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**Monitoring of the  
Eyjafjallajökull  
volcanic aerosol  
plume**

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## Abstract

Lidar and sun-photometer measurements were performed intensively over the Iberian Peninsula (IP) during the eruption of Eyjafjallajökull volcano (Iceland) in April–May 2010. The volcanic plume hit all the IP stations for the first time on 5 May 2010. A thorough study of the event is conducted for the period 5–8 May. Firstly the spatial and temporal evolution of the plume is described by means of lidar and sun-photometer measurements supported with backtrajectories. The volcanic aerosol layers observed over the IP were rather thin ( $< 1000$  m) with a top height up to 11–12 km. The mean optical thicknesses associated to those layers were rather low (between 0.013 and 0.020 over the whole period). Punctually on 7 May the optical thickness reached peak values near 0.10. Secondly the volcanic aerosols are characterized in terms of extinction and backscatter coefficients, lidar ratios, Ångström exponents and linear particle depolarization ratio. Lidar ratios at different sites varied between 30 and 50 sr without a marked spectral dependency. Similar extinction-related Ångström exponents varying between 0.6 and 0.8 were observed at different sites. The temporal evolution of the backscatter-related Ångström exponents points out a possible decrease of the volcanic particle size as the plume moves from west to east. Particle depolarization ratios on the order of 0.06–0.08 confirmed the coexistence of both ash and non-ash particles. Additionally profiles of mass concentration were obtained with a method using the opposite depolarizing effects of ash particles (strongly depolarizing) and non-ash particles (very weakly depolarizing), and sun-photometer observations. In Granada the ash mass concentration was found approximately 1.5 higher than that of non-ash particles, and probably did not exceed the value of  $200 \mu\text{g m}^{-3}$  during the whole event.

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## 1 Introduction

The eruption of the Eyjafjallajökull volcano, an ice-covered stratovolcano with a summit elevation of 1666 m a.s.l. situated in Southern Iceland, started on 14 April 2010 and stopped on 21 May 2010. This eruption threw volcanic aerosols with variable intensity several kilometers up in the atmosphere (Langmann et al., 2011) which were transported mostly towards Europe and led to air travel disruption in Northern and Central Europe from 15 April onwards and in South Europe in May 2010. Many major Northern and Central European countries closed their airspace completely during several days. In total more than 100 000 flights were canceled that affected more than 10 millions passengers. The event might have contributed relatively quickly to a decrease of the global surface temperature since ash particles reflect the solar radiation, and the emission of greenhouse gases decreased significantly during the air travel disruptions in April and May. In the long term volcanic aerosols are well-known to have a notable impact on the Earth radiative budget because of their large scale dispersion and their long residence times in the atmosphere.

The lidar technique is one of the most relevant remote sensing tools to study atmospheric aerosols. In the past volcanic aerosols (VA) have been observed by lidars a long time after they have been ejected in the stratosphere (Langford et al., 1995; Borrmann et al., 1995; Wandinger et al., 1995; Di Girolamo et al., 1996) and less frequently in the troposphere (Pappalardo et al., 2004; Villani et al., 2006; Wang et al., 2008). The Eyjafjallajökull volcanic plume was followed in near-real time by many scientists from different fields of atmospheric sciences all over the world but especially in Europe since the very first day of the eruption. While many results obtained in Northern and Central Europe have already been published (Ansmann et al., 2010, 2011; Flentje et al., 2010; Wiegner et al., 2011; Gasteiger et al., 2011; Schumann et al., 2011; Emeis et al., 2011) only very few results about the situation in Southern Europe can be found in peer-reviewed literature (Balis et al., 2010; Mona et al., 2011; Toledano et al., 2011).

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water vapor (H<sub>2</sub>O), hydrochloric acid (HCl) and hydrofluoric acid (HF) among others (<http://volcanoes.usgs.gov/hazards/gas/s02aerosols.php>). Ash particles fall out quite rapidly due to their large mass so that over the IP, far away from the source, their contribution may be significantly reduced. On the contrary the residence time of sulfate aerosols is much longer for they can reside in the atmosphere for several months.

This paper is organized as follows: Sect. 2 briefly presents the instrumentation; in Sect. 3 the spatial and temporal evolution of the volcanic plume over the IP is described; and in Sect. 4 the vertical characterization of the VA is presented in terms of their optical properties and an estimate of their mass concentration is shown. Conclusions are made in Sect. 5.

## 2 Instruments

### 2.1 Lidars

The four lidar systems involved in this study all belong to EARLINET and SPALINET. A short description of them can be found in Sicard et al. (2009, 2011). The principal characteristics of those systems are presented in Table 1. The institutions involved are:

- Centro de Geofísica de Évora, Évora (Portugal, 38.57° N, 7.91° W, 293 m above sea level, a.s.l.);
- Centro de Investigaciones Energéticas Medioambientales y Tecnológicas, Madrid (Spain, 40.46° N, 3.72° W, 665 m a.s.l.);
- Universidad de Granada, Granada (Spain, 37.16° N, 3.58° W, 680 m a.s.l.);
- Universidad Politécnica de Cataluña, Barcelona (Spain, 41.39° N, 2.11° E, 115 m a.s.l.).

All four systems participated in an intercomparison campaign in Madrid from 18 October to 5 November 2010 in the framework of the EARLINET quality assurance program

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and were satisfactorily compared to the reference system of Potenza, Italy (for more details see Molero et al., 2011). Coordinated measurements started on 15 April 2010 and intensified on 5 May accordingly with the intrusion. All measurements were inverted using the two-component elastic lidar inversion algorithm (Fernald, 1984; Sasano and Nakane, 1984; Klett, 1985) and a constant lidar ratio of 50 sr. All nighttime measurements were also inverted using the Raman lidar inversion algorithm (Ansmann et al., 1990; 1992).

## 2.2 Sun-photometers

Some columnar aerosol properties of interest were measured by CIMEL sun-sky photometers in Évora, Granada and Barcelona. For Madrid the sun-photometer in Cáceres (located approximately 250 km southwest of Madrid) was used. All sun-photometers are part of the Aerosol Robotic Network (AERONET, <http://aeronet.gsfc.nasa.gov/>) (Holben et al., 1998). The instrument provides information about, among other properties, the aerosol optical thickness (AOT) at 340, 380, 440, 500, 675, 870 and 1020 nm in Évora, Madrid and Granada, and at 440, 675, 870 and 1020 nm in Barcelona, as well as the single scattering albedo and the Ångström exponent between pairs of wavelengths. A common product of AERONET is the fine and coarse mode AOT at 500 nm (O'Neill et al., 2003). Inversion products such as the volume size distribution are also provided.

In this paper we calculated the Ångström exponent between the wavelengths of 440 and 675 nm from Level 1.5 (cloud screened data) at Évora, Granada and Barcelona and from Level 2.0 (quality assured data) at Madrid. For Évora, Madrid and Granada we used the AOT at 500 nm and for Barcelona the AOT at 500 nm was calculated with the one at 440 nm and the Ångström exponent calculated between the wavelengths of 440 and 675 nm.

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## 3 Evolution of the Eyjafjallajökull VA plume over the IP: 5–8 May 2010

### 3.1 Synoptic situation

A complex system developed during the period 5–8 May 2010. Figure 1 shows the synoptic situation over Western Europe in terms of sea level pressure for every day of the period 4–9 May at 12:00 UTC. The start of the period is characterized by a deep anticyclone located south of Iceland and west of Ireland, while a low pressure system affected Southern France. On 5 and 6 May a front formed in the Atlantic west of the IP's coasts as a new low pressure system appeared at the 40° W longitude. During 6–8 May this new low pressure system swept over the Atlantic Ocean eastwards towards the IP leading to pronounced easterly winds starting on 8 May. From 9 May on, this low pressure gradient settled over the north of the IP producing several days of instable conditions with broken clouds and low intensity rains.

