMT Discussion) provided only the correction scheme based on the volume median diameter of the collected particle and not the one based on the Angstrom exponent. During our study period the volume median diameter data were not available, thus the parameterization provided by Müller et al. in their paper under discussion (2010 AMT Discussion) was not applicable to our data.

Consequently, we had to find another way to correct our total scattering data collected with the Ecotech nephelometer.

Fortunately, Müller et al. (2010 AMT Discussion) provided a useful comparison between both uncorrected and corrected total scattering data from Ecotech and TSI nephelometers, with the TSI data corrected by means of the Angstrom-based scheme of Anderson and Ogren (1998) and the Ecotech data corrected with the median-diameter-based scheme proposed by Müller et al. (2010 AMT Discussion).

Müller et al. (2010 AMT Discussion) observed very small differences between the both uncorrected and corrected TSI and Ecotech scattering data (between 2% and 5 % depending on the wavelength) and, most importantly, they reported coefficients of determination (R^2) higher than 0.99. Thus, excellent linear relationships were observed between the TSI and Ecotech scattering data (both corrected and uncorrected).

To sum up, the coefficients of these perfect linear regressions ($R^2 > 0.99$) reported in Müller et al (2010 AMT Discussion) can be used to "simulate" Ecotech scattering data starting from TSI scattering data and *vice versa*.

Consequently, in our manuscript we corrected our Ecotech data by applying the following procedure (named *Old procedure* from now on):

a) Firstly, we simulated "uncorrected TSI data" starting from the uncorrected Ecotech data (by using the regression coefficients provided by Müller et al. 2010 AMT Discussion),
b) Then, we corrected the "uncorrected TSI data" by means of the Angstrom-based correction scheme provided by Anderson and Ogren (1998),
c) Finally, we calculated the corrected Ecotech data (used in our manuscript) by using the linear relationship between TSI and Ecotech corrected data provided by Müller et al. 2010 AMT Discussion.

Even if a bit convoluted, this procedure led to a very good correction of the Ecotech scattering data as shown in the following of this text.

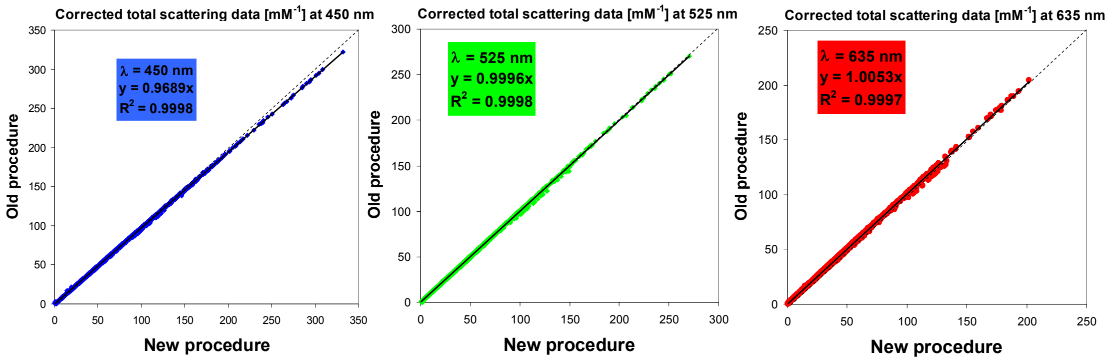
Moving on with the comment, during the open discussion phase of the paper from Müller et al.(2010 AMT Discussion), Mr. Ogren posted the following comment: "*Ecotech neph users are going to be knocking down your doors for a simplified correction scheme for truncation/illumination errors. It would be very useful if you would evaluate the feasibility of the Anderson&Ogren scheme for estimating the correction factors for the Ecotech neph using the measured Ångström exponents. If the approach is feasible, I suggest including the parameters for the correction equations in your paper.*"

In the revised version of the paper from Müller et al., published in AMT on 29 June 2011, the correction factors based on the measured Ångström exponents were provided.

