Raman lidar observations of aerosol emitted during the 2002 Etna eruption

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[1] Results of a lidar measurement campaign performed at IMAA-CNR (Potenza, Italy) during the 2002 Etna volcano eruption, are reported. With combined Raman elastic-backscatter lidar, independent measurements of aerosol extinction and backscatter coefficients at 355 nm are retrieved, whereas aerosol backscatter coefficient at 532 nm is retrieved from elastic lidar signal. A volcanic aerosol layer has been observed in the free troposphere on 1–2 November. AVHRR images show direct plume transport from Etna to Potenza. Optical depth of 0.1 and lidar ratio of 55 sr at 355 nm have been observed at the moment of direct transport. For the first time, a measurement of the lidar ratio for tropospheric volcanic aerosol has been performed. This lidar ratio value and the estimation of the wavelength dependence of the aerosol backscatter coefficient of 2.4 indicate the presence of young sub-micron sulfate particles, a low soot content, and the absence of large ash particles. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0370 Atmospheric Composition and Structure: Volcanic effects (8409); 3360 Meteorology and Atmospheric Dynamics: Remote sensing. **Citation:** Pappalardo, G., A. Amodeo, L. Mona, M. Pandolfi, N. Pergola, and V. Cuomo (2004), Raman lidar observations of aerosol emitted during the 2002 Etna eruption, *Geophys. Res. Lett.*, *31*, L05120, doi:10.1029/2003GL019073.

1. Introduction

[2] The impact on the climate caused by volcanic emissions and in particular by volcanic aerosol has been recently investigated in a review paper [*Robock*, 2000]. Previous studies of volcanic aerosol have been focused on characterizing of stratospheric volcanic aerosol in terms of microphysical properties and on evaluating its contribution to radiation budget [*Hansen et al.*, 1992; *Jäger*, 1992; *Graf et al.*, 1993]. In contrast, the radiative effects of tropospheric volcanic aerosol are insufficiently modeled and poorly quantified. During quiescent volcanic emissions (including degassing, fumaroles and small eruptive events), sulfate and soot are added to the troposphere. Sulfate particles reflect solar radiation, act as cloud condensation nuclei, and modify the radiative properties and lifetime of clouds, and therefore influence the precipitation cycle. Even if volcanoes globally emit only 14% of total tropospheric sulfur [*Graf et al.*, 1997], this percentage is important from a climatological point of view, because volcanic aerosol is injected in the free troposphere, where the removal processes are relatively slow and the resulting residence time of tropospheric volcanic aerosol is about 1–3 weeks.

[3] A realistic estimation of the climate effects of tropospheric volcanic emission needs more comprehensive observational data on chemical, physical and radiative properties of this kind of aerosol. In particular, data on optical properties of tropospheric volcanic aerosol are needed as input data for improvement of climate prediction models and of satellite retrieval algorithms. In this paper, we show and discuss lidar observations of tropospheric aerosol emitted during the volcanic eruption of Mt. Etna in 2002. The aerosol layer has been characterized in terms of aerosol backscatter coefficient at 355 and 532 nm, and aerosol extinction coefficient and lidar ratio at 355 nm. Lidar ratio, i.e., the ratio between aerosol extinction and backscatter coefficient, is an important optical parameter in aerosol characterization, because it depends on intensive aerosol properties such as chemical composition, size distribution of the particles, and particle shape [*Evans*, 1988; *Mishchenko et al.*, 1997; *Ackermann*, 1998]. What we report is, as far as we know, the first measurement of lidar ratio profile for tropospheric volcanic aerosol. Measured lidar ratios are needed as input parameters for the retrieval of optical properties from standard elastic-backscatter lidar, widely used worldwide. In addition, these observations could be very important to improve satellite retrieval algorithms also for next satellite missions as those involving backscatter lidar onboard as CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) [*Winker et al.*, 2002]. Moreover, lidar ratio measurements together with multiwavelength backscatter observations provide information on the size/composition characteristics of particles [*Franke et al.*, 2001; *Ansmann et al.*, 2002; *Müller et al.*, 2003].

2. Results and Discussion

[4] In 2002, from the end of October until the end of December, volcanic activity of Mt. Etna, Sicily, Italy (37°44′N, 15°E, 3350 m above sea level), the largest European volcano, was characterized by numerous strong eruptive and seismic events. Approximately 100 kt of SO_2 were injected into the free troposphere in the period from 27 October to 2 November 2002 (data are available from the World Wide Web server for the Istituto Nazionale di Geofisica e Vulcanologia at http://www.ct.ingv.it/Etna2002/main.htm) with a maximum value of about 20 kt/d on 29 October, against a value of about 1 kt/d observed in the pre-eruptive phase [*Salerno et al.*, 2003].

