

Long range transport and mixing of aerosol sources during the 2013 North American biomass burning episode: analysis of multiple lidar observations in the Western Mediterranean basin

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Abstract. Long range transport of biomass burning (BB) aerosols between North America and the Mediterranean region took place in June 2013. A large number of ground based and airborne lidar measurements were deployed in the Western Mediterranean during the Chemistry-AeRosol Mediterranean EXperiment (ChArMEx) intensive observation period. A detailed analysis of the potential North American aerosol sources is conducted including the assessment of their transport to Europe using forward simulations of the FLEXPART Lagrangian particle dispersion model initialized using satellite observations by MODIS and CALIOP. The three dimensional structure of the aerosol distribution in the ChArMEx domain observed by the ground-based lidars (Menorca, Barcelona and Lampedusa), a Falcon-20 aircraft flight and three CALIOP tracks, agree very well with the model simulation of the three major sources considered in this work: Canadian and Colorado fires, a dust storm from Western US and the contribution of Saharan dust streamers advected from the North Atlantic trade wind region into the Westerlies region. Four aerosol types were identified using the optical properties of the observed aerosol layers (aerosol depolarization ratio, lidar ratio) and the transport model analysis of the contribution of each aerosol source: (I) pure BB layer, (II) weakly dusty BB, (III) significant mixture of BB and dust transported from the trade wind region (IV) the outflow of Saharan dust by the subtropical jet and not mixed with BB aerosol. The contribution of the Canadian fires is the major aerosol source during this episode while mixing of dust and BB is only significant at altitude above 5 km. The mixing corresponds to a 20%-30% dust contribution in the total aerosol backscatter. The comparison with the MODIS AOD horizontal distribution during

20 this episode over the Western Mediterranean sea shows that the Canadian fires contribution were as large as the direct northward dust outflow from Sahara.

1 Introduction

Forest fires are a significant source of tropospheric aerosol particles at northern latitudes in Spring and Summer (Generoso et al., 2003; Warneke et al., 2009) and many studies project higher temperatures and longer growing season e.g. (Flannigan et al., 2009; Liu et al., 2014). The focus of biomass burning emission impact on the atmospheric composition is often on the effect of these fires on the aerosol distribution in North America and Siberia (Eck et al., 2009; Warneke et al., 2010). Long range transport of biomass burning plumes has been also recognized as a significant source of aerosol in the mid-latitude free troposphere over Europe (Müller et al., 2005; Fiebig et al., 2003; Sciare et al., 2008; Adler et al., 2011). Air mass aging related to long range transport also leads to aerosol optical and chemical properties different from results obtained when looking at observations close to the fire region (Liousse et al., 1995; Müller et al., 2007; Bougiatioti et al., 2014). As an example, the absorbing efficiency in the visible spectral range is known to significantly increase in case of internally mixed BC (coating with secondary compounds) compared to externally mixed BC (Schneider et al., 2005). So far little attention has been paid to the frequent mixing of dust and biomass burning (BB) aerosol occurring during their transatlantic long range transport while lidar data analysis has shown that such a mixing will likely modify the extinction to backscatter ratio often called Lidar Ratio (LR) and then the aerosol optical depth (AOD) (Cattrell et al., 2005; Gross et al., 2011). Results of Paris et al. (2010) also show that the solubility of iron is enhanced by the mixing with biomass burning aerosols, while aerosol deposition may influence the rate of nitrogen fixation by microorganisms, and subsequently the global carbon cycle (Guieu et al., 2014). Although episodic, such long-range transport of smoke aerosols over the Mediterranean can also impact the regional energy budget by changing the distribution of solar energy. Indeed, for an aged BB plume, Formenti et al. (2002) report a net shortwave radiative forcing over the sea (daytime average) up to -64 W m^{-2} , at the surface and up to -22 W m^{-2} , at the top of the atmosphere (for an AOD of 0.40 at 550 nm). The large concentration of absorbing material (BC particles) within smoke plumes leads to significant absorption of solar radiations within the atmospheric layer where smoke resides, that could perturb the relative humidity and temperature vertical profiles. In the framework of the Chemistry-Aerosol Mediterranean Experiment/Aerosol Direct Radiative Impact in the Mediterranean (ChArMEx/ADRIMED) experimental campaign, many aerosol lidar and aircraft measurements have been made in June-July 2013 in the Mediterranean region during a case of intense biomass burning transport from North America to Europe (Mallet et al., 2016; Chazette et al., 2015; Pelon et al., 2015). Only a few studies report such long-range transport observations from North America to Europe (Forster et al., 2001; Petzold et al., 2007) or even the eastern Mediterranean (Formenti et al., 2002).

55 The purpose of this paper is to analyze the transatlantic long range transport of BB and dust
aerosol sources from North America during this period. The context of our study is described in sec-
tion 2 by describing the main characteristics of the summer 2013 BB episode in North America and
the observation network considered for the analysis of the aerosol distribution in the Mediterranean
region. The aerosol sources are identified using satellite observations and the transport of dust or
60 BB plumes is calculated with the FLEXPART Lagrangian model (see section 3). The aerosol lidar
observations are discussed in section 4, where the contribution of the different aerosol sources is
assessed using the comparison of the spatial distribution of the layers with the FLEXPART model
simulations (forward from the sources region and backward to calculate the potential emission sensi-
tivity for each observed aerosol layers). The mixing between dust and BB plumes is mainly derived
65 from the analysis of the aerosol layer optical properties. The Menorca and aircraft lidar observations
during ChArMEx are thoroughly described in a companion paper (Chazette et al., 2015) submitted
with this paper and in a paper in preparation by J.Pelon and coworkers.

2 Context

2.1 The 2013 North American biomass burning period

70 June 2013 was on the drier side in the USA High Plains Region with most areas receiving less
than 70% of normal precipitation. It was especially dry for most of Colorado and Wyoming which
received less than 50% of normal precipitation and many locations in the western areas of those
states received little to no precipitation. As a consequence many fires took place in North America.
Fire started in Colorado State on 10 June and lasted until 22 June 2013 (Colorado HSEM, 2013).
75 Two large fires burning in Southern Colorado even produced pyrocumulonimbus clouds and very
large smoke plumes on 19 and 20 June 2013 in the West Fork Complex, and in the East Park.

In Canada there have been also many fires (334) during the period 13 to 26 June 2013 burning
632,000 ha. The seasonal fire occurrence was below average while the area burned was more than
twice the 10-year average, due to large fires burning in Quebec. The majority of fires were spread
80 between Manitoba, Alberta, Yukon and North Western Territories and Quebec, while 75% of the
area burned was in Quebec and 20% in Manitoba (CIFFC, 2013). The total amount of area burned
was around 500,000 ha for the period 12 to 25 June 2013, i.e. more than twice the 10-year summer
average for the same period. The East Canadian fires at -80°W and -100°W took place during 4-6
days between 18 and 24 June while the fires west from -120°W took place during 2-3 days starting
85 on 17 June in Alaska and 22 June in the Mackenzie mountains.

2.2 The 2013 Mediterranean lidar observation network

During ChArMEx an intensive observation period took place in Western Mediterranean region from
11 June to 5 July 2013 (SOP-1a) when airborne measurements were made by two aircraft (ATR42

and F20) and ground based observations at 4 sites in Lampedusa, Corsica, Barcelona and Menorca
90 (Mallet et al., 2016). During ChArMEx, aerosol backscatter vertical profiles were made by airborne
and ground based lidar systems which provide a very good opportunity to characterize the verti-
cal distribution of the North American BB plume over the Mediterranean region. The map of the
ChArMEx lidar observation network is shown in Fig.1. The Falcon 20 aircraft was equipped with
an airborne lidar LNG (Pelon et al., 2002) providing attenuated backscatter vertical profiles at three
95 wavelength (1064, 532 and 355 nm). It was based in Cagliari, Sardinia. The LNG lidar has been
mainly used with a downward looking mode. Two tracks have been made in late June during the
passage of the BB plume over the Western Mediterranean: a transect between Cagliari and Menorca
on 27 June 2013 and a loop around Sicily on 28 June. Only the loop on 28 June is considered in
this work because the 27 June data will be discussed in a future paper by J. Pelon and co-workers on
100 the airborne observations during ChArMEx. The ground based lidar are located in Menorca (40°N,
4°E), Barcelona (41.4°N, 2°E) and Lampedusa (35.5°N, 12.5°E). The Menorca lidar works at 355
nm, while the Barcelona and Lampedusa lidar measure the atmospheric backscatter signal at 532
nm. The ground based lidar systems are respectively described in Chazette et al. (2014), Kumar
et al. (2011), Di Iorio et al. (2009). The airborne lidar LNG was also run every morning in Cagliari
105 (39°N, 9°W) from 24 to 30 June 2013 pointing upward from the surface. All the lidars can record
also the depolarization ratio between the signal polarized parallel and perpendicular to the plane of the
outgoing beam. For the ground based lidar, the uncertainty on the aerosol depolarization ratio is or
the order 1-2% as explained in Chazette et al. (2012). For the airborne lidar LNG, the depolarization
ratio is measured at 355 nm and it is calibrated on molecular scattering using a value of $1.5 \pm 0.3\%$
110 for clean air, corresponding also to 1-2% error on the aerosol depolarization ratio.

In addition to ground based and airborne lidar, the observations of the spaceborne Cloud-Aerosol
Lidar with Orthogonal Polarization (CALIOP) are known to be very useful to track aerosol plumes
(Winker et al., 2009). Three CALIOP night-time tracks shown in Fig.1 on 27 and 28 June 2013 are
ideally located above the ChArMEx area when the BB plume is expected over Europe.

115 **3 Aerosol sources and transport**

3.1 Methodology

Satellite remote sensing were considered for the BB aerosol sources identifications: both Moderate
Resolution Imaging Spectroradiometers (MODIS) on Terra and Aqua platforms, and CALIOP. The
distribution of the fires was taken from the NASA Fire Information for Resource Management Sys-
120 tem (FIRMS) which provides the analysis of the MODIS hot spots in terms of fire radiative power
(FRP) given in MW. Only fire areas with $FRP > 0.8$ GW are included in this analysis. The MODIS
0.5 μm AOD daily product is also considered to estimate the horizontal extent of BB plume when
large AOD > 0.3 is found near the spots with elevated FRP. Both MODIS instruments on Aqua and

Terra are considered to derive the daily mean. The mean error on the MODIS AOD daily product is
125 0.03 with a root mean square error of 0.14 according to Ruiz-Arias et al. (2013). When a CALIOP
overpass is found near the MODIS BB plume, the lidar vertical cross section is used to specify the
vertical extent of the MODIS BB plume.

For the dust aerosol sources, two main information sources were considered: (i) North American
dust storms identified in the NAAPS (Navy Aerosol Analysis and Prediction System) Global Aerosol
130 Model simulations and (ii) $0.5 \mu\text{m}$ AOD anomalies from the MODIS daily products. AOD streamers
transported from the tropical Atlantic belt of elevated $0.5 \mu\text{m}$ AOD to the mid-latitudes are related
to the transport of Saharan dust across the Atlantic. CALIOP overpasses near the AOD anomalies
again provide the vertical extent of dust aerosol layers.