Consequently, we have now applied to our uncorrected total scattering data the Angstrom-based correction scheme (named as *New procedure* from now on) recently published by Muller et al. (2011 AMT) for the Ecothech nephelometer.

The data corrected with the *New procedure* are those presented in this revised version of the manuscript.

We must point out that our *Old procedure* and the *New procedure* are almost equivalent and lead to similar corrections. Thus, neither our interpretation of the results, neither the comments raised by the Anonimous Referee #1 (as well as those from the Anonimous Referee #2 and those from Mr. John Ogren) change after applying the *New procedure*. To demonstrate this, the following Figure shows the comparison between the Ecotech total scattering data corrected with the two procedures (*Old procedure* and *New procedure*):



Differences were estimated in -0.4%, +0.3%, and +3.7% between scattering data corrected with the *New procedure* and *Old procedure* for wavelengths of 635, 525 and 450 nm, respectively. These differences could be due to the fact that in the *Old procedure* we corrected the data as in Anderson and Ogren (1998) without rescaling our data (collected at 450 nm, 525 nm, and 635 nm) to the wavelengths of the TSI nephelometer (collected at 450 nm, 550 nm, and 700 nm).

The excellent correlation observed in the above Figures is a consequence of the almost perfect linear correlation discussed in Müller et al. (2010 AMT Discussion and 2011, AMT) between Ecotech and TSI scattering data.

The observed differences between the *Old procedure* and the *New procedure* lead to differences for SSA and Angstrom exponent of 0.3 % and 9%, respectively.

Consequently, we changed the section of the manuscript where we describe the correction procedure (Section 2.2) taking into account that the Angstrom-based correction scheme for the Ecotech nephelometer is now available (Müller et al. (2011, AMT). Text, Figure and Tables were accordingly changed in this revised version of the manuscript.

Consequently, the following sentence (Pag. 14096, Lines 8-22):

"Müller et al. (2010a) provided the parameterization needed to calculate Csp,λ once the volume median diameter of the particle size distribution is known and compared corrected and non-corrected data from Aurora 3000 to another commercial instrument (TSI model 3563) that was fully described by Anderson et al. (1996) and Anderson and Ogren (1998). Anderson and Ogren (1998) provided a correction factor based on the measured Ångström exponent which is often used for aerosol size characterization. As shown in the next paragraph the Ångström exponents can be easily calculated by means of the multi-wavelength total scattering measurements from nephelometers. After correction, both instruments agreed very well with differences for total scattering between 2% and 5% and coefficients of determination (R2) higher than 0.99

(Müller et al., 2010a). Given that the measurements of the volume median diameter of the particle population was not available, in the present work the data from the Aurora 3000 were corrected by using both the Ångström-based parametrization of Anderson and Ogren (1998) and the experimental comparison provided by Müller et al. (2010a) between Ecotech and TSI nephelometers after correction.",

was replaced with:

"Müller et al. (2011a) provided parameterized correction factors $C_{sp,\lambda}$ as linear relationship of the measured Ångström exponents which are often used for aerosol size characterization and which can be easily calculated by means of the multi-wavelength total scattering measurements from nephelometers. Thus, the correction factors were calculated as $C_{sp,\lambda} = a + b \times å$. The coefficients *a* and *b* at each wavelength were taken from Müller et al. (2011a) while the Ångström coefficients (*å*) were calculated starting from the uncorrected Ecotech scattering data. Similar correction scheme was developed for another commercial nephelometer (TSI model 3563; Anderson et al., 1996) by Anderson and Ogren (1998).".

5) Page 14096, Lines 23-25 – Briefly explain why it is necessary to measure with a RH less than a certain threshold. Some references should be also included.

The heater prevents the effects of hygroscopicity and consequently it prevents the scattering enhancement of particles.
The Aurora 3000 nephelometer provides information on the amount of relative humidity inside the sampling cell. During the study period the mean relative humidity inside the cell was 28% with a standard deviation of 12% (minimum and maximum RH values of 2% and 60% respectively). Thus, we studied dried aerosols and not ambient aerosols.