Backtrajectories have been calculated with the HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory Model) (Draxler and Rolph, 2003; Rolph, 2003) model provided by NOAA–ARL (National Oceanic and Atmospheric Administration–Air Resources Laboratory) to check the air-masses origin. Figure 2 shows 120-h backtrajectories arriving at the four stations at 6 altitude levels between 500 and 5500 m with a resolution of 1000 m for each day at 12:00 UTC. The length of the trajectories corresponds to 5 days. In Évora and Madrid a transport over Iceland is visible every day at two or more altitude levels. In Évora the backtrajectories are similar on 5 and 6 May: all altitude levels show a transport over Iceland 3 to 4 days prior to the plume arrival. On 7 May the same pattern persists except for the 500-m altitude level. On 8 May only the 500- and 1500-m altitude levels show transport over Iceland approximately 5 days prior to the plume arrival. In Madrid on 5 May only air masses at altitude levels above 3500 m travelled over Iceland 2 to 3 days prior to arrival. On 6 and 7 May the situation is similar to that of Évora. On 8 May only the altitude levels below 2500 m show transport over Iceland approximately 4 days prior to arrival. The situation in Granada and Barcelona is slightly different. On 5 May mesoscale transports dominate. On 6 May

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the air masses at altitude levels below 3500 m have all been transported over Iceland 5 days (at 500 and 1500 m) and 3–4 days (at 2500 and 3500 m) prior arrival. On 7 May all trajectories travel over Iceland 3 days (Granada) and 4–5 days (Barcelona) prior to arrival. On 8 May only the altitude levels below 2500 m show clearly a transport over Iceland with travelling times of 4 to 5 days.

In summary, according to the synoptic situation and the backtrajectory analysis, the VA plume entered the IP from west-northwest at all altitude levels and with relatively short transport times from Iceland between 2 and 4 days. Along the intrusion the plume transported from Iceland reaches the IP at lower altitudes and longer transport times. In the southern and eastern IP the plume appears on 6 May at lower altitudes.

### 3.2 Spatial and temporal evolution of the VA plume over the IP

The spatial and temporal evolution of the VA plume over the IP is analyzed through the time series of vertical profiles of range-square corrected lidar signals (RSCS).

Figure 3 shows the RSCS time series of the four Iberian stations over the whole period. All the measurements performed by the four stations are plotted. The height, if not otherwise stated, is reported above sea level (a.s.l.). Below the lidar time series the sun-photometer total, coarse and fine mode AOT at 500 nm, as well as the Ångström exponent calculated between the wavelengths of 440 and 675 nm are reported for each station. Figure 4 shows the AOT of the lofted layers of VA, hereinafter called  $AOT_{VA}$ , at 532 nm measured by the lidar stations as a function of height and time with a time resolution of 1 h. In this section  $AOT_{VA}$  was calculated by the integration of the backscatter profiles inverted with the two-component elastic lidar inversion algorithm multiplied by a constant lidar ratio of 50 sr. The layers below 2 km that might have been coupled to the PBL have been discarded in order to minimize the influence of local aerosols in the analysis. The measurements with clouds or optically extremely thin VA layers (leading to large errorbars in the retrieval of the optical coefficients) are discarded in Fig. 4. The thickness of lofted VA layers is hereinafter called  $\Delta h_{VA}$ . Table 2 gives the daily mean  $AOT_{VA}$ ,  $\overline{AOT_{VA}}$ , and layer thickness,  $\overline{\Delta h_{VA}}$ , of the lofted VA layers. The minimum and

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maximum values of  $AOT_{VA}$  and  $\Delta h_{VA}$  are also reported in Table 2 to show their large variability.

The uncertainty of  $AOT_{VA}$  can be calculated as the sum of the systematic error and the statistical error. Both terms are given in Eqs. (A3) and (A6) of Appendix A. The systematic error has been calculated using the relative errors of the backscatter coefficient given in Sicard et al. (2009) while the statistical error has been calculated directly with the statistical error of the backscatter coefficient provided for each inversion (EARLINET products such as the backscatter coefficient are provided with their statistical error). For all three wavelengths of 355, 532 and 1064 nm, the uncertainty of  $AOT_{VA}$  is 15–20 % for  $AOT_{VA} \geq 0.01$  (the systematic errors dominate) and 20–30 % for  $0.01 > AOT_{VA} \geq 0.002$  (both errors are similar). Taking into account the low values of  $AOT_{VA}$  found, those uncertainties are reasonable.

On 5 May the first VA plume is detected in Évora just before noon, in Madrid around 16:00 UTC and in Granada around 17:00 UTC. The Barcelona station was covered with clouds part of the afternoon. Clearly marked VA layers are observed above the PBL and below 5–6 km (Fig. 3). However over the 3 stations the rest of the troposphere is not aerosol-free (Fig. 4) indicating a strong vertical dispersion of the plume. No vertical motion is noticeable until around midnight where the plume shows a downward motion at Granada. The sun-photometer AOT does not present any significant increase which also indicates optically thin VA layers. Figure 4 shows clearly the large vertical dispersion in Évora with discontinued layers between 2.3 and 9.3 km. In Madrid the first lofted layer of VA detected above the PBL is very broad (up to 6600 m thick). This difference compared to the other stations can be explained by the Iberian thermal low (Millán et al., 1992, 1997). In the warm season the formation of the thermal low forces the convergence of surface winds from the coastal areas towards the central plateau of the IP where Madrid is located. Once over the central plateau the surface air masses are injected into the middle troposphere “pushing” upwards the lofted layers. The first lofted layer of VA is found between 4.0 and 11.4 km without clear discontinuity while in Granada much thinner VA layers ( $\Delta h_{VA} = 422$  m) are detected. Both in Granada and

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in Évora the mean optical thickness of the VA layers is very low around 0.007–0.008 which represents less than 10% of the columnar AOT. In Madrid the first VA layers detected has an  $AOT_{VA}$  of 0.010.

At the beginning of 6 May the same situation persists in Évora and Madrid whereas Granada is covered by mid- and high-altitude clouds. VA layers are clearly visible above 2.5 km in Évora and between 2.5 and 7.0 km in Madrid throughout the day. The sun-photometer AOT increases substantially during the day: it passes from 0.09 at noon in both stations to 0.20 at 18:15 UTC in Évora and to 0.16 at 18:00 UTC in Madrid. In both stations the Ångström exponent slightly decreases compared to the previous day. The coarse mode AOT is constant in both stations which indicates that the total AOT increase is due to fine particles only. Slightly after 17:30 UTC a sudden increase of the AOT is visible in both stations. It is associated to an increase of the coarse mode AOT and a decrease of the Ångström exponent which indicates the intrusion of a new layer formed by coarse particles. The Évora RSCS time series indicate the intrusion of a well defined VA layer above 5 km with a clear downward motion. The same intrusion is observed in Madrid after 21:00 UTC but there a cirrus cloud is embedded in the descending VA layer between 5 and 8 km. In terms of  $AOT_{VA}$ , the main layer observed in Évora on 5 May remains during the first half of 6 May as very thin layers appear below. The new layers which appear above 5 km after 18:00 UTC have higher optical thicknesses. In Madrid the VA layers are more stable than on the previous day:  $AOT_{VA}$  oscillates around 0.008 and  $\Delta h_{VA}$  between 600 and 1500 m. Both in Évora and in Madrid the mean optical thickness of the VA layers is around 0.008–0.009, i.e. similar to the values on the previous day. One can easily deduce that the columnar AOT increase is due to aerosols trapped in the layers below 2 km which according to the backtrajectory analysis have been transported over Iceland. Once again the mean vertical dispersion of the VA layers is stronger in Madrid ( $\Delta h_{VA} = 1000$  m) than in Évora ( $\Delta h_{VA} = 543$  m). In Granada very thin layers ( $AOT_{VA} \approx 0.004$ ,  $\Delta h_{VA} = 325$  m) with a downward motion are observed between 2 and 5 km before noon. No sun-photometer data is available.