In this work we use the new CALIOP level-1 (L1) version 4.0 attenuated backscatter coefficients
135 β_{1064} and β_{532} because they correspond to a better calibration of the lidar data. They are averaged
using a 10 km horizontal resolution and a 60 m vertical resolution (Vaughan et al., 2012). Before
making horizontal or vertical averaging, the initial 333 m horizontal resolution (1 km above the
altitude 8.2 km) are filtered to remove the cloud layer contribution (Winker et al., 2009). This cloud
mask makes use of the Version 3 level-2 (L2) cloud layer data products (Vaughan et al., 2009). Our

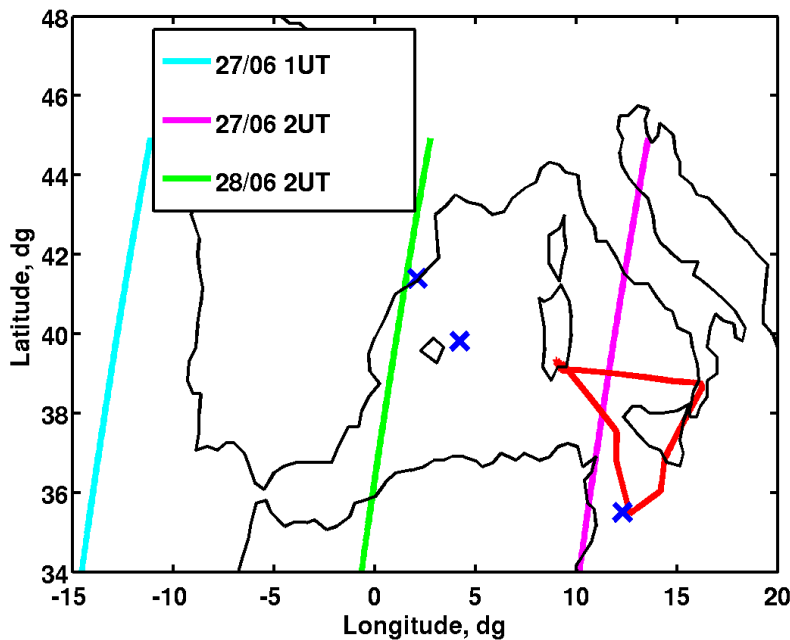


Figure 1. Map of the ChArMEx lidar observations. The colored vertical line are the positions of the nighttime CALIOP tracks on 27 and 28 June 2013. The red thick line shows the loop followed by the Falcon 20 aircraft on 28 June 2013 from 13 UT to to 15 UT, while the blue crosses are for the Menorca, Barcelona and Lampedusa ground based lidar.

140 scheme for distinguishing cloud and aerosol is described in Ancellet et al. (2014). Although the
lidar ratio (LR) is available from the CALIOP Version 3 L2 aerosol layer data products, it is often
based on an aerosol classification algorithm (Omar et al., 2009). In our work the lidar ratio (LR)
is recalculated by using the aerosol layer transmittance and the integrated attenuated backscatter in
the aerosol layer following the method described in Young (1995). To reduce the error when using
145 high horizontal resolution CALIOP profiles, β_{532} is averaged over 80 km to compute the plume
transmittance whenever it is possible. The attenuated backscatter is then corrected for the molecular
and aerosol attenuation using a forward Fernald inversion (Fernald, 1984) before calculating the
backscatter ratio $R(z) = (\beta_a + \beta_R)/\beta_R$ at 532 nm and 1064 nm using the CALIOP atmospheric
density model to calculate the β_R Rayleigh backscatter vertical profiles. The aerosol depolarization
150 ratio δ_{532} is also calculated using the perpendicular- to the parallel plus perpendicular polarized
aerosol backscatter coefficient. The calibration of the relative ratio between the two 532 nm channels
is based on regular use of a pseudo depolarizer located ahead of the beamsplitter which separates the
signal polarized parallel and perpendicular to the plane of the outgoing beam (Winker et al., 2009).
We have also derived the color ratio defined as the ratio of the aerosol backscatter coefficients at 1064
155 and 532 nm ($C_a(z) = \beta_{a1064}/\beta_{a532} = (R_{1064}(z) - 1)/[16(R_{532}(z) - 1)]$). The aerosol color ratio
can be also written as $C_a(z) = 2^{-k}$, where k is an exponent depending on the aerosol microphysical
properties (Cattrell et al., 2005). The exponent k varies from 0 to 2 when increasing the fine mode
aerosol contribution. These two ratios are provided only for $R(z) > 1.3$ because the uncertainty on
the depolarization and color ratios are large for weak aerosol layers. Whenever it is possible, the use
160 of nighttime overpasses are preferred to improve the signal-to-noise ratio (SNR).

The transport of the aerosol sources is analyzed using the FLEXPART model version 8.23 (Stohl
et al., 2002) driven by 6-hourly ECMWF analysis (T213L91) interleaved with operational forecasts
every 3 hours. The model is run using a forward simulation with a tracer released within a volume
estimated from the satellite observations. The release time period ranges from 1-3 days according to
165 the MODIS AOD observations. The total mass of the tracer emitted is estimated using the aerosol
concentration given in the NAAPS Global Aerosol Model simulations and FLEXPART calculates the
gridded tracer concentration in $\text{ng}\cdot\text{m}^{-3}$. Considering the uncertainty in the estimate of the emitted
tracer mass, the tracer distribution in the ChArMEx domain is analyzed using a relative mass fraction
between the emitted mass and the calculated mass within the model grid cell. A factor is applied to
170 calculate this ratio in order to take into account on one hand the difference between the emission
volume ($\approx 5 \cdot 10^5 \text{ km}^3$) and the grid cell volume of the tracer concentration field ($\approx 2 \cdot 10^3 \text{ km}^3$), and
on the other hand, the time difference between the emission period (1-3 days) and the integration time
(6 h) used for the calculation of the tracer gridded concentration. The relative mass fraction is 100%
when the air mass is advected above the $0.5^\circ \times 0.5^\circ$ grid cell chosen for the gridded concentration
175 calculation, without dilution (< 100%) or concentration (> 100%) of the tracer.

3.2 North American biomass burning aerosol

The MODIS FRP distributions are plotted in Fig.2 from 17 to 25 June 2013 showing the 6 main fire regions over Canada and Colorado. The map of the $0.5 \mu\text{m}$ daily AOD MODIS also show aerosol plumes on 22 June near Hudson Bay, Colorado and over the Atlantic ocean where the AOD is > 0.4 .

180 The white area on the daily mean MODIS map often correspond to the cloud distribution which was high over Quebec explaining the lack of large AOD daily mean values near this strong BB source. Nearby CALIOP tracks on 21 June over Colorado and 22 June over Canada show the vertical extent of the aerosol layers related to the fires (Fig.3). The aerosol layers reach 8 km over Colorado, while they remain below 4 km over Canada. The aerosol depolarization ratio is less than $7\pm 3\%$ for the
185 layers over Canada, while it is near $9\pm 3\%$ in the mid troposphere over Colorado. The uncertainty on the CALIOP aerosol depolarization ratio averaged over the two layers are calculated using the error on the 532 nm backscatter signals. Notice also the high depolarization ratio ($> 15\%$) over Colorado below 3 km showing that the BB plume overlays dust layers in the lower troposphere. The 6 areas shown in blue in Fig.2 are considered for a forward run of FLEXPART in order to study the long
190 range transport of the Canadian and Colorado biomass burning tracer. The depth of the volume is set according to the CALIOP vertical distributions shown in Fig.3. The parameters of the different BB sources considered in the FLEXPART simulations are given in Table 1.

The map of the biomass burning tracer plume over the ChArME_x domain on 27 and 28 June is shown in Fig.4 using the relative fraction between the emitted mass and the simulated mass in
195 the grid cell of the tracer field as explained before. Two different maps are given for the Canadian and Colorado fire contribution respectively. The Canadian plume has crossed the whole Western Mediterranean basin being over Menorca already on 27 June 06 UT and passing over Sicily on 28 June during the day. The Colorado fires do not play a major role in the aerosol layers observed on 27 June, but according to the transport model they could be observed on 28 June mainly over Spain and
200 also in a 200- km wide strip parallel to a line from Gibraltar to Messina. The vertical cross sections (Fig.5) show that the front edge of the Canadian fires are above 4 km on 27 June while the tails bring aerosol at lower altitudes in the 1-4 km altitude range on June 28. The Colorado fires can be only detected above 5 km. The relative mass fraction is larger than 30% in the Canadian fire plume showing that a significant part is indeed advected above the Mediterranean while the remaining part
205 is transported to Central Europe as observed by the EARLINET lidar network in Germany Gross et al. (2015).The relative fraction for the Colorado fires remains in the range 20-30% because the major part of the plume remains over Spain and the Atlantic ocean.

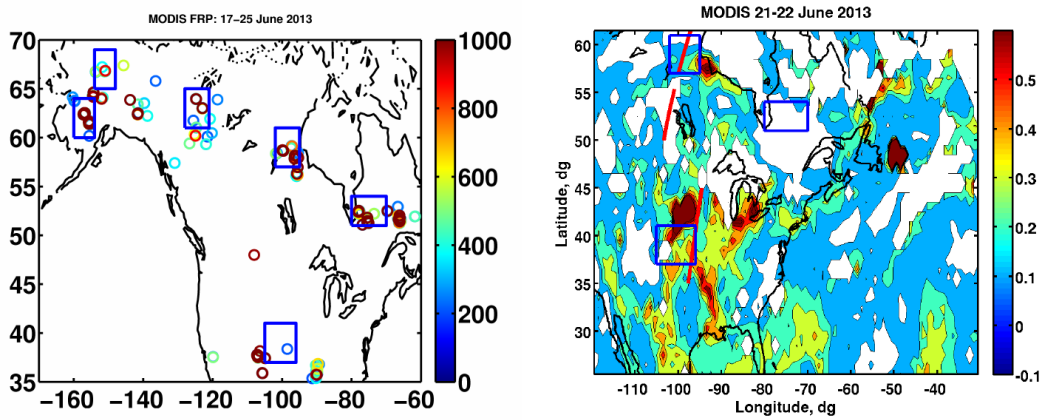


Figure 2. (Left panel) MODIS Fire Radiative Power from 17 to 25 June 2013. Areas with red dots are considered as significant fires. The blue boxes correspond to area chosen for the release of particles in the FLEXPART forward simulation. (Right panel) Daily AOD $0.5 \mu\text{m}$ measured by MODIS on 22 June 2013. The CALIOP tracks used to estimate the height of layer influenced by the fires are shown in red on 22 June near Hudson Bay fires and 21 June 2013 near Colorado fires