Consequently, the following sentence:
"In order to prevent the presence of liquid particles inside the sampling cell a relative humidity (RH) threshold of 60% was set by using a processor-controlled automatic heater inside the nephelometer.",

was replaced with:
"In order to prevent the presence of liquid particles inside the sampling cell, and consequently the effects of hygroscopicity enhancing the scattering properties of particles, a relative humidity (RH) threshold of 60% was set by using a processor-controlled automatic heater inside the nephelometer. This experimental procedure was applied elsewhere (see for example Pereira et al., 2011 or Anderson and Ogren, 1998). Thus, during the study period the particles were dried to a mean relative humidity of 28% with a standard deviation of 12%. In their technical paper on Ecotech nephelometer Muller et al. (2011a) dried the sampled aerosols to a RH lower than 25%."

6) Page 14097, Line 1 – MAAP measures the absorption coefficients and the BC is derived from MAAP data. But the authors talk about 'Black carbon (BC) mass concentrations at 637 nm was measured with MAAP,: : :'. It can cause confusion to the reader and therefore it should be corrected in the manuscript (abstract and diverse sections).

The following sentence (Pag. 14097, Line 1):
"Black carbon (BC) mass concentrations at 637nm (Müller et al., 2010b) and particle number concentrations during the study period were measured with Multi Angle Absorption Photometers (MAAP, model 5012, Thermo) and a Condensation Particle Counters (CPC), respectively.",

was replaced with:

"Aerosol absorption coefficients at 637nm (Müller et al., 2011b) and particle number concentrations during the study period were measured with Multi Angle Absorption Photometers (MAAP, model 5012, Thermo) and a Condensation Particle Counters (CPC, Model TSI 3772, D50 = 10 nm), respectively.".

All the sections of the manuscript have been accordingly modified.

Moreover, also following the suggestion of Mr. Ogren and the Anonymous Referee #1, the term "black carbon concentration*"* (or BC) was replaced with "equivalent black carbon concentration" (or EBC) throughout the manuscript in order to take into account that MAAP "calculates" (i.e. does not measure) the concentration of BC.

7) Page 14097, Line 4 - information about the CPC model and about the D50 should be included.

The following sentence has been added:

"(CPC, Model TSI 3772 with D50 of 10 nm.)".

8) Page 14097, Lines 6:10 – the inlet description should be more detail (flows, Reynolds number, humidity control, etc).

The following sentence was added:

"The inlet flow was 1 m^3/h and humidity control was performed by connecting a drier to the sampling inlet. The Reynolds number for the described inlet was around 1300."

9) Page 14098, Lines 1:20 – I have some troubles going through this section. It is very confused and should be improved. I would suggest the following structure but I leave this choice to the authors: begin with the explication about how the SSA is estimated

(from scat–neph and absor–MAAP). After that, discuss how BC is obtained from absor- MAAP and absorption cross section, which was estimated from absor–MAAP and the concentrations of EC in the collected PM10 filters. Finally, the improvement of the BC values obtained with respect on those derived from MAAP manufacturer can be described.

As suggested by the Anonymous Referee #2 (and also taking into account one of the comments of the Anonymous Referee #1), the following sentence (Pag. 14098, Lines 1-20):

"Two aerosol optical parameters, the Single Scattering Albedo (SSA) and the Ångström (Å) exponent, were calculated by using the nephelometer and MAAP data. As known the atmospheric particles have a cooling or warming effect on climate depending on the SSA value. Non-absorbing particles such as sulfate have an SSA of one while lower SSA values indicate the presence of more absorbing particles. The SSA at a given wavelength λ is given by:

$$SSA(\lambda) = \frac{\sigma_{sp}(\lambda)}{\sigma_{sp}(\lambda) + \sigma_{ap}(\lambda)} \qquad (1)$$

