Anonymous Referee #1

1) Specific comments:
The authors state to explain the variability in aerosol optical properties by the origin and ageing of air masses (Page 14902 line 21). The reviewer thinks that the effects of aging on aerosol properties were not discussed sufficiently. The authors should discuss the aerosol aging alongside with mass specific absorption coefficients and aerosol mixing state. In chapter 2.3 the mass specific absorption coefficient was determined to be 10.4 m2/g. Is there a dependence of the mass absorption coefficient on the origin of the air mass?

We thank the Anonymou Referee #1 for this comment. We calculated the mass absorption cross sections (MAC) as a function of the 4 scenarios (AA, WAE, NAF and REG) we defined. We observed a statistically significant difference between the MACs calculated under Atlantic Advection (AA) and Winter Anticyclonic Episodes (WAE), with MAC under AA lower than the MAC under WAE mainly as a consequence of the ageing of the sampled particles.

Consequently, the following sentence was added to the Section 3.5 (Cluster analysis):

"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 episodes, from one side, and WAE and REG episodes, from the other side, represent 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."

Moreover, the following sentences were added to the Abstract:

"The MAC values appear dependent of particles aging: similar to the expected absorption cross-section for fresh emissions under Atlantic Advection episodes and higher under aerosol pollution episodes.",

and to the Conclusions:

"The aerosol absorption cross section (MAC) also changed as a function of the origin of air masses. A mean value of 10.4±2.0 m^2/g for MAC was calculated during the study period. Relatively small MAC value of 7.5±1.8 m^2/g at 635 nm was calculated under Atlantic Advection episodes, while higher MAC (11.6±2.0 m^2/g) was estimated during winter anticyclonic episodes (WAE) leading to an accumulation PM and sulfate aerosols over the WMB with consequent ageing of the particles."

2) Page 14092 line 7: MAAP does not measure BC but absorption coefficient.

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 14042 line 14: The manuscript is not accessible to the reviewer.

The paper of Pandolfi et al., (2011) [Pandolfi, M., Querol, X., Alastuey, A., Jimenez, J., Jorba, O., Stohl, A., Comerón, A., Sicard, M., Pey, J, vanDrooge, B. and the DAURE team: Source and origin of PM in the Western Mediterranean Basin: An Overview of the DAURE campaign, in preparation, Atmos. Chem. Phys. Discuss, 2011.] is almost ready to be submitted to ACPD. We think that once the aforementioned manuscript from Pandolfi et al. will be published in ACPD it will be a useful reference for the readers of the present manuscript. With the permission of the Anonymous Referee#1 we would like to leave this reference in our manuscript.

4) Page 14096 lines 20-22: It is not entirely clear how the correction was done. What is the result of the experimental comparison between Ecotech and TSI nephelometers?

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.

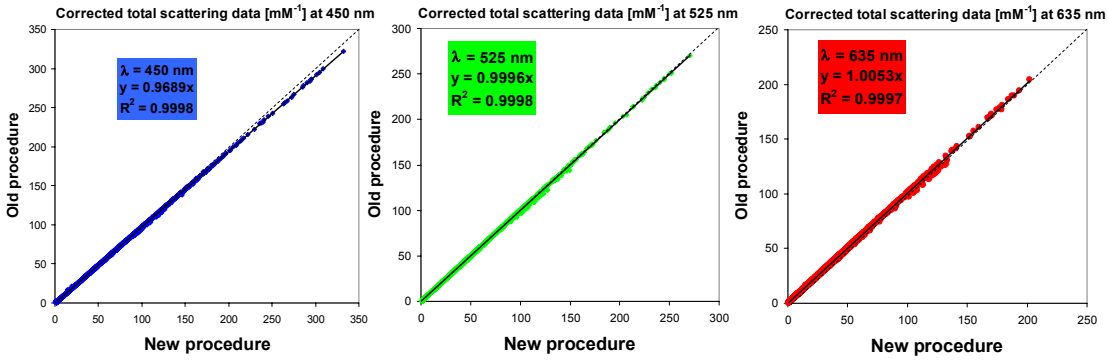
Moving on with the comment, during the open discussion phase of the paper from Müller et al.(2010 AMT Discussion), Mr. Ogren posted the following comment: "*Ecotech neph users are going to be knocking down your doors for a simplified correction scheme for truncation/illumination errors. It would be very 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 23-24: The heater prevents the presence of liquid particles in the nephelometers. Does it also prevent effects of hygroscopicity induced scattering enhancement of particles? The authors should discuss humidity effects and give ranges of humidity in the measurements systems.

Yes, 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 measure primarily the absorption coefficients.

The following sentence:
"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.".

7) Page 14097 line 1: Reference must be Mueller et al. 2011.

Müller et al., 2010b was replaced with Müller et al., 2011b.

8) Page 14098 lines 9 to 23: The section might be misunderstood by the readers. MAAP measures absorption coefficient. The black carbon concentration calculated from MAAP is an equivalent BC concentration assuming a specific mass absorption coefficients. In that section the authors derive a mass specific absorption coefficient of 10.4 m2/g, which is more appropriate for the aerosols in the WMB. All following BC concentrations in the manuscript are linked to the absorption coefficient by this value. The reviewer suggests to consequently use the term equivalent black carbon throughout the manuscript if the values are derived from MAAP.