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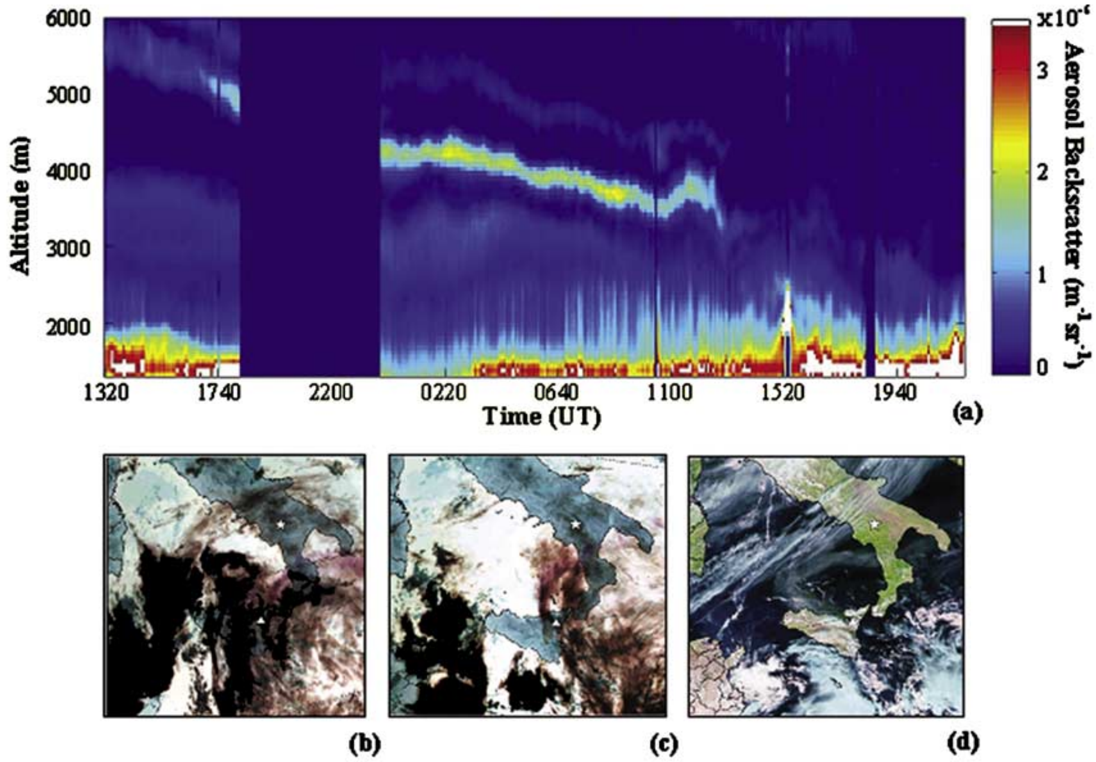

Figure 1. Temporal evolution of aerosol backscatter coefficient profiles at 532 nm (a). Profiles have been integrated over 5 minutes with a vertical resolution of 60 m (from 1 November, 1320 UT to 2 November, 2200 UT). False colors AVHRR images captured at three different times: (b) 1 November at 1658 UT, NOAA-15; (c) 2 November at 0050 UT, NOAA-16; (d) 2 November at 1216 UT, NOAA-16. The star indicates the city of Potenza, while the position of Mt. Etna is indicated by the triangle.

[5] Since the beginning of this eruptive activity, lidar observations of atmospheric aerosols have been performed at the lidar station of IMAA-CNR (Istituto di Metodologie per l'Analisi Ambientale - Consiglio Nazionale delle Ricerche), located in Tito Scalo, Potenza, Italy (40°36′N, 15°44′E, 820 m above sea level), about 330 km north-east of the Etna volcano. This system is part of EARLINET (European Aerosol Research LIdar NETwork), the first lidar network for tropospheric aerosol study on continental scale [*Bösenberg et al.*, 2003]. Following EARLINET protocols, three systematic measurements per week are performed since May 2000, leading to a characterization of the aerosol typically present over our station [*Pandolfi et al.*, 2004]. Additional measurements are performed in the occasion of special events (e.g., Saharan dust outbreaks and forest fires). The IMAA lidar system [*Pappalardo et al.*, 2003] is based on an Nd:YAG laser equipped with second and third harmonic generators. Signals elastically backscattered at 355 and 532 nm are detected, moreover, an inelastic channel is utilized for Nitrogen Raman signal at 386.6 nm. A combined Raman elastic-backscatter lidar permits the independent measurement of the aerosol extinction coefficient and the aerosol backscatter coefficient at 355 nm [*Ansmann et al.*, 1990; *Ferrare et al.*, 1998], and thus of the lidar ratio (*LR*). Aerosol backscatter coefficient at 532 nm is retrieved by using an iterative approach with an assumption on lidar ratio profile; the typical values of lidar ratio used are taken from literature with the support of 3 years of *LR* direct measurements at 355 nm performed at our site [*Pappalardo et al.*, 2003]. Both lidar system and retrieval algorithms have been successfully tested during intercomparison experiments performed within EARLINET [*Böckmann et al.*, 2004; *Bösenberg et al.*, 2003; *Matthias et al.*, 2004].