where $\sigma_{ap}(\lambda)$ is the particles absorption coefficient at the wavelength λ given by $\sigma_{ap}(\lambda)$ [m^{-1}] = BC [gm^{-3}] × $\sigma(\lambda)$ [m^2g^{-1}] (Petzold and Schönlinner, 2004) where $\sigma(\lambda)$ is the absorption cross section. We calculated the hourly SSA values from equation 1 by using the σ_{sp} at 635 nm obtained with the nephelometer and the σ_{ap} at 637 nm taken with the MAAP. Thus, the σ_{ap} was calculated by dividing the equivalent BC concentration given by the MAAP by 6.6 m^2/g which is the value of the absorption cross section recommended by the manufacturer. Then, following the experimental procedure described in Fernández-Camacho et al. (2010) and Reche et al. (2011), the actual aerosol absorption cross section $\sigma(\lambda)$ was determined by comparing the absorption coefficient $\sigma_{ap}(\lambda)$ measured by the MAAP with the concentrations of EC in the collected PM_{10} filters as reported in Figure 2. An average value of $\sigma(\lambda) = 10.4\pm2.0$ m^2/g was obtained and used to correct the equivalent concentration of BC given by the MAAP instrument." ,

was replaced with:

"In this work, two additional aerosol optical parameters, the Single Scattering Albedo (SSA) and the Ångström exponent (å), were calculated by using the nephelometer and MAAP data. As known the atmospheric particles have a cooling or warming effect on climate depending on the SSA value. Non-absorbing particles such as sulfate have an SSA of one while lower SSA values indicate the presence of more absorbing particles. The SSA at a given wavelength λ is given by:

$$SSA(\lambda) = \frac{\sigma_{sp}(\lambda)}{\sigma_{sp}(\lambda) + \sigma_{ap}(\lambda)} \qquad (1)$$

where $\sigma_{ap}(\lambda)$ is the particles absorption coefficient. Consequently, we calculated the hourly SSA values from equation 1 by using the σ_{sp} at 635 nm obtained with the nephelometer and the σ_{ap} at 637 nm measured with the MAAP. It must be taken into account that the information provided by the MAAP is an equivalent black carbon concentration (EBC) which is calculated by the instrument's software by dividing the measured $\sigma_{ap}(\lambda)$ by 6.6 m^2g^{-1} which is the MAC recommended by the manufacturer. Thus, the following equation is applied: $\sigma_{ap}(\lambda)$ [m^{-1}] = EBC [gm^{-3}] × $\sigma(\lambda)$ [m^2g^{-1}] (Petzold and Schönlinner, 2004) where $\sigma(\lambda)$ is the mass absorption cross section (MAC). Consequently, we calculated the measured absorption coefficient $\sigma_{ap}(\lambda)$ by multiplying the EBC given by the MAAP by the MAC value of 6.6 m^2g^{-1}. Then, $\sigma_{ap}(\lambda)$ and $\sigma_{sp}(\lambda)$ were used in Eq. 1 for the calculation of SSA.

In order to determine a MAC value more appropriate for the aerosols in the WMB we compared the measured absorption coefficients $\sigma_{ap}(\lambda)$ with the concentrations of EC in the collected PM_{10} filters as reported in Figure 2. The uncertainty for the measured EC concentration was calculated by adding one half of the minimum measured EC concentration to the 10% of the concentration ($Err_{[EC]} = \min[EC]/2 + 0.1\cdot[EC]$). This formula gives higher uncertainty to low EC concentrations (Polissar et al., 1998). An average value of $\sigma(\lambda) = 10.4\pm2.0$ m^2/g was obtained and used to calculate the EBC presented in this work. Absorption cross sections between 7 m^2/g and 11 m^2/g are usually reported in literature (see for example Bond & Bergstrom's, 2005; Fernández-Camacho et al., 2010; He et al., 2009; Barnard et al., 2008; Arnott et al., 2003, 2005)."

10) Definitions of aerosol parameters. (backscattering fraction, scattering to backscattering ratio, mass scattering cross section,: : :) should be described in this sections. A physical interpretation of each of them should be also discussed.