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On 7 May several descending VA layers are detected over Évora between 2 and 6 km, which are optically thicker than those observed on 6 May. Shortly before noon clouds start developing on top of the PBL preventing a further analysis. In Madrid clouds are present on top of the PBL from 04:00 to 05:00 UTC, VA layers are observed between 07:00 and 08:00 UTC and later clouds are again embedded in the VA layers between 2 and 5 km. In Granada a complex situation similar to that in Évora is observed in the morning. In both Évora and Madrid the sun-photometer AOT (and essentially the fine mode AOT) has increased compared to the previous day while the Ångström exponent has no marked trend. In Granada two AOT increases associated with a slight increase of the Ångström exponent are observed after 09:30 and 15:30 UTC. In both cases the Granada RSCS time series indicate the intrusion of new well defined VA layers. In Évora while the optical thickness of the VA layers has increased ( $AOT_{VA} \approx 0.025$ ) compared to 6 May, their thickness has decreased ( $\Delta h_{VA} = 360$  m). This indicates smaller and optically thicker VA layers. In Granada the VA layers present roughly the same characteristics ( $AOT_{VA} \approx 0.026$ ,  $\Delta h_{VA} = 412$  m). As the day goes by, the lowest lofted VA layers get coupled to the PBL both in Évora (in the morning) and Granada (in the afternoon).

On 8 May single-layer stratifications predominate: in Madrid a layer is detected between 5.0 and 7.0 km in the first hours of the day, in Granada between 2.2 and 4.2 km and in Barcelona between 1.9 and 3.1 km. Except in Madrid (again for the same explanation given above about the formation of the Iberian thermal low), the VA layers are observed at lower altitudes than at the beginning of the period. The total AOT decreases following the variations of the fine mode back to its seasonal mean value (0.10) in Granada (Alados-Arboledas et al., 2003) and oscillates around 0.15 in Barcelona, which also comes to be a seasonal mean value (Sicard et al., 2011). In Granada the VA layers have approximately the same  $AOT_{VA}$  (0.027) than on the previous day while their thickness has decreased ( $\Delta h_{VA} = 343$  m). However  $AOT_{VA}$  and  $\Delta h_{VA}$  are not constant throughout the day: the relatively thick layers (spatially and optically,  $AOT_{VA} > 0.060$ )

observed at the beginning of the day get thinner as the day goes by.

#### 4 Characterization of the volcanic aerosols

The VA characterization is made through the retrieval of their optical coefficients from nighttime Raman and daytime elastic lidar data, as well as through an estimate of the mass concentration of ash and non-ash particles. The fact that the lofted VA layers of interest in this study were optically very thin (in average  $AOT_{VA} < 0.020$  over all stations and over the whole period) induced lidar signals with very low signal-to-noise ratios which made the inversions quite difficult. For nighttime measurements the analysis was limited because in the optically thinnest lofted VA layers the Raman inversion yielded to large errors. After a careful review of all the good quality inversions available, three cases have been selected and are presented next (indicated in the time scale of Figs. 3 and 4 by gray bars):

- Raman inversion comparison between Évora and Madrid on 7 May at 01:00 UTC,
- Raman inversion at Granada on 8 May at 04:00 UTC, and
- Elastic inversion comparison between Granada and Barcelona on 8 May at 16:00 UTC.

The optical characterization is made in terms of aerosol extinction,  $\alpha$ , and backscatter,  $\beta$ , coefficients, lidar ratios at 355 nm,  $S_{355}$ , and at 532 nm,  $S_{532}$ , the  $\alpha$ -related Ångström exponent at 355/532 nm ( $AE_{355-532}^{\alpha}$ ) and the  $\beta$ -related Ångström exponents at 355/532 nm ( $AE_{355-532}^{\beta}$ ) and at 532/1064 nm ( $AE_{532-1064}^{\beta}$ ), as well as the linear particle depolarization ratio,  $\delta$ , when available. In this section  $AOT_{VA}$  was calculated as the integration of the extinction profiles when Raman measurements were available and otherwise as the integration of the backscatter profiles multiplied by a constant lidar ratio of 50 sr.

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## 4.1 Retrieval of the optical coefficients

### 4.1.1 7 May 2010, 01:00 UTC, Évora – Madrid

Those two measurements reflect some of the optically thickest layers observed during the period 5–8 May 2010 (see Fig. 4). The optical characterization is shown in Fig. 5. The VA layer in Évora is about 1-km thick and is located between 2.65 and 3.70 km.  $AOT_{VA}$  is 0.099, 0.070 and 0.050 at 355, 532 and 1064 nm, respectively. The bottom part of the optical coefficient ( $\alpha$  and  $\beta$ ) profiles is shown as reference. In Madrid the VA layer is observed roughly between 4.2 and 6.8 km and has an  $AOT_{VA}$  of 0.095 at 532 nm. For the sake of clarity the Évora profiles are not represented above 4.0 km and those of Madrid below 4.0 km. The numbers in the plots of the lidar ratio and Ångström exponent indicate the mean values plus standard deviation of the profiles in the VA layers. The lidar ratios retrieved in the VA layer in Évora are  $39 \pm 10$  and  $32 \pm 4$  sr at 355 and 532 nm, respectively, Those values are rather small compared to recent studies (Ansmann et al., 2010; Mona et al., 2011; Wiegner et al., 2011). According to the backtrajectories (Fig. 2) all the air masses arriving over Évora above 2500 m travelled above the Atlantic Ocean at altitudes well above 3000 m. In absence of emission sources along its way the VA plume probably did not mix with other aerosols during its transport. Therefore one possible explanation for the low values of  $S_{355}$  and  $S_{532}$  is that the air masses dehydrated along their path. This is suggested by Mona et al. (2011) who found that  $S_{355}$  decreases when the relative humidity decreases. In Madrid the lidar ratio at 532 nm is larger: its mean value is  $52 \pm 27$  sr. The backtrajectories arriving over Madrid above 3500 m are very similar to those arriving over Évora above 2500 m. The most probable hypothesis is that in Madrid between 4.2 and 6.8 km proportionally more small particles are present than in Évora (the convection due to the Iberian thermal low raises proportionally more small particles than large particles, see Sect. 3.2). Therefore small non-ash and small aged ash particles may dominate resulting in an increased contribution of non-ash particles which have a larger lidar ratio than ash par-

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ticles (Tesche et al., 2011; Ansmann et al., 2011). From Fig. 5, the mean  $AE_{355-532}^{\beta}$  and  $AE_{532-1064}^{\beta}$  in Évora are  $0.22 \pm 0.40$  and  $1.05 \pm 0.43$ , respectively. This indicates that the backscatter coefficient changes more sensitively at longer wavelengths. The mean  $\alpha$ -related Ångström exponent,  $0.68 \pm 0.63$ , is relative low. Such values are representative of rather medium-size particles. They range between very low values of 0.0 to 0.1 observed in Germany (Ansmann et al., 2010) and higher values of 1.0 to 1.4 observed in Italy (Mona et al., 2011).

#### 4.1.2 8 May 2010, 04:00 UTC, Granada

This measurement reflects also one of the optically thickest layers observed during the period 5–8 May 2010. The optical characterization is shown in Fig. 6. The VA layer is located between 2.6 and 2.9 km. The bottom part of the optical coefficient ( $\alpha$  and  $\beta$ ) profiles is shown as reference. The extinction and backscatter coefficient profiles reach values in the VA layers on the same order of magnitude than in the layer centered around 1.50–1.75 km which is supposedly a mixture of local aerosols and VA.  $AOT_{VA}$  is 0.106, 0.078 and 0.028 at 355, 532 and 1064 nm, respectively, which are quite high values compared to the rest of the period. The lidar ratios retrieved in the VA layer are  $47 \pm 7$  and  $48 \pm 16$  sr at 355 and 532 nm, respectively. Those values are higher than those observed in Évora the day before and are in agreement with former and ongoing VA studies (Ansmann et al., 2010; Mona et al., 2011; Pappalardo et al., 2011). According to Mona et al. (2011) a ratio of lidar ratio values around 1 suggests the presence of both aerosol types: non-ash and aged ash. Note, en passant, that aerosols such as mineral dust or fresh smoke can also have a ratio of lidar ratio around 1 (Alados-Arboledas et al., 2011); however such kind of particles are not present in the measurements considered here. If we consider that pure ash and non-ash depolarization ratios are 0.36 and 0.01, respectively, (Tesche et al., 2011; Ansmann et al., 2011; see Sect. 4.2) the mean particle depolarization ratio of 0.06 clearly confirms that both aerosol types are present. This value is much lower than what was observed