We agree with the Anonymous Referee #1. Consequently, the term "black carbon concentration" (or BC) was replaced with "equivalent black carbon concentration" (or EBC) throughout the manuscript. Moreover, the following sentence:

"…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±2.0 m^2/g was obtained and used to correct the equivalent concentration of BC given by the MAAP instrument."

was changed and replaced with:

"…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·[EC]). This formula gives higher uncertainty to low EC concentrations (Polissar et al., 1998). An average value of $\sigma(\lambda)$ = 10.4±2.0 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)."

9) Line 14100 line 25: "These relatively high values… " which high values ? It is not clear to what this is referring to.

We refer to the values registered in the Western Mediterranean Basin. In order to clarify this point in the manuscript, the following sentence:

"These relatively high values reflected from one side the effect of the Saharan dust events frequently observed in the Mediterranean Basin and from the other side the impact of continental pollution on the Eastern Mediterranean coast.",

was replaced with:

"The relatively high values registered in the Western Mediterranean Basin reflected from one side the effect of the Saharan dust events frequently observed in the Mediterranean Basin and from the other side the impact of continental pollution on the Eastern Mediterranean coast."

10) Page 14103 Line 14: Typo in "-3:-1 Mm-1".

Done.

11) Page 14103 line 22: "absorption" or "adsorption".

"absorption" was replaced with "adsorption".

12) Page 14106 lines 9-12: What is the source of the increased atmospheric BC?.

The increase in EBC concentration at MSY under sea breeze circulation was due to the transport, driven by the sea breeze, of polluted air masses from the highly urbanized/industrialized coastline and valleys toward inland rural areas where the MSY measurement station is located.

In order to clarify this point the following sentence (Pag. 14106, Lines 9-12):

"The increase of the absorption coefficient was instead higher and corresponding to about 100% (from 1.9 Mm^{-1} to 3.9 Mm^{-1}) as a consequence of the strong increase in BC concentrations at MSY observed under sea breeze circulation (~ 400 ngm^{-3}, Fig. 8d)."

was replaced with:

"The increase of the absorption coefficient was instead higher and corresponding to about 100% (from 1.9 Mm^{-1} to 3.9 Mm^{-1}) as a consequence of the strong increase in EBC concentrations observed at MSY under sea breeze circulation (~ 400 ngm^{-3}, Fig. 8d). Thus, EBC increased in the afternoon at MSY station as a consequence of the transport, driven by the sea breeze, of polluted air masses from the highly urbanized/industrialized coastline and valleys toward inland rural areas where the MSY station is located.".

13) References: Mueller et al. 2010a has been published in 2011.

The reference Müller et al. 2010a (AMTD) was replaced with the recently published paper on AMT (Müller et al. 2011a).

14) Table 1: Is the statistics based on the hourly mean values? Row BC: Is that the equivalent BC? If yes, it should be noted that it was calculated from sigma_abs using

the mass absorption coefficient of 10.4 m2/g. What does the wavelength of 670 nm mean for BC? Value for maximum of BC seems to be too high?

Yes, the statistics is based on the hourly mean values. The sentence "Statistics is based on hourly mean values" has been added to the caption of Table 1.

Yes, the concentration reported in Table 1 is the concentration of the equivalent black carbon (EBC). Consequently, "BC" was replaced with "EBC" in Table 1 and corresponding caption.

The nominal wavelength given by MAAP manufacturer is 670 nm. However, this wavelength differs significantly from the wavelength measured during two intercomparison workshops dedicated to the characterization of aerosol absorption photometers [Müller, T., Henzing, J. S., de Leeuw, G., Wiedensohler, A., Alastuey, A., Angelov, H., Bizjak, M., Collaud Coen, M., Engstr¨om, J. E., Gruening, C., Hillamo, R., Hoffer, A., Imre, K., Ivanow, P., Jennings, G., Sun, J. Y., Kalivitis, N., Karlsson, H., Komppula, M., Laj, P., Li, S.-M., Lunder, C., Marinoni, A., Martins dos Santos, S., Moerman, M., Nowak, A., Ogren, J. A., Petzold, A., Pichon, J. M., Rodriquez, S., Sharma, S., Sheridan, P. J., Teinil, K., Tuch, T., Viana, M., Virkkula, A., Weingartner, E., Wilhelm, R., and Wang, Y. Q.: Characterization and intercomparison of aerosol absorption photometers: result of two intercomparison workshops, Atmos. Meas. Tech., 4, 245–268, doi:10.5194/amt-4-245-2011, 2011.].

Müller et al (2011) found that the optical wavelength of MAAP is 637 nm instead of 670 nm.

Throughout the manuscript we refer to the wavelength of 635 nm. By mistake we reported a wavelength of 670 nm in Table 1. Consequently, "670" in Table 1 was replaced with "637".

Yes, the maximum value of EBC reported in Table 1 is expressed in ng/m^3 and not in $\mu g/m^3$. Consequently, the value "3294" was replaced with "3.294".

15) Figure 7: (i) and (f) are the same considering the mass absorption coefficient of 10.4 m2/g.

Yes. But given that also in Table 1 we give both values (EBC and Absorption coefficient), we prefer, with the permission of the Anonymous Referee #1, to leave the Figure 7 as is. The label of Figure 7i was opportunely changed from "Black Carbon [ng/m^3]" to "Equiv. Black Carbon [ng/m^3]" .