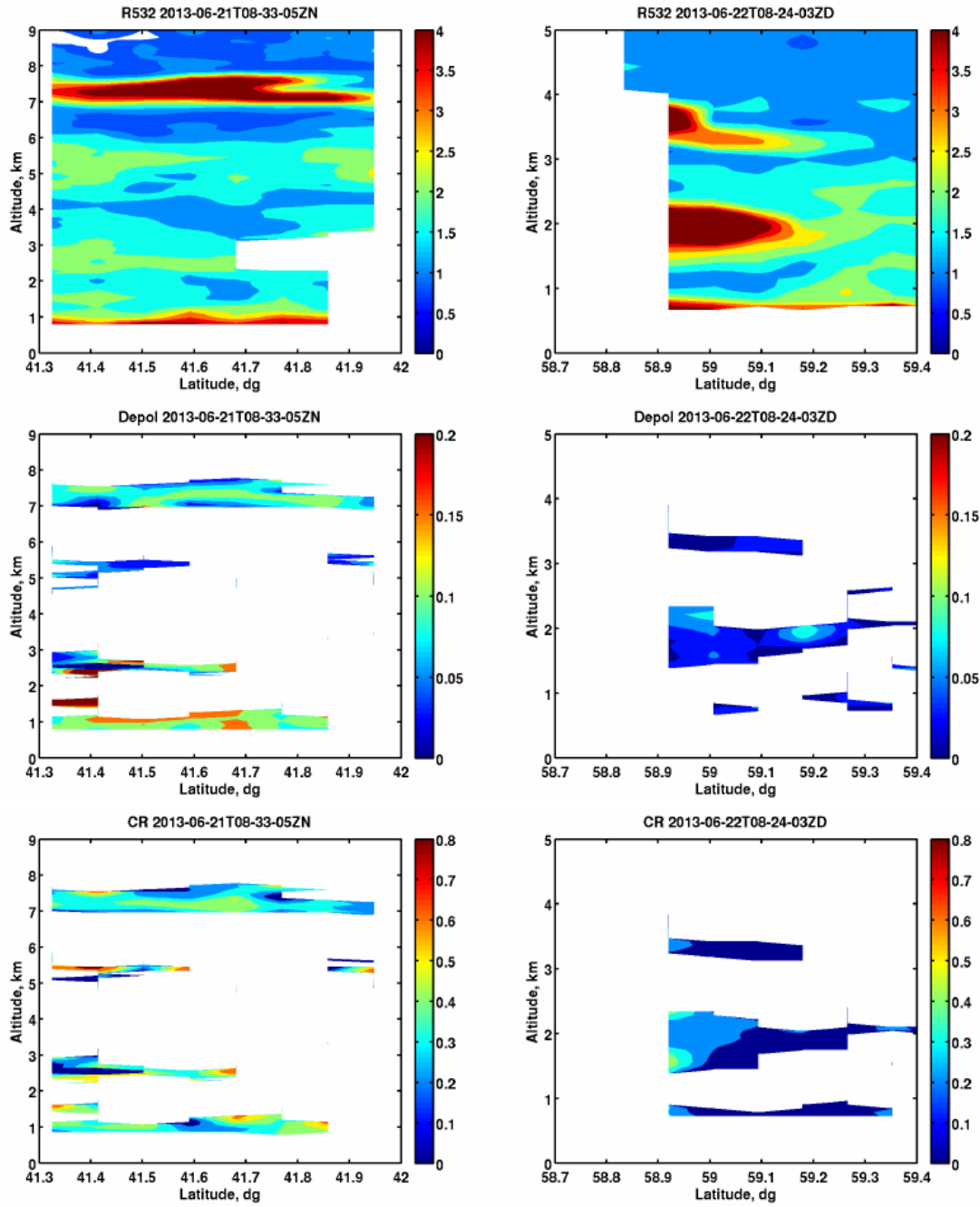


Figure 3. CALIOP vertical cross section of backscatter ratio (top), aerosol depolarization ratio (middle) and aerosol color ratio (bottom) for the two tracks shown in Fig.2 on 21 (left) and 22 (right) June 2013. Depolarization and color ratios are only reported for backscatter ratio >2 . Aerosol layers on 21 and 22 June 2013, are near the Colorado and Canadian fires, respectively.

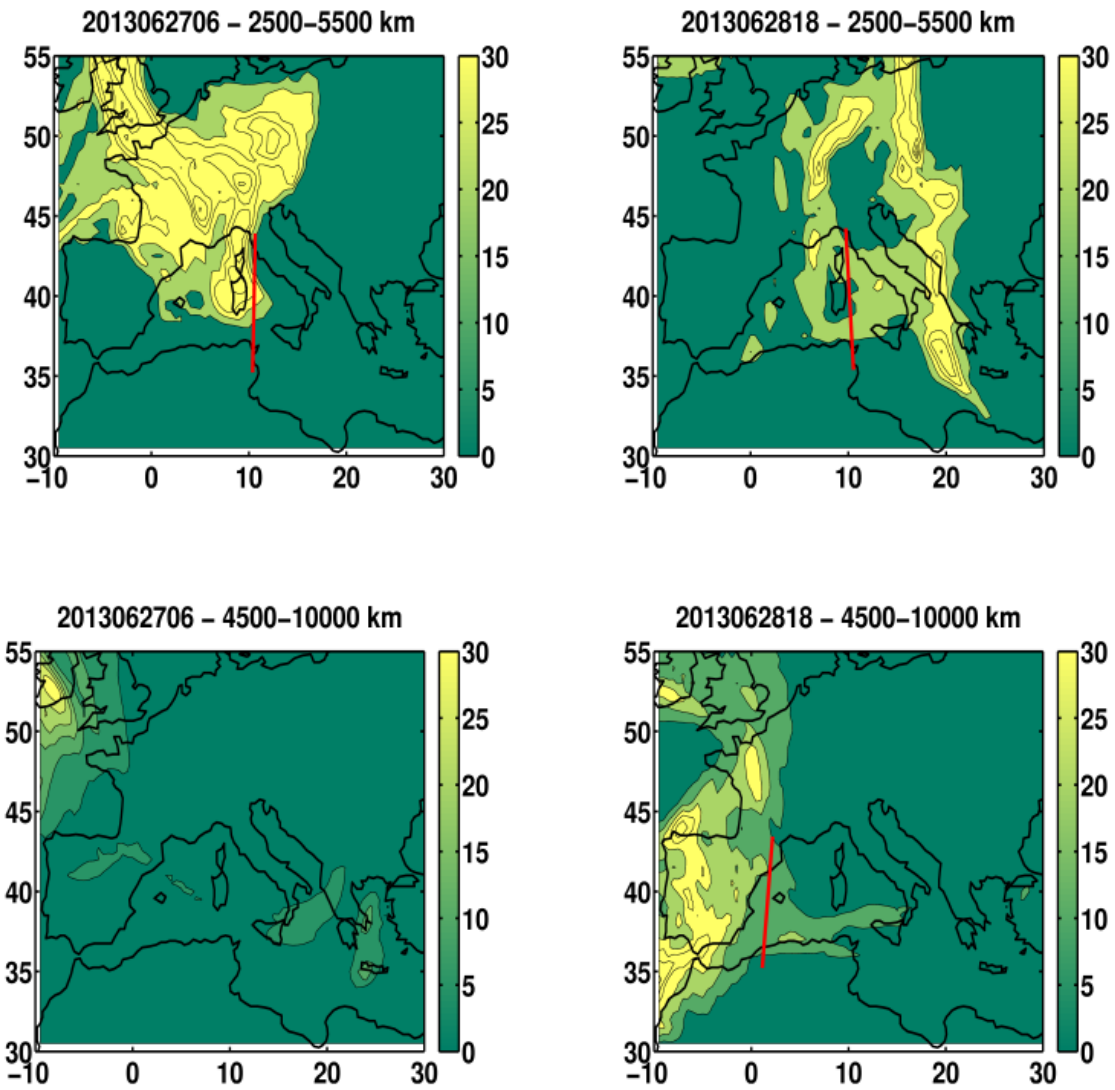


Figure 4. Map of the relative fraction of the FLEXPART biomass burning tracer plume in % for the Canadian (top) and Colorado (bottom) fires on 27 June 2013 06 UT (left) and 28 June 18 UT (right). The altitude range corresponds to the vertical levels included in the calculation of the tracer relative fraction.

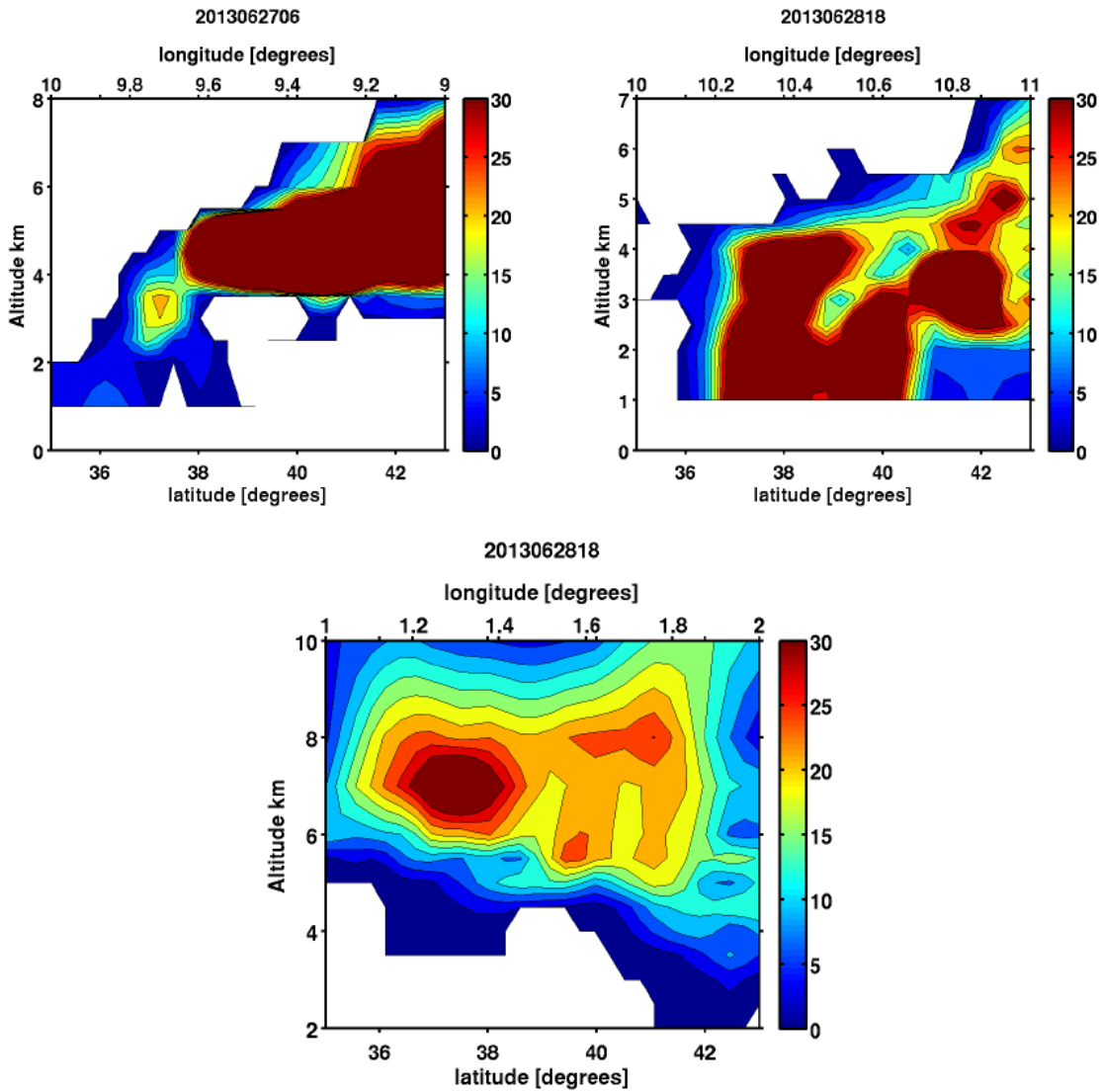


Figure 5. Vertical cross section of the relative fraction of the FLEXPART biomass burning tracer in % for the Canadian fires on 27 June 2013 06 UT (top), 28 June 18 UT (middle) and the Colorado fires on 28 June 18 UT (bottom).