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in Germany (Ansmann et al., 2010) or in Italy (Mona et al., 2011) and suggests the coexistence of both aerosol types. Below 2.5 km (not shown) the particle depolarization ratio is around 0.045 which is a typical value at Granada when only local aerosols are present (Bravo-Aranda and Navas-Guzmán, 2011). However the backtrajectories below 2.5 km and the synoptic analysis (Sect. 3) indicated that VA are present and mixed with local aerosols in the lowermost layer. The mean value of 0.045 suggests then that the depolarization effect of the VA in the lowermost layer is also on the order of 0.045, which leads to a higher contribution of non-ash particles in the lowermost layer (see Sect. 4.2) than in the 2.6–2.9 km layer. From Fig. 6, the mean  $AE_{355-532}^{\beta}$  and  $AE_{532-1064}^{\beta}$  are  $0.73 \pm 0.07$  and  $1.68 \pm 0.31$ , respectively. This indicates again that the backscatter coefficient changes more sensitively at longer wavelengths. The similar values of 0.73–0.79 found for the  $\alpha$ - and the  $\beta$ -related Ångström exponents at 355/532 nm clearly reflects the fact that the lidar ratios at both wavelengths are nearly equal. The mean  $\alpha$ -related Ångström exponent,  $0.79 \pm 0.54$ , is similar to that in Évora the day before and is also representative of rather medium-size particles.

#### 4.1.3 8 May 2010, 16:00 UTC, Granada – Barcelona

In the afternoon of 8 May almost all the downward moving VA layers have mixed with the upward developing local PBL. Two lofted layers are still visible in Fig. 7: in Granada an extremely thin VA layer is observed between 3.00 and 3.35 km while in Barcelona a VA layer is visible around 2.2–2.9 km. For the sake of clarity the Granada profiles are not represented above 2.9 km and those of Barcelona below 3.0 km. The sun-photometer AOT and the fine mode fraction are, respectively around 0.15 and 80 % in Granada and 0.13 and 70 % in Barcelona. In Granada  $AOT_{VA}$  is 0.005 at 355 nm and 0.002 at 532 nm. At 1064 nm  $AOT_{VA}$  is less than 0.001. In Barcelona  $AOT_{VA}$  is higher: 0.025 at 355 nm and 0.010 at 532 nm. In both cases the AOT of the lofted VA represents a small fraction of the columnar AOT (between 1 and 7 %). Because those values are extremely low their interpretation should be done very cautiously.

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The backscatter coefficients reach very low values which do not exceed  $1 \text{ Mm}^{-1} \text{ sr}^{-1}$ . The  $\beta$ -related Ångström exponents at 355/532 nm oscillate around similar values at both sites ( $2.35 \pm 0.20$  and  $2.37 \pm 0.27$  in Granada and in Barcelona, respectively). This result indicates that the backscatter coefficient is highly wavelength-dependent at short wavelengths. In Granada the mean  $AE_{532-1064}^{\beta}$  ( $1.45 \pm 0.17$ ) and the mean particle depolarization ratio (0.075) are similar to their respective values at 04:00 UTC.

## 4.2 Estimate of the mass concentration

Mass concentration is one of the most critical parameters for airspace restrictions related to volcanic aerosol plumes. Shortly after the end of the Eyjafjallajökull eruption the UK Meteorological Office distinguished between 3 contamination levels: low ( $< 200 \mu\text{g m}^{-3}$ ), medium (200 to  $4000 \mu\text{g m}^{-3}$ ) and high ( $> 4000 \mu\text{g m}^{-3}$ ) (Schumann et al., 2011). At present  $2000 \mu\text{g m}^{-3}$  is considered as the maximum tolerable concentration for continuous flight operation.

Mass concentrations have been calculated from backscatter coefficients for ash and non-ash particles. The method used to distinguish between both types of particles is based on the work by Tesche et al. (2009) and refined recently for ash and fine-mode particles by Tesche et al. (2011) and Ansmann et al. (2011). The method uses the opposite depolarizing effects of ash particles (strongly depolarizing) and non-ash particles (very weakly depolarizing). Only the Granada system had a depolarization channel (at 532 nm) operative during the period 5–8 May 2010, so that profiles of mass concentration were only calculated for this station. For the sake of clarity the wavelength dependency of all coefficients has been omitted in this section. We remark that the method is applied under the assumption of external mixing only. As the Eyjafjallajökull eruption was sub-glacial, it is unlikely that internal mixing such as sulfur coating on ash occurred due to the high concentration of water vapor present in the ash cloud (Thomas and Prata, 2011).

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If we call  $\beta$  the total aerosol backscatter coefficient, the backscatter coefficients of ash,  $\beta_a$ , and non-ash particles,  $\beta_{na}$ , can be calculated, respectively as:

$$\beta_a = \beta \frac{(\delta - \delta_{na})}{(\delta_a - \delta_{na})} \frac{(1 - \delta_a)}{(1 - \delta)} \quad (1)$$

and

$$\beta_{na} = \beta \frac{(\delta - \delta_a)}{(\delta_{na} - \delta_a)} \frac{(1 - \delta_{na})}{(1 - \delta)} \quad (2)$$

where  $\delta_a$  and  $\delta_{na}$  represent the ash and non-ash linear particle depolarization ratios, respectively. Pure ash and pure non-ash particle depolarization ratios are reasonably well known as  $\delta_a = 0.36$  and  $\delta_{na} = 0.01$  (Ansmann et al., 2010; Groß et al., 2010; Tesche et al., 2011), respectively. The mass concentrations of ash,  $m_a$ , and non-ash particles,  $m_{na}$ , are given in terms of backscatter coefficient, respectively as:

$$m_a = \rho_a \frac{C_c}{AOT_c} \beta_a S_a \quad (3)$$

and

$$m_{na} = \rho_{na} \frac{C_f}{AOT_f} \beta_{na} S_{na} \quad (4)$$

where  $\rho_a$  and  $\rho_{na}$  are the ash and non-ash particle mass density, and  $S_a$  and  $S_{na}$  are the ash and non-ash particle lidar ratio. The two ratios,  $\frac{C_f}{AOT_f}$  and  $\frac{C_c}{AOT_c}$ , also called mean extinction-to-mass conversion factors represent the ratio of volume concentration to AOT for the fine (non-ash) and the coarse (ash) mode, respectively.

For pure ash and non-ash particles, respectively,  $\rho_a = 2.6 \text{ g cm}^{-3}$  (<http://volcanoes.usgs.gov/ash/properties.html#density>; Schumann et al., 2011) and  $\rho_{na} = 1.6 \text{ g cm}^{-3}$  (Bukowiecki et al., 2011), and  $S_a = 50 \pm 10 \text{ sr}$  and  $S_{na} = 60 \pm 20 \text{ sr}$  (Tesche et al., 2011; Ansmann et al., 2011). The ratios of volume concentration to AOT were calculated

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from AERONET-derived level 1.5 inversion products on 8 May 2010, at 06:35 and 07:00 UTC. The size distributions associated to those inversions are shown in Fig. 8. They are very similar and exhibit an enhanced fine (non-ash) mode. Note en passant that similar size distributions were obtained in Madrid by ground-based in-situ measurements (Revueña et al., 2011). This result is completely different to similar observations made in Germany at the beginning of the eruption (Ansmann et al., 2011) where the fine (non-ash) and coarse (ash) mode predominance were inverted, the enhanced mode being that of coarse (ash) particles, and where the volume concentration was higher by a factor 10. The mean extinction-to-mass conversion factors for the fine (non-ash) and the coarse (ash) modes are  $\frac{C_f}{AOT_f} = 0.255 \times 10^{-6} \text{ m}$  and  $\frac{C_c}{AOT_c} = 0.89 \times 10^{-6} \text{ m}$ , respectively.