[6] Starting from the morning after the first eruption of 27 October 2002, at IMAA a special intensive lidar measurement campaign has been performed. In particular, we performed measurements in a nearly continuous way from 1 to 3 November.

[7] Figure 1a shows the evolution of aerosol backscatter profiles at 532 nm starting from the afternoon of 1 November until the night of 2 November. Backscatter profiles at 532 nm were obtained by integrating lidar signals over 5 minutes and with a spatial resolution of 60 m. Looking at the strong gradient in the false colors image of Figure 1a, it can be seen that the Planetary Boundary Layer (PBL) extends up to a maximum of 2–2.5 km above sea level (a.s.l.), in perfect agreement with PBL height typically observed in autumn over Potenza [*Pappalardo et al.*, 2003]. The figure also shows the presence of a considerable aerosol layer at higher altitudes. This observed layer is present at about 5.5 km a.s.l. at the beginning of our measurements (1320 UT, 1 November 2002), then it decreases in altitude, becoming almost stable at about 4 km between midnight and 1100 UT of 2 November and it seems to join the underlying aerosol residual layer at about 1940 UT. This aerosol layer presents a falling speed as a function of altitude that cannot be explained if only sedimentation is considered [*Kasten*, 1968]; air mass motion

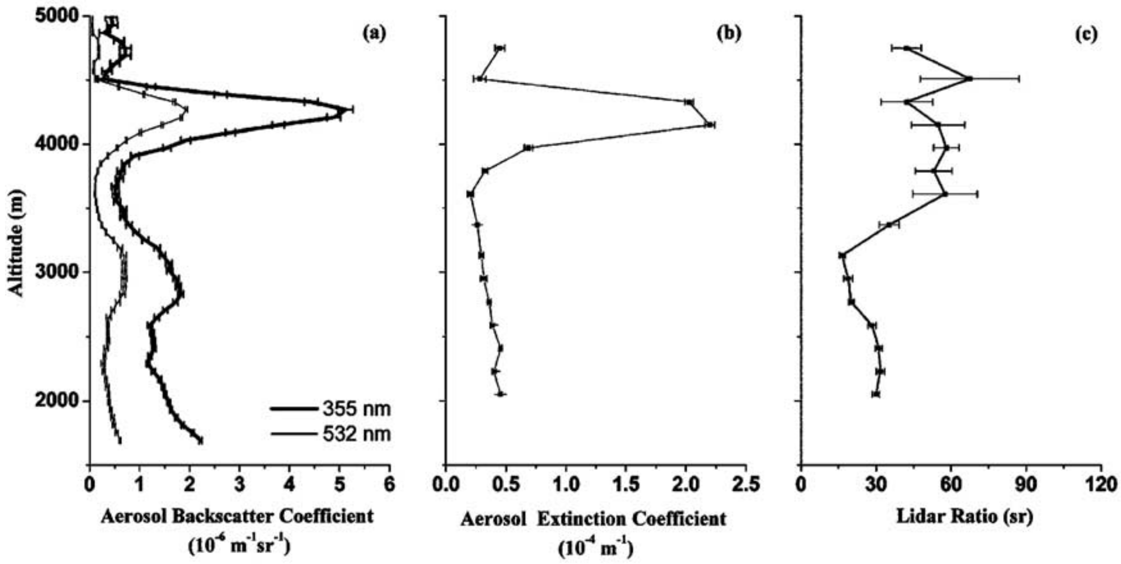

Figure 2. Aerosol backscatter coefficient profiles at 355 and 532 nm (a), aerosol extinction coefficient (b) and lidar ratio (c) profiles at 355 nm, measured on 1 November 2002, from 2320 to 2350 UT. The aerosol backscatter coefficient profiles have a vertical resolution of 60 m, while spatial resolution of the aerosol extinction coefficient and lidar ratio profiles is 180 m.

could explain this behavior and the observed rapid descent with a mean falling speed of about 0.017 m/s. A weaker aerosol layer is present at higher altitudes and shows the same temporal evolution of the underlying aerosol layer. Backward trajectories, available on the EARLINET database [*Bösenberg et al.*, 2003], show, for these altitudes, air mass transport over the Mediterranean Sea and they do not indicate any air mass transport from the Sahara region. In absence of dust events and forest fires, as in this case, aerosol layers at our site typically do not exceed 2.5 km in altitude during autumn and winter [*Pandolfi et al.*, 2004]; therefore the observed aerosol layers can be related to the Etna eruption.