The following sentence was added at the beginning of the Section 2.3:

"The aerosol total scattering σ_{sp} (λ) and hemispheric backscattering σ_{bsp} (λ) coefficients from nephelometer describe the interaction of light with the particles in the atmosphere as a function of the wavelength. Thus, σ_{sp} (λ) is a measure of the elastic diffuse reflection of radiation at all angles (0-360º) while σ_{bsp} (λ) represents the radiation elastically scattered by particles back to the direction where the radiation come from. The σ_{bsp} (λ) from nephelometer is called hemispheric backscattering given the large angular distribution of the backscattered radiation measured by nephelometers (90º to ≈170º). These optical parameters are function of aerosol properties such as size, shape, composition, refractive index and both are fundamental parameters for estimating the effect of atmospheric aerosol on climate (IPCC, 2007). The ratio hemispheric backscatter-to-scatter can be used to estimate the asymmetry parameter of airborne particles used in radiative transfer calculations (Andrews et al., 2006). The attenuation of light during wave propagation in the atmosphere is determined also by the absorption properties of particles described by the particle absorption coefficient σ_{ap} (λ). A major role in absorbing radiation is played by the light-absorbing carbon (LAC; Bond and Bergstrom, 2006) called elemental carbon (EC) or black carbon (BC) depending on the analytical methods used to quantify its atmospheric concentration: thermal/optical techniques for EC and light-absorption measurements for BC (Subramanian et al., 2010). The determination of optical and chemical properties of LAC is important for climate studies as LAC can change its optical properties by absorbing up to 50% more light if coated with non-absorbing matter such as ammonium sulfate (Bond et al., 2006). The σ_{sp} (λ) and σ_{ap} (λ) are linked to the concentration of scattering or absorbing particles by their mass scattering cross section (MSC) and mass absorbing cross section (MAC) respectively."

11) Page 14099, Lines 12:15 – Consider including a brief discussion of the skew-ness parameters obtained.

A brief discussion was included and the following sentence (Pag. 14099, Lines 12-15):

"The skewness measures the asymmetry of a distribution function and the higher the skewness, the higher the probability of measuring high levels for the considered aerosol component or parameter."

was replaced with:

"The skewness measures the asymmetry of a distribution function and the higher the skewness, the higher the probability of measuring levels higher than the mean for the considered aerosol component or parameter. All variables reported in Table 1, apart from SSA and å, show positive skewness with high values being more frequent than low ones, as is typical for many positive defined meteorological parameters (This work, Table 1; O'Neill et al., 2000; Matthias and Bösenberg, 2002; Querol et al., 2009; Pereira et al., 2011). Thus, as shown in the section 3.3, a positive skewness leads to a frequency distribution with a tile toward positive values. As already observed, the radiation scattered by particles in the forward direction increases more rapidly than the backscattered radiation with increasing particle size (van de Hulst, 1957). Consequently, there is a higher probability of measuring values much higher than the mean for total scattering compared with backscattering, thus leading to higher skewness for σ_{sp} rather than for σ_{bsp} (Table 1). Exceptions from this behaviour were observed for the SSA and the å exponent showing negative skewness indicating the presence of a tile toward values lowers than the mean in the frequency distributions."

Consequently, the following sentence was removed (Section 3.3, Pag. 14104, Lines 15-23):

"The observed distribution of σ_{sp} is typical for many positive defined meteorological parameters (This work, Table 1; O'Neill et al., 2000; Matthias and Bösenberg, 2002; Querol et al., 2009; Pereira et al., 2011). Exceptions from this behaviour in Table 1 was observed for the SSA and the Å exponent showing negative skewness indicating the presence of a tile toward values lowers than the mean in the frequency distributions.”.

12) Page 14101, Lines 14-16 – This sentence should be deleted. It was mentioned previously in Sect. 2.3.

Done. Moreover, the reference *Schuster et al., 2006* was moved to the section 2.3

13) Page 14101, Lines 16-25 – Consider moving the relation between PM and Angstrom parameter to other better location.