In order to justify the use of the columnar values of the mean extinction-to-mass conversion factors in the VA layers, the non-ash and the ash AOT fractions are calculated for the lidar at 04:00 and 16:00 UTC and compared, respectively, to the fine (non-ash) and coarse (ash) mode fractions of the sun-photometer at 06:35 and 07:00 UTC. In this section the lidar-derived  $AOT_{VA}$  was calculated as:

$$AOT_{VA} = \int_{VA \text{ layer}} (\beta_{na} S_{na} + \beta_a S_a) . \quad (5)$$

Table 3 summarizes the values found for both instruments. Even though the AERONET AOT uncertainty is known to be  $\leq \pm 0.01$  for wavelengths greater than 440 nm (Holben et al., 1998; Dubovik et al., 2000), here the AOT at 500 nm is expressed with three digits in order to minimize differences due to truncation in the calculation of the fine and coarse mode fractions. For the same reason the lidar-derived  $AOT_{VA}$  is expressed with four digits. The VA layers at 04:00 and 16:00 UTC are found around 2.6–2.9 km and 3.00–3.35 km, respectively. In those layers, the ratio of lidar-derived non-ash AOT to  $AOT_{VA}$  is nearly constant between both lidar measurements around 80–82 %. In the atmospheric column the sun-photometer fine (non-ash) mode fraction is around 87–89 %. Given the relatively good agreement between those numbers and besides the

lack of size distribution measurements in the VA layers, the above-mentioned method can be further applied to our lidar measurements.

The results are shown in Fig. 9 for the VA layers identified at 2.6–2.9 km at 04:00 UTC and at 3.00–3.35 km at 16:00 UTC. As a consequence of the low particle depolarization ratios observed around 0.06–0.075 (see Figs. 6 and 7) compared to Central Europe (Ansmann et al., 2010, 2011; Tesche et al., 2011) or Southeastern Europe (Mona et al., 2011) the ash and non-ash particle mass concentrations appear approximately in the same order of magnitude. They roughly fluctuate in the range 60–180  $\mu\text{g m}^{-3}$  at 04:00 UTC and 1.5–3.5  $\mu\text{g m}^{-3}$  at 16:00 UTC. In the afternoon the mass concentration is around 30 to 50 times smaller than what is observed at 04:00 UTC. Those proportions are also reflected by the difference observed between both  $\text{AOT}_{\text{VA}}$ . It is worth noting that the  $\text{AOT}_{\text{VA}}$  at Granada reaches the highest values of the period in the night between 7 and 8 May (see Fig. 4) without a significant change neither in the thickness of the layers nor in the particle depolarization ratio. Thus it leads to high values of the backscatter coefficient and also of the mass concentration. It is quite probable that the mass concentration of neither the ash nor the non-ash particles found in lofted VA layers exceeded the value of 200  $\mu\text{g m}^{-3}$  in Granada in the period 5–8 May 2010.

## 5 Conclusions

During the Eyjafjallajökull eruption the strongest intrusion of volcanic aerosols in the Iberian Peninsula occurred during 5–8 May 2010. Volcanic aerosols were first observed at the westernmost lidar station in Évora. Lofted VA layers showed a downward motion from 6 May onwards. The mean AOT of those layers was rather low (between 0.013 and 0.020 in all stations over the whole period) with a peak on 7 May. Even though the thickness of the VA layers was spatially and temporally quite variable, rather thin layers (< 1000 m) with a top height up to 11–12 km were observed. A significant increase in the total AOT, as well as in the fine mode AOT, is observed along the intrusion which indicates an increase of the aerosol load of rather small size in the PBL since

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the lofted VA layers optical thickness did not change significantly during the intrusion. The backtrajectory analysis which shows transport time from Iceland on the order of 3 days at the beginning of the period and on the order of 5 days at the end corroborates this result: the size of the lofted VA decreases with increasing age. Contrarily to lidar stations from Northern and Central Europe that detected optically very thick lofted VA layers most of the VA that reached the IP were already coupled to the PBL.

Lidar ratios at different sites varied between 30 and 50 sr without a marked spectral dependency between 355 and 532 nm. Values near 50 sr were observed on 7 May over Madrid in high VA layers above 4 km ( $AOT_{VA} = 0.095$  at 532 nm) and on 8 May over Granada in VA layers between 2.5 and 3.0 km ( $AOT_{VA} = 0.078$  at 532 nm). Smaller values between 30 and 40 sr were observed over Évora on 7 May in a VA layer between 2.5 and 4.0 km ( $AOT_{VA} = 0.070$  at 532 nm). Similar extinction-related Ångström exponents varying between 0.6 and 0.8 were observed in Évora and Granada. In terms of backscatter-related Ångström exponents an increase is observed between sites with time: e.g. at 355/532 nm, 0.22 is measured in Évora on 7 May, 0.73 is measured in Granada on 8 May (morning), and  $\sim 2.35$  is measured in Granada and Barcelona on 8 May (afternoon). This result points out a possible decrease of the volcanic particle size as the plume moves from west to east. The relatively low linear particle depolarization ratio at 532 nm measured in the VA layers in Granada around 0.065–0.075 further indicates the coexistence of ash and non-ash particles.

An estimate of the mass concentration of ash and non-ash particles has been performed in Granada by using the opposite depolarizing effects of ash particles (strongly depolarizing) and non-ash particles (very weakly depolarizing), and sun-photometer observations. In the morning of 8 May both the ash and non-ash mass concentration varied between 60 and  $180 \mu\text{g m}^{-3}$  in a VA layer with an optical thickness of 0.101. Later the mass concentration varied between 1.5 and  $3.5 \mu\text{g m}^{-3}$  in a VA layer with an optical thickness of 0.0025. In both cases the ash mass concentration was approximately 1.5 higher than the non-ash mass concentration. Given that the case selected in the morning of 8 May had one of the highest optical thicknesses it is quite probable

that the mass concentration of neither the ash nor the non-ash particles found in lofted VA layers exceeded the value of  $200 \mu\text{g m}^{-3}$  in Granada in the period 5–8 May 2010.

## Appendix A

5 The uncertainty of  $\text{AOT}_{\text{VA}}$  can be deduced from the uncertainty of the backscatter coefficient profiles. There are two terms to consider:

- The systematic error ( $d\beta_{\text{syst}}$ ): the sum of the error due to a range-dependent lidar ratio and the error due to an erroneous backscatter coefficient at the calibration height;
- 10 – The statistical error ( $d\beta_{\text{stat}}$ ) due to the observation noise.

On the one hand, the systematic errors,  $d(\text{AOT}_{\text{VA}})_{\text{syst}}$ , are correlated with range, so that the variance associated to  $\text{AOT}_{\text{VA}}$  due to systematic errors falls like  $1/N^2$  (Barlow, 1989), being  $N$  the number of samples in the VA layer:

$$V(\text{AOT}_{\text{VA}})_{\text{syst}} = \frac{[d(\text{AOT}_{\text{VA}})_{\text{syst}}]^2}{N^2} \quad (\text{A1})$$

15 and

$$d(\text{AOT}_{\text{VA}})_{\text{syst}} = N \cdot \sqrt{V(\text{AOT}_{\text{VA}})_{\text{syst}}} = N \cdot \Delta z \cdot S \cdot \sqrt{V(\beta_{\text{syst}})} \quad (\text{A2})$$

where  $\Delta z$  represents the range resolution and  $S$  the lidar ratio. If we call  $d\beta_{\text{syst}}^{\text{max}}$  the maximum of  $d\beta_{\text{syst}}$  in the VA layer, then a conservative approximation of the uncertainty of  $\text{AOT}_{\text{VA}}$  due to systematic errors can be written as:

$$20 \quad d(\text{AOT}_{\text{VA}})_{\text{syst}} \leq N \cdot \Delta z \cdot S \cdot d\beta_{\text{syst}}^{\text{max}} \quad (\text{A3})$$

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On the other hand, the statistical error,  $d(\text{AOT}_{\text{VA}})_{\text{stat}}$ , is uncorrelated with range, so that the variance associated to  $\text{AOT}_{\text{VA}}$  due to the statistical error falls now like  $1/N$  (Barlow, 1989):

$$V(\text{AOT}_{\text{VA}})_{\text{stat}} = \frac{[d(\text{AOT}_{\text{VA}})_{\text{stat}}]^2}{N} \quad (\text{A4})$$

5 and

$$d(\text{AOT}_{\text{VA}})_{\text{stat}} = \sqrt{N} \cdot \sqrt{V(\text{AOT}_{\text{VA}})_{\text{stat}}} = \sqrt{N} \cdot \Delta z \cdot S \cdot \sqrt{V(\beta_{\text{stat}})}. \quad (\text{A5})$$

Again if we call  $d\beta_{\text{stat}}^{\text{max}}$  the maximum of  $d\beta_{\text{stat}}$  in the VA layer, a conservative approximation of the uncertainty of  $\text{AOT}_{\text{VA}}$  due to the statistical error can be written as:

$$d(\text{AOT}_{\text{VA}})_{\text{stat}} \leq \sqrt{N} \cdot \Delta z \cdot S \cdot d\beta_{\text{stat}}^{\text{max}}. \quad (\text{A6})$$

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 25 because of the closure of the Barcelona airspace.