3.3 North American dust layers

Modeling and satellite observations suggest that the Western USA is a significant contributor to the global mineral dust aerosol budget (Ginoux et al., 2001) and mineral dust emissions from this source region may have increased during the last 20 years (Brahney et al., 2013). Several dust blows hit Utah, Colorado and Wyoming in June 2013 due to the very dry conditions and strong winds which were also the cause of the Colorado forest fires (Hahnenberger and Nicoll, 2012). The NAAPS aerosol transport model simulations indicate elevated surface dust concentrations ($> 300 \mu\text{g}\cdot\text{m}^{-3}$) from 19 to 22 June 2013 in a region almost similar to the large MODIS AOD area related to the Colorado fires. It also explains the aerosol layers with large depolarization seen by CALIOP on 21 June 2013 at 41°N at 3 km below the Colorado fires (Fig.3). In addition to the local sources coming from Western USA, the MODIS maps on 20 and 21 June also show that dust streamers are transported at latitudes north of 30°N from the large scale Saharan dust plume, crossing the Atlantic because of the trade winds. Three streamers are shown in Fig.6 over the Atlantic ocean where the 0.5 μm AOD is enhanced with values > 0.3 . Nearby CALIOP tracks on 20 and 21 June show that the AOD enhancement are indeed related to the contribution of aerosol layers with large depolarization $> 20\%$ (Fig.7). The uncertainty on the average depolarization ratio for the dust layers is of the order of 5%. The 3 areas shown in blue in Fig.6 are considered in our study in order to analyze the role of dust layers over the Atlantic in the aerosol distribution over the Mediterranean Sea. According to the CALIOP vertical cross sections, the northern layer at 42°N was already uplifted in the altitude range 3-5 km while the dust plumes near 30°N remains below 3 km. Four areas are then selected for a FLEXPART forward run of dust tracers (see Table 2). The emission volume is set according to the MODIS AOD anomalies horizontal extent and the CALIOP vertical distribution of the dust layers. The emission period is chosen between 20 and 22 June for the dust layers over the Atlantic when the AOD anomalies are observed with MODIS, while the time frame for the High Plain region dust source is set according to the NAAPS model simulations.

The amount of tracer related to the High Plains dust sources was found to be negligible over the ChArMEx area during the period 27 and 28 June (mass fraction $< 10\%$) and it will not be considered any further. It may have been however mixed with the lower boundary of the Colorado fire plume seen at higher altitudes as shown in the previous section. The maps of the Atlantic dust tracer plume over the ChArMEx campaign domain is shown on 27 and 28 June in Fig.8. The values of the mass fraction is significant ($> 30\%$) showing that the contribution of long range transport of dust cannot be neglected even during the event of biomass burning aerosol transport to Europe. A first plume of dust was advected across the Western Mediterranean basin already before 27 June and a second crossed the basin on 28 June. The tail of the first one is at relatively low altitude (< 4 km) on 27 June while the second one is above 5 km on 28 June.

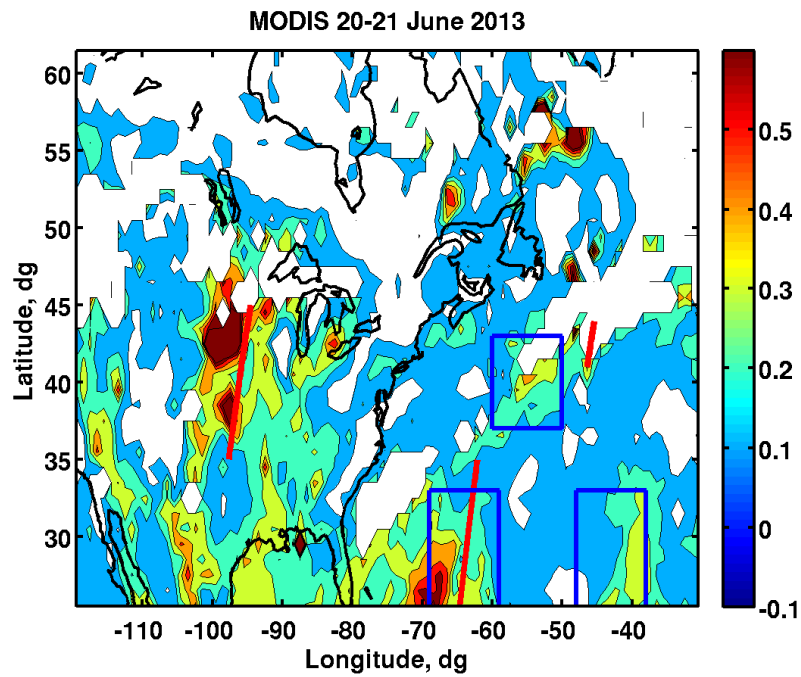


Figure 6. Daily AOD $0.5 \mu\text{m}$ measured by MODIS on 20 June 2013. The CALIOP tracks used to estimate the heights of the dust layers over the Atlantic ocean are shown in red on 20 June at 42°N and 21 June at 30°N . The blue boxes correspond to areas chosen for the release of particles in the FLEXPART forward simulation.

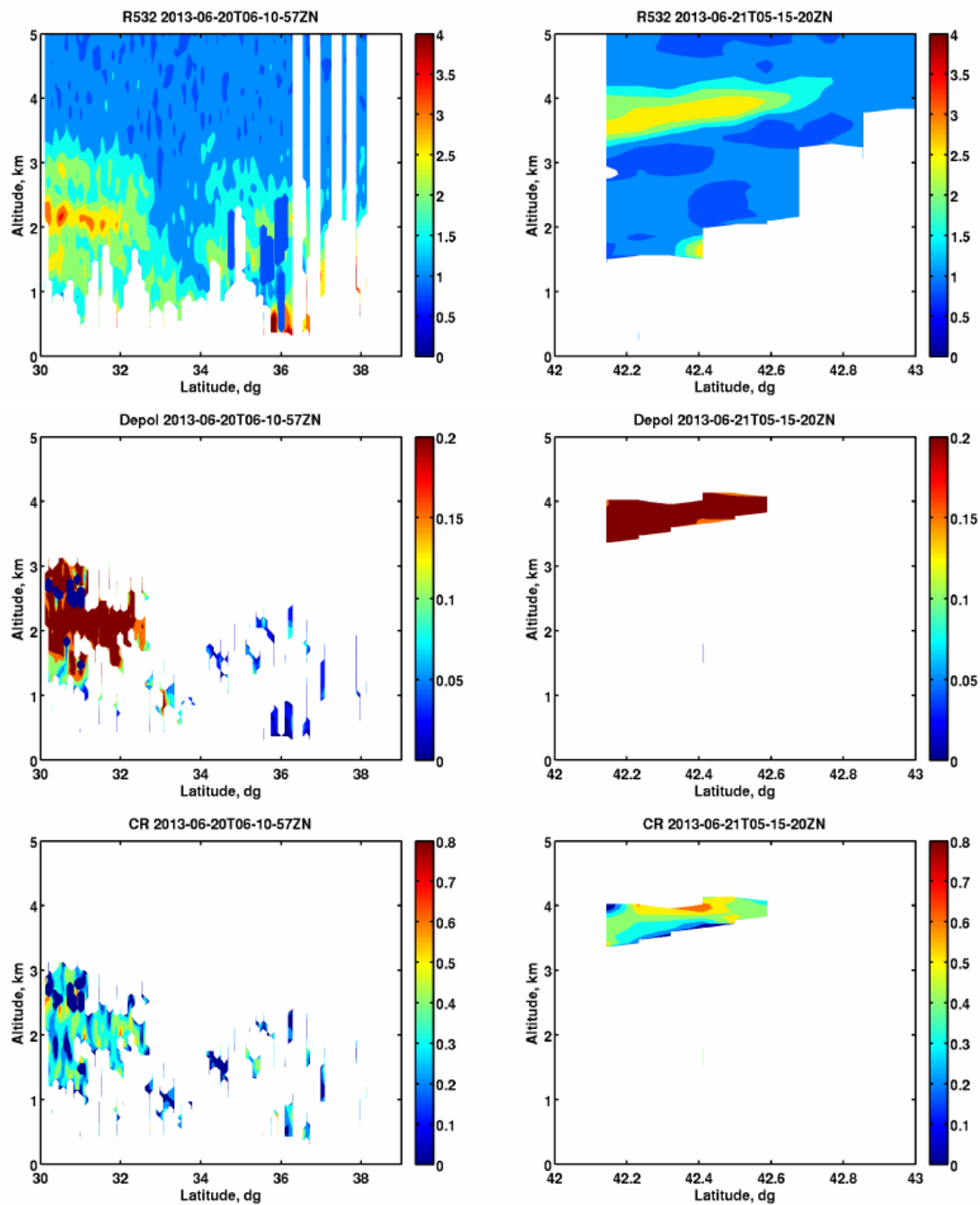


Figure 7. Same as Figure 3 for the two tracks shown in Fig. 6 on 20 (left) and 21 (right) June 2013. Dust layers are seen above the Atlantic ocean in the altitude range 1-4 km, near 32° and 42°N, on 20 and 21 June 2013, respectively.