Given that, following the next comment of the Anonymous Referee #2, we have added the mean concentration of PM1, PM2.5 e PM10 at Pag. 14101, Line 16, we would like, with the permission of the Anonymous Referee #2, to not move the relationship between PM and å.

14) Page 14102, Lines 13-14 – The mean PM10 and PM1 concentrations, as well as PM2.5, should be indicated in Page 14101, Line 16. A brief comment about the comparison with other sites should be also mentioned (or at least some reference of previous studies in northeaster Spain).

Done. The following sentence was added in Pag. 14101, Line 16:

“Mean PM_1, $PM_{2.5}$ and PM_{10} were 10.1±7.5 μg/m^3, 13.0±8.8 μg/m^3, and 16.6±11.9 μg/m^3, respectively. The measured PM_x concentrations were consistent with those typically registered in northeaster Spain (Querol et al., 2008; Pérez et al., 2008a; Pey et al., 2009; Pey et al., 2010).”

Consequently, the following sentence (Pag. 14102, Lines 14-15):

“Over the entire study period the mean PM_{10} concentration was 16.6±11.9 μg/m^3.”,

was removed.

15) Page 14102, Lines 15:17 – In page 14096 Line 27, the measured detection limits of the nephelometer were exposed, being about 0.2 Mm-1. But In Fig. 5, if the measured extremely negative Angstrom exponents were due to instrumental noise, the detection limits look like about 10 mM-1. What could be the reason?

Firstly, we point out that the Muller et al. (2011, AMT) in their technical paper on the Ecotech nephelometer present new detection limits values (with respect to those published in the Discussion paper).
Consequently, the following sentence (Pag. 14096, Lines 25-28):

“Müller et al. (2010a) measured detection limits of Aurora 3000 over a one minute averaging time at wavelengths 450 nm, 525 nm, and 635nm of 0.20, 0.12, 0.29 Mm−1 for total scattering, and 0.06, 0.07, 0.04Mm−1 for backscattering, respectively.”,

was replaced with:

“Müller et al. (2011a) measured detection limits of Aurora 3000 over a one minute averaging time at wavelengths 450 nm, 525 nm, and 635nm of 0.11, 0.14, 0.12 Mm^{-1} for total scattering, and 0.12, 0.11, 0.13 Mm^{-1} for backscattering, respectively.”.

Moreover, we must point out also that after the *New correction* applied in this revised version of the manuscript, Figure 5 slightly changed.

In the new Figure 5, the mean value of the total scattering at 635 nm calculated for Angstrom exponents smaller than -2 is 1.4 Mm^{-1} (and 5.8 Mm^{-1} for Angstrom exponents smaller than 0). A value of 1.4 is lower than the value (10 Mm^{-1}) visually identify by the Anonymous Referee #2 from the old Figure 5, but anyway it is higher than the detection limit reported by Muller et al. (2011, AMT). There are two possible explanations. Firstly, we compare the total scattering with a parameter (the Angstrom exponent) which is calculated starting from the three wavelength total scattering from nephelometer, and consequently subject to higher errors in presence of low scattering. Secondly, negative å could be actually due to the presence of relatively larger particles during low aerosol concentration. Also Figure 7e indicate this reduction of the Angstrom exponent for aerosol scattering coefficient lower that 35 Mm^{-1}.

Consequently, the following sentence (Pag. 14102, Lines 4-18):

“In the present case 57 values of the Angstrom exponent out 5 of 7834 hourly values were smaller than −1 (0.7%) and 5 values were higher than 4 (0.06 %). Moreover, Table 1 indicates that the 1-th and the 99th percentiles for the Angstrom exponent were −0.7 and 2.1, respectively. Figure 5 shows the distribution of the scattering coefficient at 635 nm as a function of the calculated hourly Angstrom exponents. As reported in the Figure both negative and positive Angstrom exponents were always related with low scattering coefficients. A closer analysis of the air mass origin revealed the absence of Saharan dust intrusions during negative Å values and the prevalence of Atlantic advection episodes leading to low PM concentrations. The mean PM10 and PM1 concentrations were in fact 4.3±1.6 µgm−3 and 2.3±1.2 µgm−3 respectively when measuring negative Å. Over the entire study period the mean PM10 concentration was 16.6±11.9 µgm−3. Consequently, the measured extremely negative and positive Angstrom exponents were likely due to instrumental noise under very low scattering conditions. Note also that, as reported in Fig. 4, on a daily basis the Angstrom exponents were always positive and smaller than 4.”