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**Table 1.** Some characteristics of the lidar systems involved in the study.

Station Country	Évora Portugal	Madrid Spain	Granada Spain	Barcelona Spain
Elastic wavelengths (nm)	1064, 532, 355	532	1064, 532, 355	532, 355
Raman wavelengths (nm)	607, 387	607	607, 387, 408	–
Depolarization	No	No	at 532 nm	No
Raw resolution (m)	30	3.75	7.5	7.5

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**Table 2.** Mean optical (at 532 nm) and spatial thickness of the lofted VA layers at the 4 stations from 5 to 8 May 2010. The minimum and maximum values (min, max) of  $AOT_{VA}$  and  $\Delta h_{VA}$  are also reported.

Station		5 May	6 May	7 May	8 May	5–8 May
Évora	$\overline{AOT_{VA}}$	0.008	0.009	0.025	–	0.013
	(min, max)	(< 0.002, 0.022)	(< 0.002, 0.065)	(0.002, 0.107)	–	(< 0.002, 0.107)
	$\Delta h_{VA}$ (m)	1104	543	360	–	614
	(min, max)	(179, 3885)	(60, 2332)	(89, 777)		(60, 3885)
Madrid	$\overline{AOT_{VA}}$	0.034	0.008	0.070	0.004	0.020
	(min, max)	(< 0.002, 0.016)	(0.004, 0.130)	(0.070, 0.070)	(0.004, 0.004)	(< 0.002, 0.130)
	$\Delta h_{VA}$ (m)	4240	1000	2250	2105	2098
	(min, max)	(750, 6600)	(600, 1500)	(2250, 2250)	(2105, 2105)	(600, 6600)
Granada	$\overline{AOT_{VA}}$	0.007	0.004	0.026	0.027	0.020
	(min, max)	(< 0.002, 0.016)	(< 0.002, 0.021)	(< 0.002, 0.137)	(< 0.002, 0.106)	(< 0.002, 0.137)
	$\Delta h_{VA}$ (m)	422	325	412	343	382
	(min, max)	(262, 705)	(82, 1147)	(82, 1230)	(113, 667)	(82, 1230)
Barcelona	$\overline{AOT_{VA}}$	–	–	–	0.015	0.015
	(min, max)	–	–	–	(0.013, 0.017)	(0.013, 0.017)
	$\Delta h_{VA}$ (m)	–	–	–	950	950
	(min, max)	–	–	–	(700, 1200)	(700, 1200)

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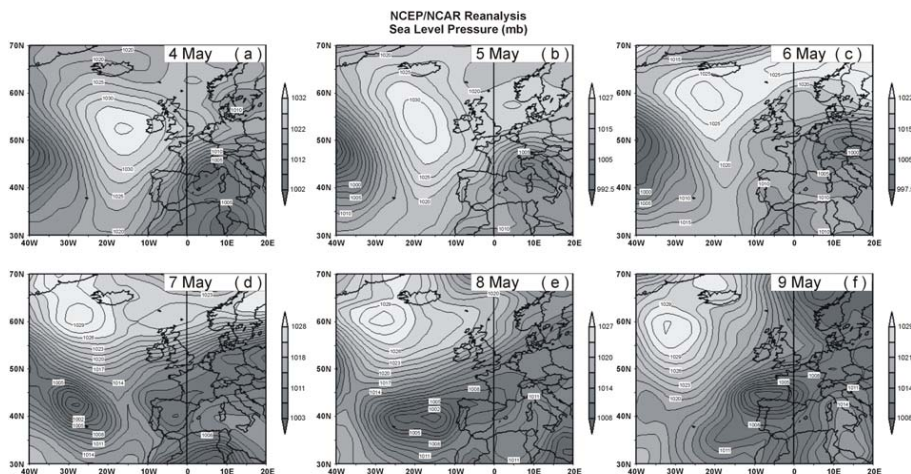
**Table 3.** Sun-photometer total, fine and coarse mode AOT at 500 nm and lidar-derived total, non-ash and ash AOT at 532 nm at Granada on 8 May 2010. The numbers in parenthesis represent the fraction of AOT of the mode considered to the AOT total (AOT for the sun-photometer and  $AOT_{VA}$  for the lidar).

	AOT	$AOT_f$	$AOT_c$	$AOT_{VA}$	$\int_{VA \text{ layer}} \beta_{na} S_{na}$	$\int_{VA \text{ layer}} \beta_a S_a$
Lidar 04:00 UTC 2.6–2.9 km				0.101	0.083 (82 %)	0.018 (18 %)
Sun-phot. 06:35 UTC	0.376	0.334 (89 %)	0.042 (11 %)			
Sun-phot. 07:00 UTC	0.358	0.313 (87 %)	0.045 (13 %)			
Lidar 16:00 UTC 3.00–3.35 km				0.0025	0.002 (80 %)	0.0005 (20 %)



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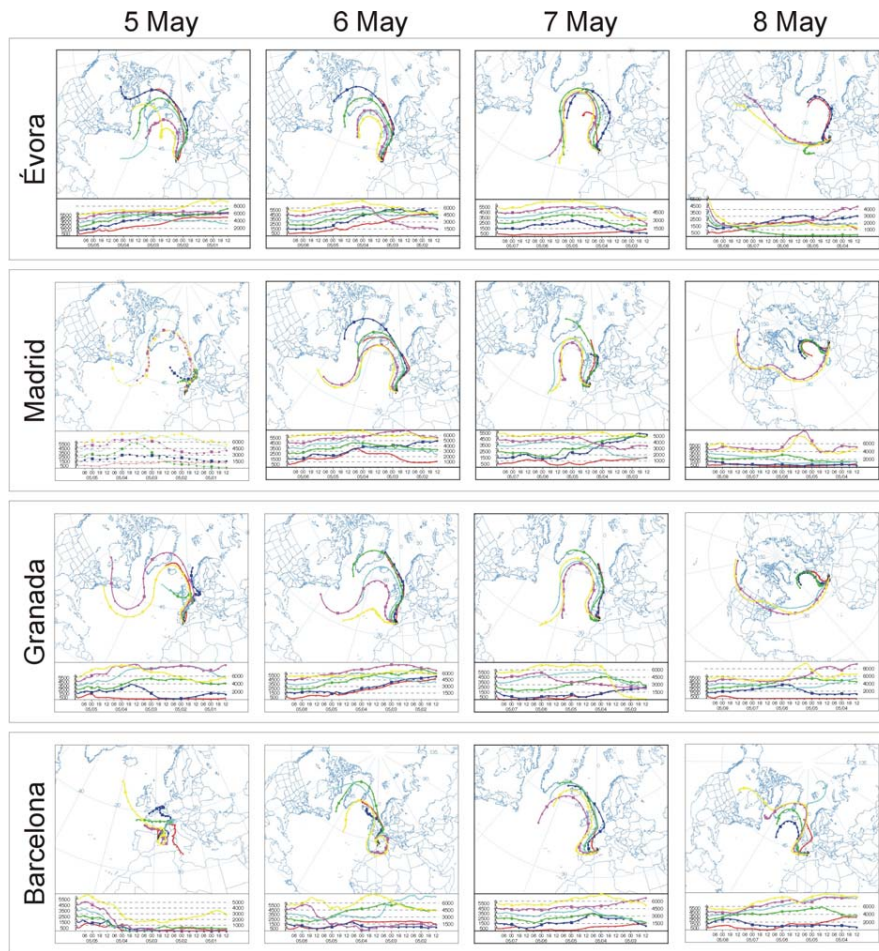


**Fig. 1.** Synoptic situation at 12:00 UTC on (a) 4, (b) 5, (c) 6, (d) 7, (e) 8 and (f) 9 May 2010. Dark and light colors represent low and high pressures, respectively.