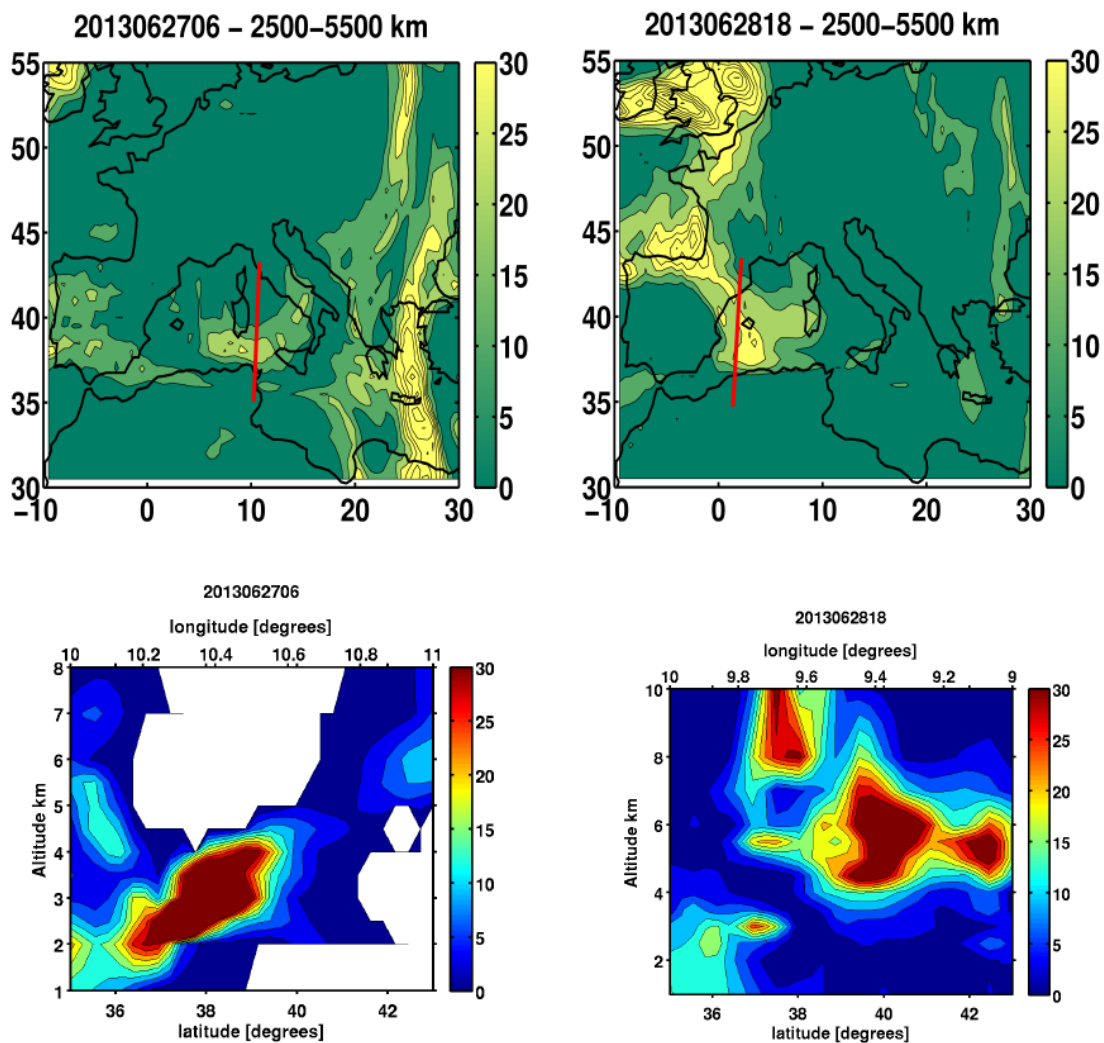


Figure 8. Map (top) and vertical cross section (bottom) of the relative fraction of the FLEXPART Atlantic dust tracer in % on 27 June 2013 06 UT (left) and 28 June 18 UT (right). The altitude ranges in the top figures correspond to the vertical levels included in the calculation of the tracer relative fraction.

Table 1. Characteristics of the biomass burning tracer emission used for the forward FLEXPART simulation. The emitted mass is only a rough estimate explaining the use of relative mass fraction in the simulation analysis. Altitude is given above ground level.

Aerosol Source	Release Time	Horizontal domain	Vertical range,km	Emitted mass, kg
Quebec BB	18-24 June 2013	-80W/-70W, 51N/54N	0-3	$3 \cdot 10^7$
Manitoba BB	20-24 June 2013	-102W/-95W, 57N/61N	0-3	$2.5 \cdot 10^7$
NWT BB	22-24 June 2013	-128W/-121W, 61N/65N	0-5	$2 \cdot 10^7$
N.Alaska BB	17-19 June 2013	-160W/-154W, 60N/64N	0-5	$1.9 \cdot 10^7$
S.Alaska BB	19-22 June 2013	-154W/-148W, 65N/69N	0-5	$3.6 \cdot 10^7$
Colorado BB	19-22 June 2013	-105W/-96W, 37N/41N	0-6	$5 \cdot 10^7$

Table 2. Same as Table 1 for the dust tracer emission

Aerosol Source	Release Time	Horizontal domain	Vertical range,km	Emitted mass, kg
Dust High Plains	19-22 June 2013	-105W/-99W, 37N/40N	0-3	$5 \cdot 10^7$
Dust over Atlantic	20-21 June 2013	-60W/-50W, 37N/43N	1-5	$5 \cdot 10^7$
Dust over Atlantic	20-21 June 2013	-69W/-59W, 25N/33N	1-4	$5 \cdot 10^7$
Dust over Atlantic	20-21 June 2013	-48W/-38W, 25N/33N	1-4	$5 \cdot 10^7$

3.4 Saharan dust

Although the synoptic wind conditions (northwesterly flow) from 25 to 29 June 2013 were not favorable for the export of Saharan dust to the basin as explained in Mallet et al. (2016), it is important to set the northern limit of the area influenced by the northward transport of Saharan dust. The characteristics of the dust emissions were estimated using the Multi-angle Imaging SpectroRadiometer (MISR) AOD maps for the period 22 to 28 June (not shown) because the multiangle observations are better suited to distinguish surface and dust contribution to the solar reflection. The depth of the Saharan dust layer has been estimated looking at several CALIOP overpasses above North Africa during the same period. A FLEXPART forward run with a Saharan dust tracer was made for a wide area over Northern Africa in the box (24°-34.5°N, 0°-10°E, 0-6 km) from 23 to 28 June 2013.

The vertical layering of the Saharan dust tracer over the ChArMEx domain is shown on 28 June in Fig.9. As expected for a nearby source, the relative mass fraction is very large ($> 100\%$). Although the dust outflow from Sahara is transported above Lampedusa, it remains south of 36.6°N between Lampedusa and Cagliari. No Saharan dust is expected above Menorca. The altitude of the dust plume is between 2.5 and 4.5 km because the uplifting in the westerly flow is very limited.

4 Aerosol observations in the Mediterranean basin

In this section, the ChArMEx aircraft or ground based lidar observations and the CALIOP vertical
260 cross sections on 27 and 28 June 2013 are compared with the expected contributions of the different
aerosol sources transported across the Atlantic.

4.1 Spatial distribution of the aerosol layers

Three nighttime CALIOP overpasses are suitable for a comparison with the different BB plumes:
27 June at -10°W and at 10°E , and 28 June at 0°W . The backscatter ratio $R(z)$ and the aerosol
265 depolarization ratio δ_{532} are shown in Fig.10 in the latitude range where cloud free sky made possible
the observations of aerosol layers. The $R(z)$ values are larger than 3 in these layers. On 27 June,
the layers are in the altitude range 5 to 7.5 km at 10°E while it is between 2 and 5 km at 10°W . In
both cases low δ_{532} values ($<10\%$) are found, showing that the plumes are not mixed with significant
amount of dust (except at 10°W where δ_{532} may reach 10% in some layers). The uncertainty for δ_{532}
270 is of the order of 3%. These results are in good agreement with the characteristics of the Canadian
fire plumes discussed in section 3. Indeed it was found that the front edge of the plume was at 10°E
on 27 June with an altitude range 4-7 km, while the tail is at -10°W in the altitude range 2-5 km
(see vertical cross section on 28 June in Fig.5). Although the Colorado fires may be present in the 27
June CALIOP cross section at -10°W according to the FLEXPART simulations, the altitude range
275 of the observed aerosol layers is not consistent with the influence of the Colorado BB plume which
is expected at altitude above 5 km. On 28 June at 0°W , the CALIOP observations show also aerosol
layers in the 3-5 km altitude range, with slightly higher depolarization ratio ($\approx 10\pm 3\%$), but still in

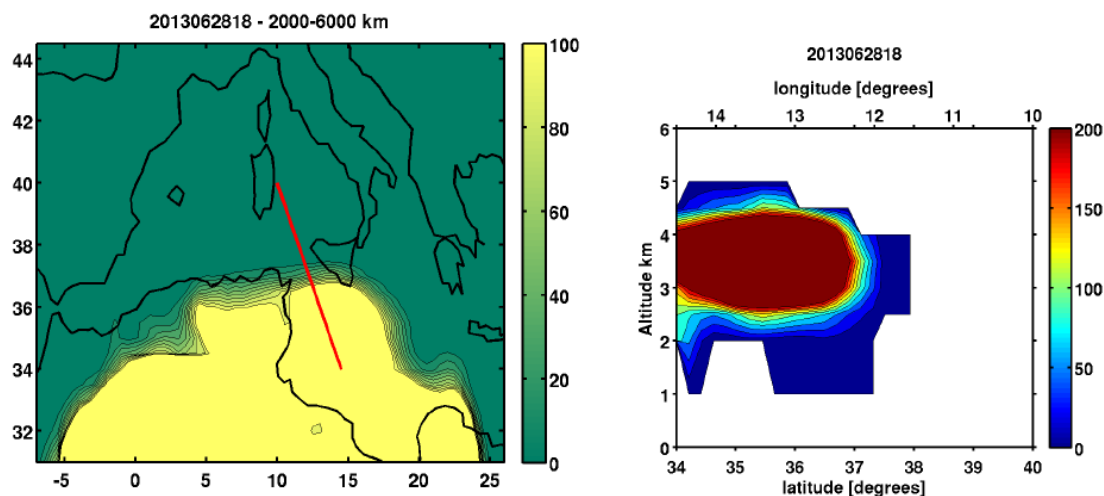


Figure 9. Same as figure 8 for the FLEXPART Saharan tracer on 28 June 2013 18 UT.

the range expected for biomass burning aerosol (Nisantzi et al., 2014). The altitude range is again in good agreement with a major role of the tail of the Canadian fire plume.

280 Several ground based lidar observations have also identified aerosol plumes possibly related to the transatlantic transport. The characteristics of the aerosol layers are summarized in Table 3. The Menorca lidar data are discussed in a companion paper by Chazette et al. (2015). An aerosol layer between 3-5 km seen in Menorca is quite similar to the CALIOP observations on 28 June. A second layer between 5-7 km is also seen in Menorca with a noticeable depolarization ($\delta_{355} > 12 \pm 1\%$).
285 The upper layer is not seen by CALIOP because it is expected at latitudes higher than 40°N and is masked by overlaying clouds. In Menorca the vertical profiles of the water vapor mixing ratio were also measured during the night (Chazette et al., 2014) showing elevated mixing ratio > 1 g/kg above 5 km and values near 0.5 g/kg in the aerosol layer observed around 4 km. The time series of the Menorca lidar is also useful to estimate the horizontal range of BB plume. The plume is observed
290 for 24 hours from 27 June 00 UT to 28 June 12 UT and the wind speed at 4 km is between 30-40 km/h, therefore the plume zonal extent is of the order of 1200 km. It is very similar to the size of the Canadian tracer plume obtained in the FLEXPART simulations (15° longitude difference between the front edge and the tail).

As expected the Barcelona lidar detects similar features: a strong layer between 5-7 km with
295 $\delta_{532} \approx 10\%$ and an optically thin layer between 3-5 km with $\delta_{532} < 10\%$. The spectral variation of the aerosol depolarization ratio between Barcelona and Menorca cannot be accurately estimated but is less than 1.5. It is consistent with a small influence of urban aerosol (Burton et al., 2012). When looking at the Lampedusa lidar data at 35°N , a layer is seen between 2 and 4 km on 28 June which is influenced by the Saharan dust outflow discussed in section 3 since $\delta_{532} \geq 30\%$, i.e. a value similar
300 to other dust layers observed over Menorca during ChArMEx (Chazette et al., 2015).