Was replaced with:

“In the present case 57 values of the Ångström exponent out of 7834 hourly values were smaller than -1 (0.2 %) and 5 values were higher than 4 (0.1 %). Moreover, Table 1 indicates that the 1th and the 99th percentiles for the Ångström exponent were -0.03 and 2.3, respectively. Figure 5 shows the distribution of the scattering coefficient at 635 nm as a function of the calculated hourly Ångström exponents. As reported in the Figure both extremely negative (< -2) and positive (> 4) hourly Ångström exponents were always related with low scattering coefficients. A closer analysis of the air mass origin revealed the absence of Saharan dust intrusions during negative å values (å < 0) and the prevalence of Atlantic advection episodes leading to low PM concentrations. The mean PM_{10}, $PM_{2.5}$ and PM_1 concentrations were 4.6±1.8 $\mu g/m^3$, 3.0±1.1 $\mu g/m^3$ and 2.6±1.1 $\mu g/m^3$ respectively when measuring negative å. Consequently, the measured negative Ångström exponents were likely due to both the presence of relatively larger particles during low aerosol concentration and the instrumental noise under low scattering conditions.”.

16) Page 14103, Lines 8:9 – Some references should be included.

The following sentence (Pag. 14103, Lines 8-9):

“The reduction of the mass scattering cross section with increasing wavelength reflects the $\lambda^{-Å}$ dependence of σ_{sp}.”,

was replaced with:

“The reduction of the mass scattering cross section with increasing wavelength reflects the $\lambda^{-å}$ dependence of σ_{sp} (van de Hulst, 1957; Kokhanovsky, 2008).”.

17) Page 14103, Lines 20:24 – Some references about the process of adsorption of SO2 and the formation of coarse SO42- could be included. Is it possible to include quantitative information on the reduction of SO2?

The sentence (Pag. 14103, Lines 22-23):

“…which favoured the absorption of species like SO_2 and the formation of coarse SO_4^{2-}”,

was replaced with:

“…which favoured the adsorption of species like SO_2 and the formation of coarse SO_4^{2-} (Vrekoussis et al., 2005; Adams et al., 2005) “.

Concerning the question of the Anonymous Referee #2, we calculated the mean SO2 concentration for the days highlighted by the orange boxed areas in Figure 6b (mean $SO_2^{(Ads)}$ = 1.00±0.00 $\mu g/m^3$) and for the days included in the fitting process of Figure 6b (mean $SO2^{(no\text{-}Ads)}$ = 1.57±1.74 $\mu g/m^3$). Thus, a lower mean SO2 concentration (SO_2

$^{(Ads)}$) was actually measured under adsorption conditions. However, the standard deviation of the mean SO_2 $^{(Ads)}$ is zero. Thus, the levels of SO_2 $^{(Ads)}$ were always around the detection limit of the instrument which, in this case, gives constant values of 1. Consequently, we are not very much confident with these SO2 measurements. Moreover, even if it is actually possible to appreciate a difference between SO_2 $^{(Ads)}$ and SO2 $^{(no\text{-}Ads)}$, the adsorption of SO2 could have took place during the transport from the SO2 sources (mainly the coastline) toward the Montseny station, thus considerably complicating the estimation of the actual SO2 reduction.

Thus, we the permission of the Anonymous Referee #2, we prefer not include these SO2 measurements.

18) Page 14104, Line 5 – "Mass" scattering cross sections for fine: : :

Done.

19) Page 14104, Line 12 (Fig. 7) – About the frequency counts of aerosol scattering coefficients, I suggest to use the frequency in percentages. It is more visual.