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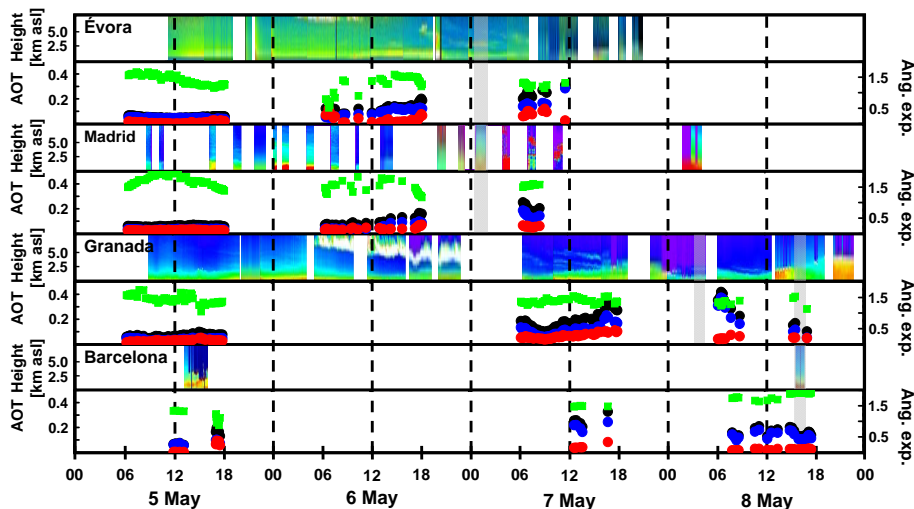


**Fig. 2.** 120-h (5 days) backtrajectories arriving in Évora, Madrid, Granada and Barcelona at 12:00 UTC on 5, 6, 7 and 8 May 2010 at 500, 1500, 2500, 3500, 4500 and 5500 m a.g.l.

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**Fig. 3.** Evolution of the lidar range-square corrected signal from 5 to 8 May 2010, at Évora, Madrid, Granada and Barcelona. Below the lidar signal time series the sun-photometer total (black circles), coarse mode (red circles) and fine mode (blue circles) at 500 nm are reported. The mean Ångström exponent is reported as green squares (right axis). The vertical gray bars indicate the time periods discussed in Sect. 4.

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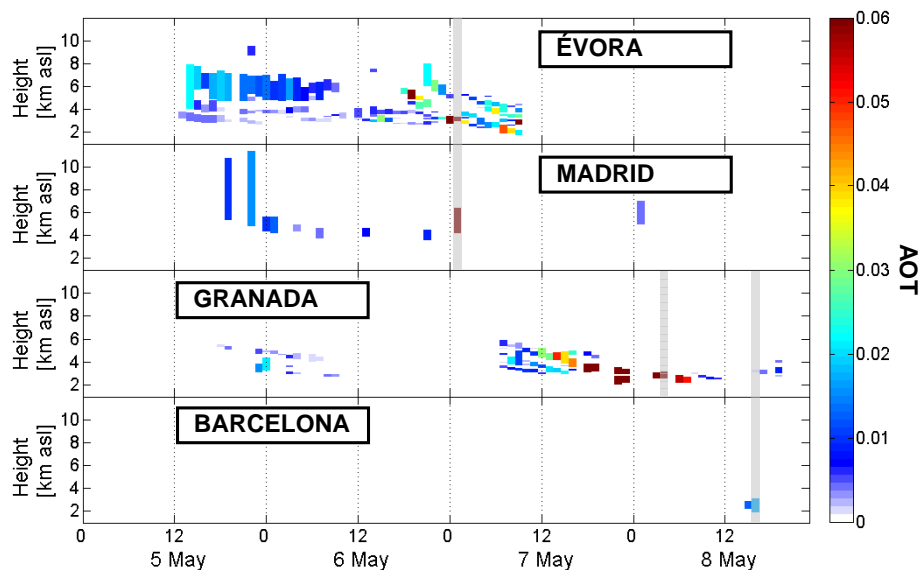
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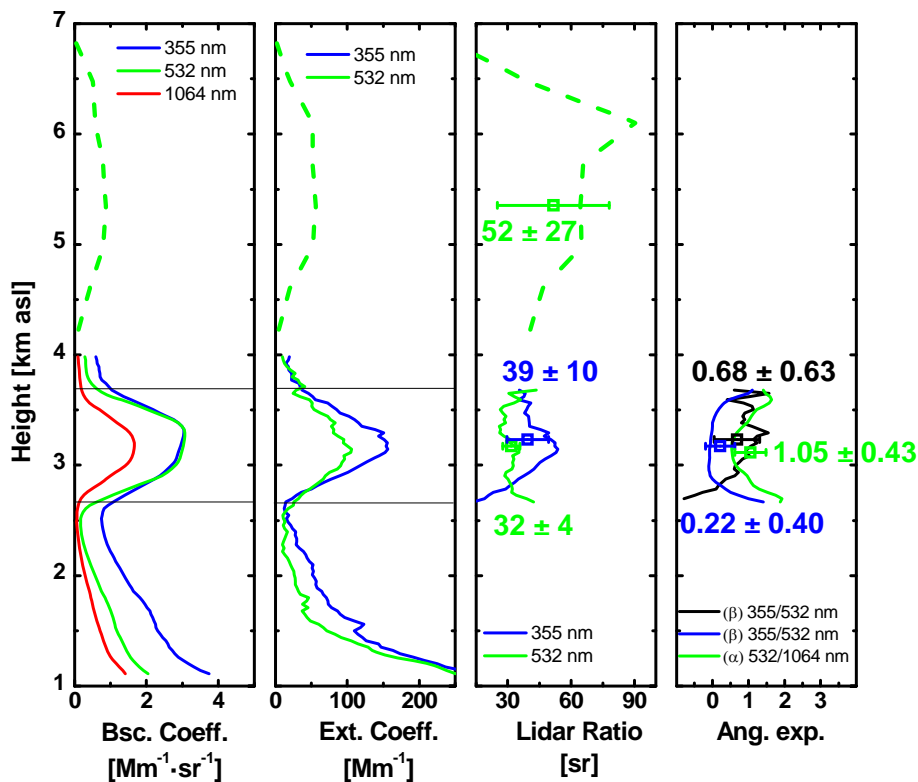


**Fig. 4.** AOT of lofted VA layers at 532 nm versus height from 5 to 8 May 2010. The vertical gray bars indicate the time periods discussed in Sect. 4.

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**Fig. 5.** Volume backscatter and extinction coefficients, lidar ratios, and  $\beta$ - (blue and green curves) and  $\alpha$ -related (black curve) Ångström exponents on 7 May 2010 at 01:00 UTC at Évora (solid lines) and Madrid (dash lines).