The Lampedusa lidar measures aerosol layers in the 2-4 km altitude range on both days, but with very different optical characteristics. A dust layer with $\delta_{532} > 30\%$ on 28 June 12 UT while a mixture of dust and BB aerosol is seen on 27 June from 8 UT to 16 UT. The aerosol layer seen by CALIOP on 27 June 01 UT near 36°N has optical characteristics close to the layer observed in Lampedusa on
305 27 June (Fig.10), i.e. a depolarization between 10%-15% and LR between 50-55 sr.

The LNG airborne lidar data obtained during ChArMEx will be thoroughly discussed in a forthcoming paper by J.Pelon and co-workers. Here we will only consider the vertical structure of the aerosol layers observed on 28 June 2013 along the loop shown in Fig.1. The 3 corresponding vertical cross sections of attenuated $R(z)$ at 532 nm are shown in Fig.11. Three interesting regions can
310 be identified:

- (A) the 38.2°N layer at 2-4 km on the Cagliari-Lampedusa section and at $11-14^\circ\text{E}$ on the return section between Messina and Cagliari,
- (B) the upper altitude layer in the 4-6 km altitude range covering a southwest (36°N , 12°E)-northeast (39°N , 15°E) band, the width of which is of the order of 100 km,

Table 3. Characteristics of the aerosol layers observed in the free troposphere by the ground based lidars listed in Fig.1 on 27 and 28 June 2013.

Lidar	Wavelength nm	Layer altitude	Time period	Scattering ratio	Depolarization ratio	Lidar ratio
Menorca	355	5-7 km	27/6 00UT to 28/6 00UT	1.87 ± 0.03	$10\pm 1\%$	42 ± 5 sr
Menorca	355	3-5 km	27/6 00UT to 28/6 12UT	1.22 ± 0.02	$4\pm 1\%$	59 ± 5 sr
Barcelona	532	5-7 km	26/6 20UT to 27/6 16UT cloudy after	4.3	$\approx 10\pm 2\%$	
Barcelona	532	3-5 km	26/6 12UT to 27/6 16UT cloudy after	1.4	$7\pm 2\%$	
Lampedusa	532	2-4 km	27/6 08UT to 27/6 16UT	1.5 ± 0.03	$12\pm 2\%$	51 ± 9 sr
Lampedusa	532	2-4 km	28/6 08UT to 28/6 14UT	2.8 ± 0.04	$30\pm 2\%$	30 ± 6 sr

315 – (C) a low altitude layer between 2-4 km south of 36 N which corresponds to the layer seen by the Lampedusa lidar.

The spatial distribution of the aerosol layers seen by the LNG lidar corresponds quite well with the position of North American BB plumes and the expected latitudinal extent of the Saharan dust
 320 Canadian BB plume. The layer B is also in the latitude range of the Canadian BB plume, possibly mixed with the Colorado BB plume present between Gibraltar and Messina (see Fig.4). Layer A and B seen by the LNG airborne lidar on 28 June are also consistent with the superposition of two different aerosol layers seen above Menorca 24 hours before. In layer B, $\delta_{355} \approx 10\pm 1\%$, i.e. higher than the low values found in layer A ($\delta_{355} \leq 5\pm 2\%$).

325 4.2 Aerosol source attribution

Although the comparison with the position of the FLEXPART tracer plumes can already help to attribute a specific source to the observed layers in the ChArMEx area, it can be further checked by calculating the potential emission sensitivity (PES) values by running the FLEXPART model in the backward mode for 10-11 days to identify the area where surface emissions may influence the
 330 observed aerosol structure seen by CALIOP, the Menorca ground based lidar and the LNG airborne instrument. The PES is given in s unit in order to be multiplied by model surface fluxes to produce

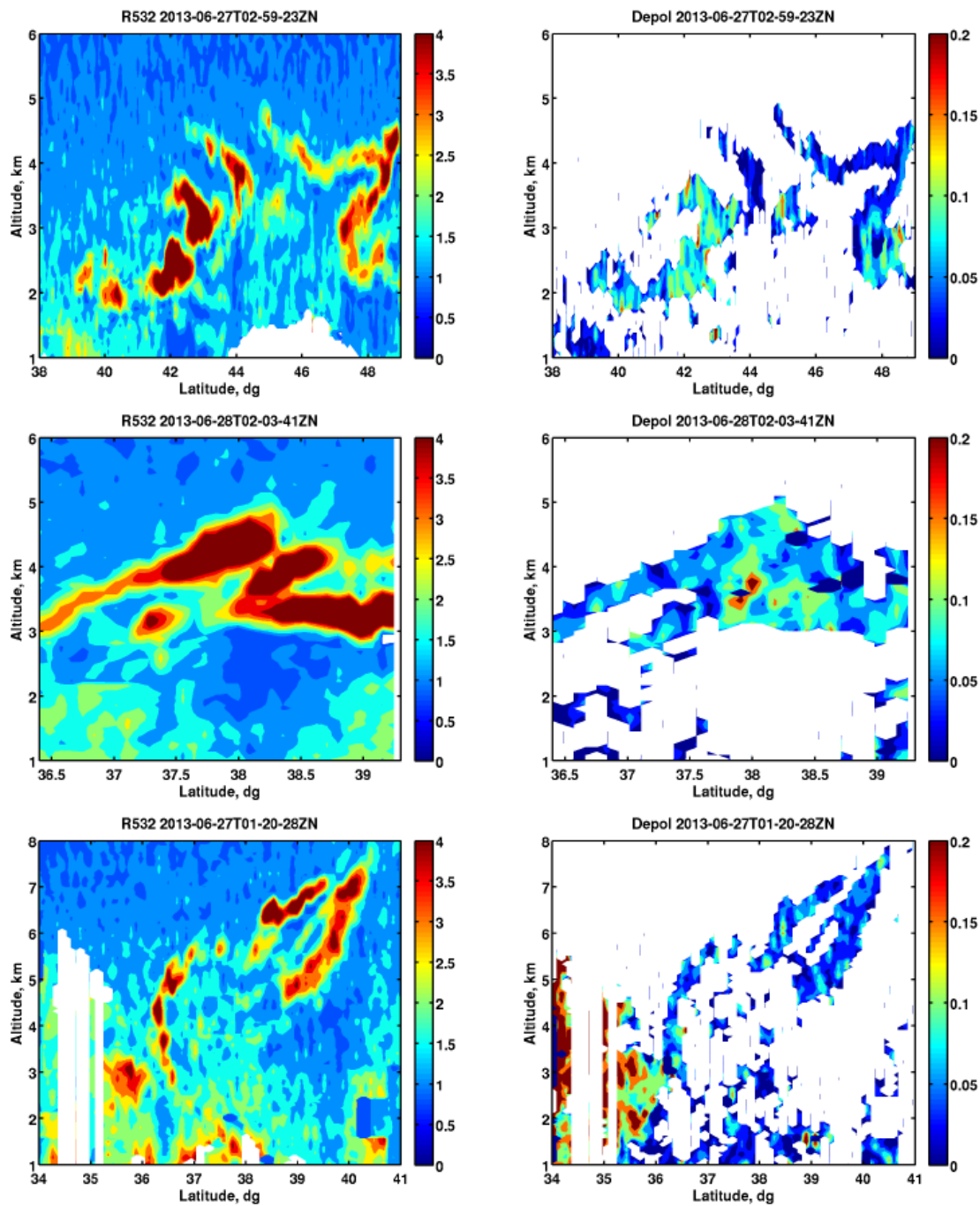


Figure 10. CALIOP vertical cross section of backscatter ratio (left panel), aerosol depolarization ratio (right panel) for the 3 tracks shown in Fig.1 on 27 June 2013 03 UT at -10° W (top panel), on 28 June 2013 2 UT at 0° W (middle panel), on 27 June 2013 01 UT at 10° E (bottom panel).

concentrations at the receptor location. The PES is calculated using 6 hours averages on a three dimensional $1^\circ \times 1^\circ \times 1$ km grid. The results are shown for the CALIOP and Menorca observations on 27 June 2013 (Fig.12). Similar calculations were also made for layer A, B and C seen by the airborne lidar on 28 June (Fig.13). The simulations for the layers seen by CALIOP on 28 June 02 UT are not shown because they are very similar to the results obtain for the Menorca lidar on 27 June at 12 UT or for layer B seen by LNG.

The aerosol layers observed by CALIOP along the two cross sections at -10°W and 10°E are indeed mainly related to aerosol sources over Canada and Alaska, but the retro-plume altitude and latitude at 60°W are quite different when reaching the Atlantic Ocean. The probability of dust and biomass mixing is higher for the CALIOP layers at -10°W which is located in the 40° - 50°N latitude band at lower altitude (5-7 km) than for the CALIOP layers at 10°E . This may explain the slight depolarization difference for the two CALIOP tracks since there are more layers with $\delta_{532} \approx 10\%$ at -10°W than at 10°E . The mixing of dust layers over the Atlantic and Canadian BB aerosol is even more explicit for the Menorca layer at 6 km since two branches of elevated PES are seen over the two aerosol source regions identified for this layer. It explains the relatively higher aerosol depolarization ratio (up to 12%) at 6 km than at 4 km in Menorca during this episode. Such a transport pathway is also consistent with the water vapor mixing ratio maximum > 1 g/kg seen by the Menorca water vapor lidar near 6 km since uplifting of air masses from the lower troposphere above the Atlantic Ocean is likely to increase the humidity in the mid-troposphere.

When considering the PES related to the airborne lidar layers, the layer A PES is similar to the 10°E plume showing a strong influence of the Canadian aerosol BB source, while the layer B PES distribution resembles the results obtained with the Menorca layer seen one day earlier. For the dusty layer C seen both by the aircraft and at the Lampedusa station, the PES distribution shows that there is no transatlantic transport for the period 17 to 28 June while the aerosol sources are mainly located above North Africa and Western Europe at low altitude (< 3 km). Although air masses are still advected from Western Europe, Saharan dust emission remains the major aerosol source since Western Europe air masses were heavily influenced by Saharan dust layers during the period 16 to 20 June (Mallet et al., 2016). It is consistent with the large depolarization seen both above Lampedusa ($\delta_{532} \approx 30\%$).

The ATR42 aircraft also flew between Cagliari and Lampedusa on 28 June around 12 UT to sample the aerosol layers with in-situ measurements (Pelon et al., 2015). The analysis of the CO and BC in-situ measurements made on-board the ATR42 shows that the layer A and B correspond to a CO excess above background of the order of 100 ppbv while ΔCO is less than 20 ppbv for layer C (not shown). The BC variability shows also the same pattern. This is in very good agreement with the conclusions derived from the lidar data analysis coupled with the Lagrangian transport model simulations.