Done. Figure 7 was changed.

20) Since aerosol SSA depends on the absorbing part of the extinction and then on the chemical species, the mean aerosol mass concentrations and chemical species as a function of the observed SSA should be a goal to obtain in this manuscript. This new information should be included in conclusions accordingly.

We thank the Anonymous Referee #2 for this comment. Given that also the Anonymous Referee #1 raised a similar comment for the aerosol mass absorption cross section, both comments were addressed in the Section 3.5, by adding the following sentence:

"Low correlation with air mass origin was observed for the SSA which was almost similar during the four observed scenarios. Despite the increase in the absorption coefficient observed under the WAE scenario, the corresponding increase in PM_1 concentration was so high to enhance the mean aerosol scattering properties thus leading to SSA values similar between WAE and AA. Moreover, the absorbing properties of Saharan dust (Vrekoussis et al., 2005) could have accounted, at least in the present case, for the similarity observed between the SSA values during the NAF and WAE episodes.
In order to study the dependence of the mass absorption coefficient (MAC) on the origin of the air masses, the correlation between MAC and EC concentration was analysed as a function of the four defined scenarios. Mass absorption cross sections of 7.5±1.8 m^2/g, 10.2±2.0 m^2/g, 10.7±1.8 m^2/g, and 11.6±2.0 m^2/g at 635 nm for AA, NAF, REG and WAE scenarios respectively were calculated. Thus, the mass absorption cross sections for NAF, REG and WAE were relatively high and not significantly different, while a statistically-significant

difference was observed for the MAC value under AA scenario. The value of MAC estimated under AA is similar to the value proposed by Bond and Bergstrom (2006) of 7.5 m^2/g (at 550 nm), or 6.5 m^2/g at 635 nm assuming a λ^{-1} dependence of MAC, for fresh light-absorbing carbon (LAC). As already stated, the AA episode, from one side, and WAE and REG episodes, from the other side, represents two extremes in terms of degree of pollution in the WMB. The strong winds blowing constantly from the West (from the Atlantic Ocean) under AA episodes assure fresh clean air with low concentrations of pollutants at regional level, while the WAE and REG episodes involve the stagnation (WAE) and recirculation (REG) of air masses around the WMB for a few days with subsequent accumulation of pollutants, which can be transported to regional areas inland by the sea breeze. Previous studies in the area based on both modelling and experiments showed that this recirculation/accumulation of pollutants can span for more than five days as long as this weather pattern is maintained (Millán et al., 1992; Gangoiti et al., 2001; Pey et al., 2010). Consequently, the aerosols transported toward the MSY station under REG and WAE scenarios were mainly aged rather than fresh. As reported in literature (see for example Knox et al., 2009; Bond and Bergstrom, 2006) an increase in mass absorption cross section can be due to difference in coating thickness as a result of the aging of the aerosols. Bond et al. (2006) estimated that absorbing carbon can change its optical properties by absorbing up to 50% more light if coated with ammonium sulfate (Bond et al., 2006). In our study, mean EC and sulfate concentrations in PM_{10} filters under WAE and REG scenarios were 0.39 $\mu g/m^3$ and 2.49 $\mu g/m^3$ and 0.27 $\mu g/m^3$ and 3.62 $\mu g/m^3$, respectively. Under the AA scenario mean EC and sulfate were 0.17 $\mu g/m^3$ and 1.04 $\mu g/m^3$ respectively. Thus, higher sulfate concentrations were on average observed under WAE and REG compared with AA, thus probably explaining the observed differences among the calculated MAC values. The relatively high sulfate burden observed under WAE and REG episode could also explain the small variations observed for SSA as a function of the air mass origin. Even if a higher absorption of light was observed under WAE compared to AA, the increase of WAE sulfate concentration enhanced the scattering of light thus probably leading to similar SSA for the WAE and AA scenarios."

21) Pereira et al., 2010 has been published in 2011.

Done.

22) Sciare et al., 2005 was published in ACP in 2005.

Done.