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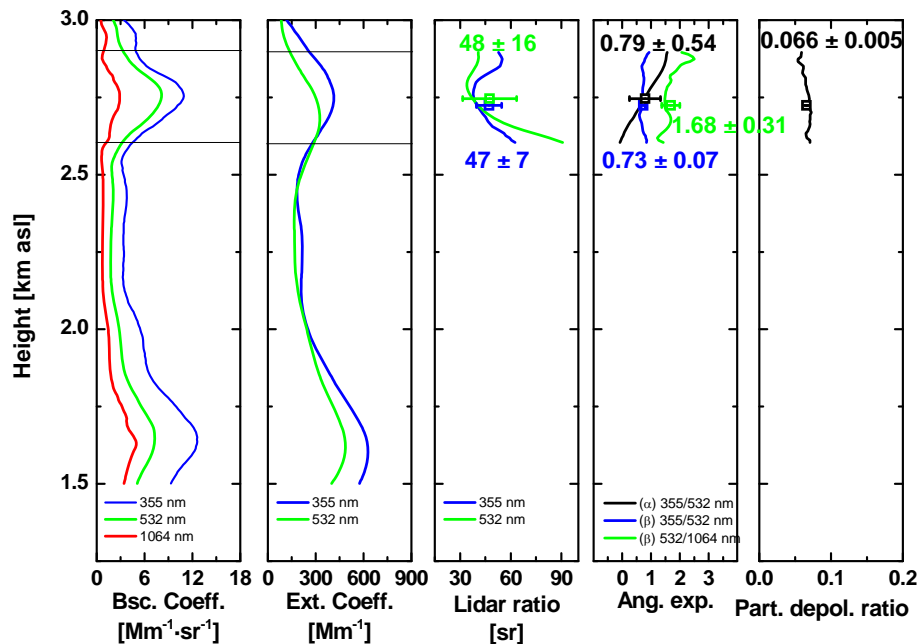
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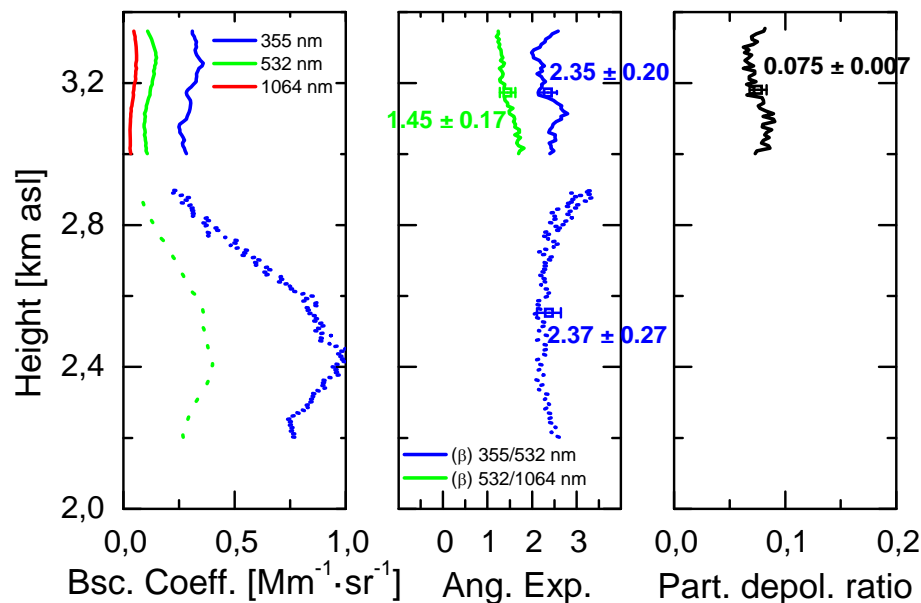


**Fig. 6.** Volume backscatter and extinction coefficients, lidar ratios,  $\beta$ - (blue and green curves) and  $\alpha$ -related (black curve) Ångström exponents and linear particle depolarization ratio on 8 May 2010 at 04:00 UTC at Granada.

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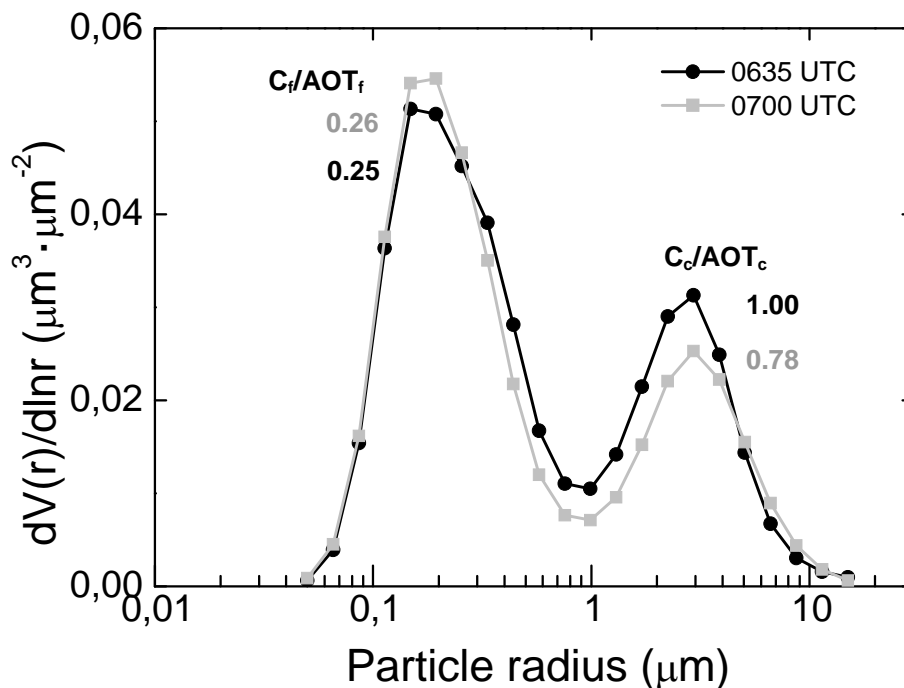


**Fig. 7.** Volume backscatter coefficients,  $\beta$ -related Ångström exponents and linear particle depolarization ratio on 8 May 2010 at 16:00 UTC at Granada (solid lines) and Barcelona (dot lines).

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**Fig. 8.** AERONET size distributions at Granada on 8 May 2010, at 06:35 and 07:00 UTC. The ratio of volume concentration to AOT for the fine (non-ash) and the coarse (ash) mode are indicated in black at 06:35 UTC and in gray at 07:00 UTC.

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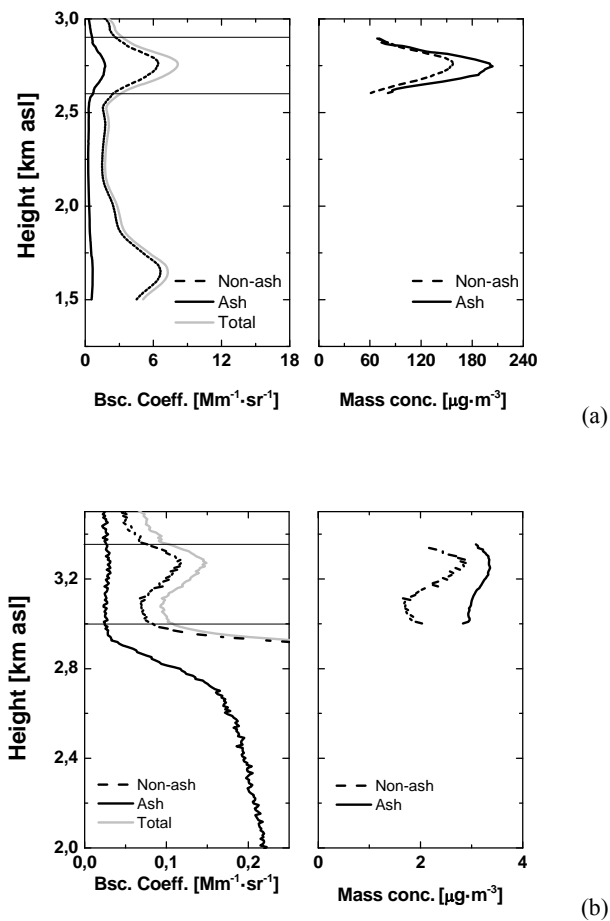
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**Fig. 9.** Mass concentration profiles of ash and non-ash particles at Granada on 8 May 2010, at (a) 04:00 and (b) 16:00 UTC.