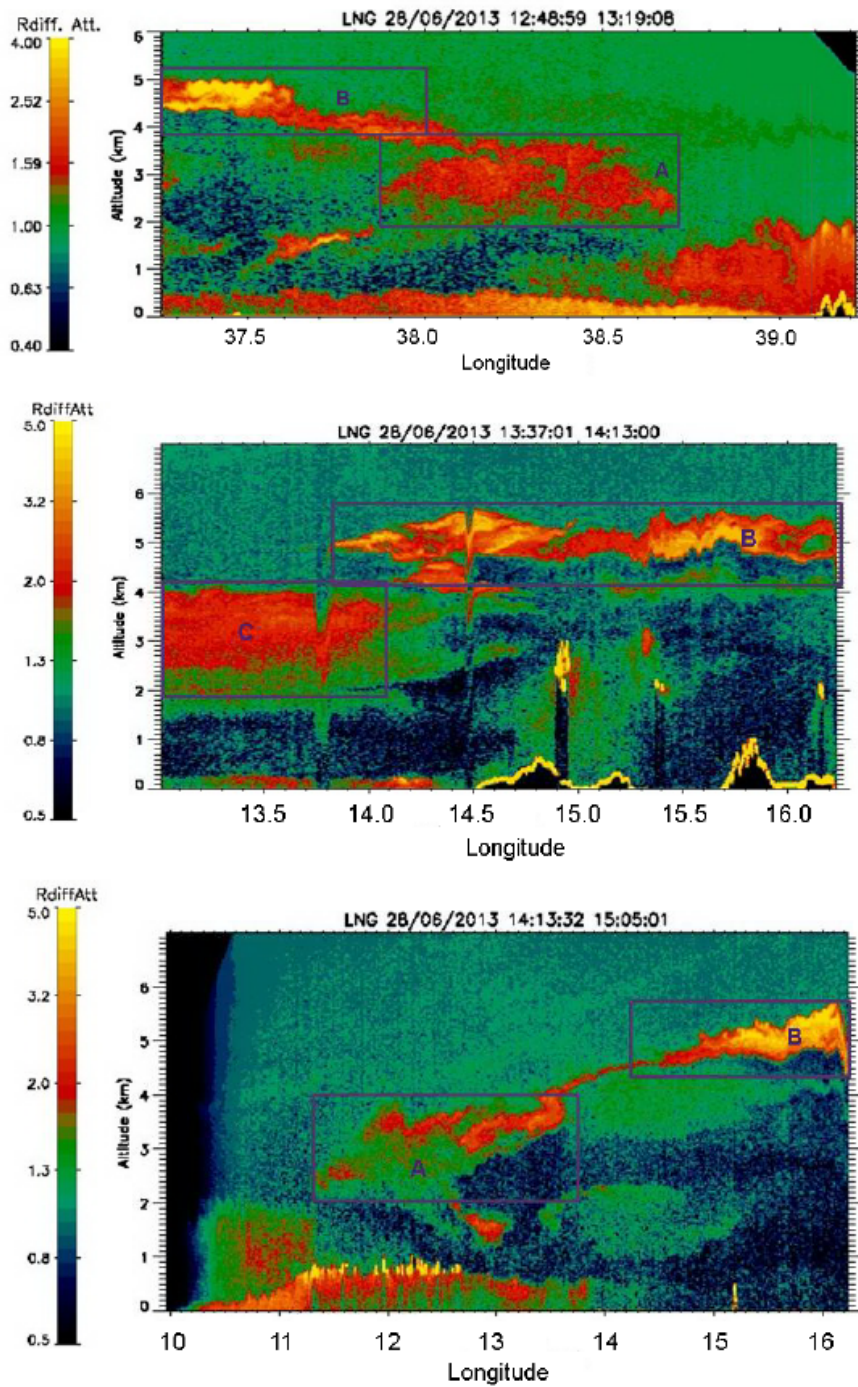


Figure 11. Airborne lidar vertical cross sections of attenuated backscatter ratio at 532 nm on 28 June 2013 along the loop shown in Fig.1: (top) Lampedusa/Cagliari around 13 UT (middle) Lampedusa/Messina around 14 UT (bottom) Cagliari/Messina around 14:40 UT.

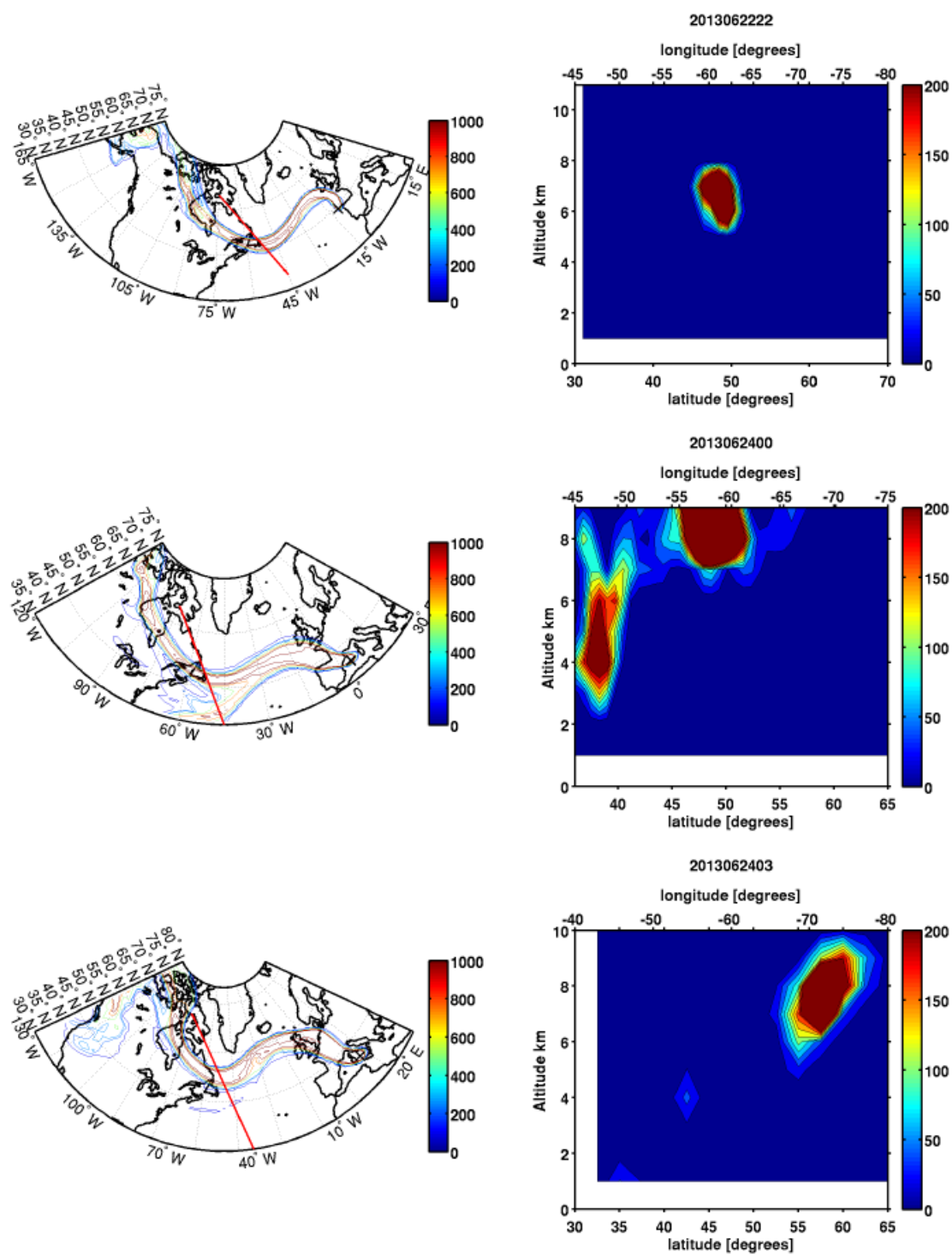


Figure 12. (Left panel) FLEXPART Potential emission sensitivity (PES) in s for 3 aerosol layers identified by CALIOP and Menorca lidar: 27 June 2013 02 UT, -10°W , 43°N (top) 27 June 12 UT, in Menorca (middle) 27 June 01 UT, 10°E , 39°N (bottom). The PES Vertical cross section are along the red line following the North American East Coast (right panel).

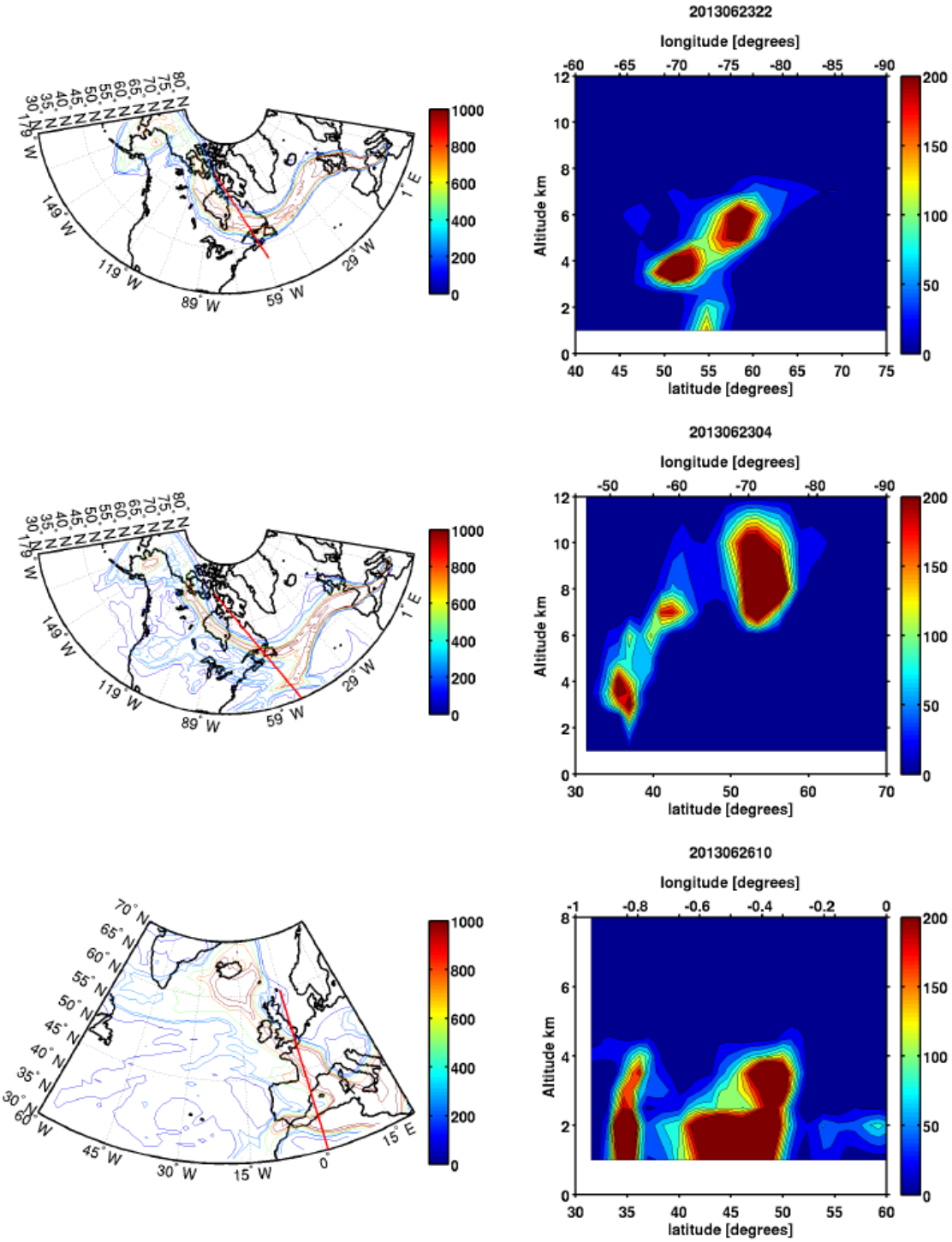


Figure 13. Same as figure 12 for the 3 aerosol layers identified by the Falcon 20 lidar on 28 June 2013: layer A (top) layer B (middle) layer C (bottom).

4.3 Aerosol optical properties

In this section, we will summarize the results about the aerosol layer optical properties and the aerosol source attribution. The analysis conducted in the previous sections leads to the identification of 4 different aerosol layers during the passage of the BB plume over the ChArMEx area:

- (I) pure BB layer at 10°E above 4 km on 27 June 2013 01 UT (CALIOP),
- (II) weakly dusty BB layer below 5 km observed between -10°W and 10°E on 27 and 28 June 02 UT by CALIOP, the Menorca lidar and layer A seen by LNG flying around Sicily on 28 June.
- (III) significant mixture of BB and dust transported across the Atlantic above 5 km at Menorca on 27 June 12 UT and layer B seen by LNG on 28 June 14 UT.
- (IV) the outflow of Saharan dust above the sea at latitudes South of 36°N on 28 June 13 UT (Lampedusa lidar and layer C seen by LNG).

For layer (I) δ_{532} is $< 5\%$, while the LR at 532 nm is 60 ± 20 sr when using the aerosol layer transmission from the averaged L1 CALIOP attenuated backscatter, and it is 65 sr in the level-2 (L2) CALIOP operational aerosol data products. The color ratio is between 0.2 and 0.4. Both LR and C_a are in the range expected for a pure BB layer (Burton et al., 2012) in agreement with our source identification.

For layer (II) δ_{532} and δ_{355} are respectively in the range 5-10% and $< 8\%$ with the lowest values on 27 June at 45°N along the -10°W CALIOP overpass. The LR calculated from the ground based and airborne lidars are more accurate and they are 59 ± 5 sr at 355 nm and 60 ± 5 sr at 532 nm, respectively. The CALIOP 27 June (-10°W) and 28 June LR at 532 nm estimated to be 60 ± 20 sr and 50 ± 20 sr, respectively, using the L1 data analysis and are of the order of 60 sr for both layers using the L2 operational products. It also gives confidence in the LR retrieval to see the largest LR is obtained where δ_{532} is minimum. The CALIOP C_a is in the range 0.4-0.5. These aerosol optical parameters are still in the range expected for a BB layer. However differences with the optical parameters found for layer (I) (higher depolarization and C_a , slightly lower LR) are consistent with a BB mixed with a small amount of dust or an increase in relative humidity. According to the small values (< 0.5 g/kg) of water vapor recorded by the Menorca lidar observations (Chazette et al., 2015), the mixing with a small amount of dust is more likely.

For layer (III) mainly seen by the Menorca ground based and airborne lidar, δ_{355} values are in the range 8-12%, while LR are 45 ± 5 sr and 42 ± 5 sr at 532 and 355 nm, respectively. The LR of the layer B seen by LNG is calculated by including also the contribution of the underlying layer between 3-4 km to get a better molecular reference. Following the methodology proposed by Tesche et al. (2009), the aerosol depolarization value for layer (III) is consistent with a contribution of 20%-30% of dust and 80%-70% of BB aerosol in the total aerosol backscatter in layer (III), if we assume that

Table 4. Optical properties of the four types of aerosol encountered during the passage of the BB plume: depolarization ratios (δ_{532} , δ_{355}), lidar ratios (LR_{532} , LR_{355}), color ratios (CR_a 1064/532, CR_a 532/355)

Aerosol type	δ_{532}	δ_{355}	LR_{532} , sr	LR_{355} , sr	CR_a 1064/532	CR_a 532/355
Pure BB (I)	<5%	-	60±20	-	0.2-0.4	-
Dusty BB (II)	5-10%	5-8%	60±5*, 51±9**	59±5	0.4-0.5	0.35
BB/dust mixture (III)	≈10±2%	8-12%	45±5	42±5	-	0.74
Saharan dust (IV)	30±2%	-	48±5*, 30±5**	-	-	-

* airborne lidar ** Lampedusa lidar

pure dust and pure BB aerosol types have δ_{355} of 25% and 5%, respectively and LR at 355 nm of 45±10 sr and 60±10 sr, respectively. It is also interesting to calculate C_a between 355 and 532 nm using the Barcelona and Menorca observations assuming that $R(z)$ is stationary during the advection of the aerosol layers between Barcelona and Menorca. The C_a value is 0.74 for layer (III) while it is only 0.35 for the layer (II). It is consistent with a larger contribution of the accumulation mode when BB is mixed with dust, but also with a larger water vapor mixing ratio (1 g/kg) for layer (III) than layer (II).

For layer (IV) larger depolarization up to 30% are seen by the Lampedusa lidar at 532 nm. The lidar ratio calculated by the LNG lidar and the ground based lidar at 532 nm are respectively 48±5 sr and 30±10 sr. The layer optical parameters are consistent with a dust plume with a large depolarization, while a large variability is observed for LR. The large depolarization ratio and the low LR value at 355 nm is quite similar to previous observation by Di Iorio et al. (2009) in fresh dust exported over the Mediterranean sea. The strong variation in the LR values between the layer C of the LNG lidar at 13.5 °E and the Lampedusa observations at 12.5°E suggests an increase of the mixing between the northward African dust outflow and the BB plume as the aircraft moved across the boundary between layer (IV) and (II) between Lampedusa and the southern cape of Sicily.

The aerosol properties and spatial distribution of the four aerosol types are summarized in, Table 4 and Fig.14 respectively. The spatial distribution of the MODIS AOD at 0.5 μ m is also shown in Fig.14, where the largest AOD values are seen before the plumes dispersion above Northern Spain. For the type (II) aerosol, i.e. an aged BB plume seen below 5 km and mixed with a small amount of dust mainly from continental origin, two areas are distinguished for the Colorado and Canadian fires using the results of the FLEXPART forward simulations. The Canadian fires significantly contributes to the AOD observed by MODIS over the Mediterranean sea. The additional contribution of the upper aerosol layers of type (III) where the BB is mixed with dust also explains the significant AOD increase over the Western Mediterranean region. The BB contribution to AOD is as large as the North African dust contribution (type IV) that dominates the southern part of the domain with AOD values in the same range of 0.3-0.4 over Northeastern Algeria and Tunisia.

430 5 Conclusions

A very interesting event of long range transport of biomass burning (BB) aerosols between North America and the Western Mediterranean region that took place in late June 2013 has been documented during the ChArMEX/ADRIMED campaign. Although the occurrence of such events has been discussed in previous publications, the contribution of this work is to take advantage of a large number of ground based and airborne lidar measurements used in conjunction with spaceborne lidar observations by CALIOP during this period. A detailed analysis of the biomass burning North American sources was conducted including the assessment of their transport to Europe using forward simulations with the FLEXPART model initialized using satellite observations. The specific question of mixing between dust and BB particles was addressed by considering the possible dust sources transported along the same transport pathway. The role of mixing was quantified by considering the optical properties of the different aerosol layers observed during two days of the ChArMEX campaign (27 and 28 June 2013) when the biomass burning aerosol load was at its maximum over the Western Mediterranean. The three dimensional structure of the aerosol distribution revealed by the lidar network and the airborne lidar flight provides a detailed assessment of the different aerosol source contributions when it is coupled with the results of the Lagrangian FLEXPART transport model. Four aerosol types were identified using the depolarization ratio and the three dimensional structure of the aerosol plume: (I) pure BB layer, (II) weakly dusty BB, (III) a significant mixture of BB and dust transported from the North Atlantic trade wind region (IV) the direct northward outflow by the subtropical jet of Saharan dust not mixed with BB aerosol. Mixing of dust and BB can correspond to a 20%-30% dust contribution in the total aerosol backscatter. The comparison with the MODIS AOD distribution during this episode over the Western Mediterranean sea shows that the Canadian fires contribution were surprisingly as large as the direct northward dust outflow from Sahara. An additional contribution from a mid-tropospheric aerosol layer due a mixture of dust and BB aerosol was found in the region of higher AOD seen by MODIS. The next step will now concern the use of all presented and analyzed data for evaluating 3-D regional models to simulate this specific event, in terms of optical properties, possible mixing and vertical extent of mineral dust and forest fire aerosol layers.

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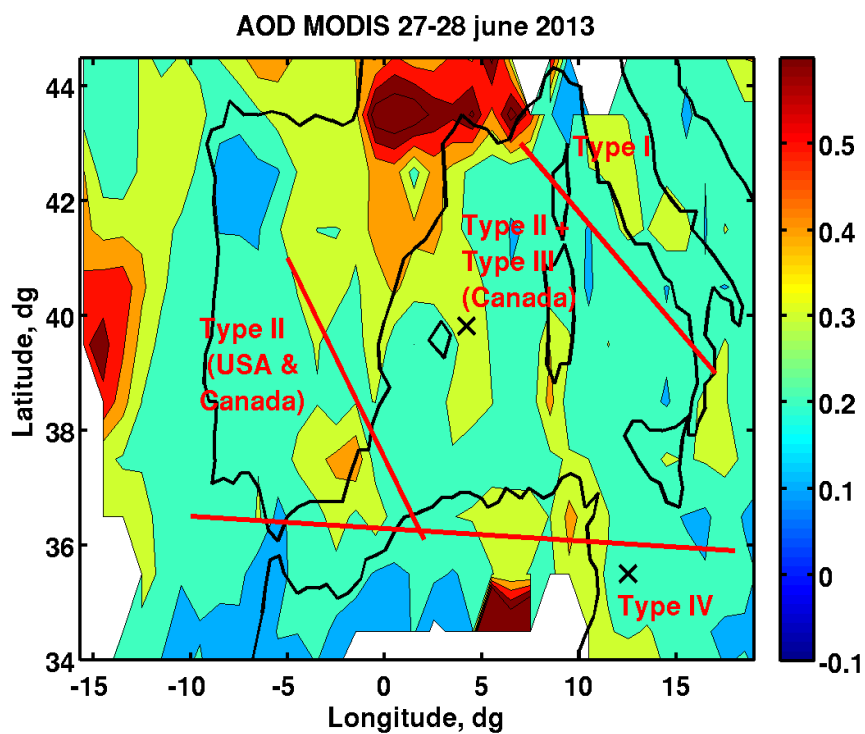


Figure 14. MODIS AOD horizontal distribution on 27 and 28 June 2013 over the Mediterranean region. The area corresponding to the aerosol types identified during our analysis of the BB plume passage are delimited by the red lines. The black crosses are for the Menorca and Lampedusa stations.

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