[8] During the 2002 Etna eruption, several AVHRR (Advanced Very High Resolution Radiometer) images have been acquired by the NOAA (National Oceanic and Atmospheric Administration) receiving station operational at IMAA. Three examples of these images, in coincidence with our lidar measurements, are reported in Figures 1b–1d. As shown in Figure 1b, in the afternoon of 1 November, the volcanic plume, seen as a purple trail in the figure, flows toward north-northeast, passing over the Puglia region. Under this condition, the observed anomalous aerosol layer is characterized by a mean aerosol backscatter value at 532 nm of 0.5×10^{-6} m^{-1}sr^{-1}. In the following hours, as shown in the image recorded at 0050 UT, 2 November (Figure 1c), the volcanic plume turns toward north in the direction of Potenza and the observed aerosol layer becomes more intense with a mean aerosol backscatter of 1.2×10^{-6} m^{-1}sr^{-1}. At 1216 UT, 2 November, the wind direction changes again and the plume flows toward western direction over the sea before turning toward east passing over the Calabria region (Figure 1d).

[9] The volcanic aerosol layer observed in the free troposphere has been characterized in terms of vertical profiles of aerosol extinction and backscatter coefficient at 355 nm, aerosol backscatter coefficient at 532 nm, and lidar ratio at 355 nm. Figure 2 reports the aerosol backscatter coefficient profiles at both 355 and 532 nm (a), and the aerosol extinction coefficient (b) and the lidar ratio profile (c), both at 355 nm. All these profiles were obtained using a time integration of 30 minutes extending between 2320 and 2350 UT, when a direct transport of the plume from Mt. Etna to Potenza occurs. A well distinguishable structure is present between 4 and 4.5 km a.s.l.. In this layer, peak values of aerosol backscatter coefficient at 355 and 532 nm are about 5.1×10^{-6} m^{-1}sr^{-1} and 1.9×10^{-6} m^{-1} sr^{-1}, respectively. These values are higher by a factor of 20 when compared to background values measured during the last three years.

[10] Starting from aerosol backscatter at 355 and 532 nm and assuming a power-law wavelength dependence of the aerosol backscatter coefficient ($\sim\lambda^{-\delta}$), we estimate the parameter δ. Within the whole layer (4–4.5 km), the mean estimated value of δ is 2.4. Even if at least three wavelengths backscatter measurements are needed for discriminating between different aerosol types [*Sasano and Browell*, 1989], our estimation can provide some qualitative information about the kind of investigated aerosol. The retrieved value is considerably different from values around 3 that we typically measure in clean tropospheric conditions, and from values around 0.5 observed in Saharan dust outbreaks occurrences. This difference is probably a consequence of the fact that detected volcanic aerosol has an effective radius smaller than that of the Saharan dust.

[11] Figure 2b reports the extinction profile measured from 2320 to 2350 UT with a vertical resolution of 180 m. It shows an enhanced peak of 2.2×10^{-4} m^{-1} at 4.2 km a.s.l., with an optical depth of the whole layer (4–4.5 km) equal to 0.1. Figure 2c shows two regions having an almost constant *LR*: from the first sounded point up to 2.6 km, with a mean value of 30 ± 1 sr, and within the layer extending between

4 and 4.5 km a.s.l., where it assumes a mean value of 55 ± 4 sr. Lidar ratio of 30 sr in the PBL is in agreement with values typically observed during three years of systematic measurements (mean value of 40 ± 12 sr) [*Pandolfi et al.*, 2004]. In fact, about 34% of the mean values calculated within the PBL ranges from 25 to 35 sr. At higher altitudes, lidar ratio measurements have been performed for more than 100 Saharan dust outbreak events, with a typical *LR* value of 47 ± 12 sr for Saharan dust layers observed at IMAA [*Pandolfi et al.*, 2004]. This mean value is in agreement with theoretical estimation obtained for spherical dust particles [*Ackermann*, 1998], while the large variability is related to the existing differences in shape and dimension [*Mishchenko et al.*, 1997]. In the present case, no Saharan dust outbreak has been observed and the aerosol layer in the free troposphere is clearly due to the volcanic eruption. The small variability of *LR* within the layer indicates a very homogeneous aerosol layer in terms of chemical-physical properties. The retrieved *LR* value of 55 sr is the first measured lidar ratio of aerosol particles emitted in the free troposphere during a volcanic eruption. Even if no measurements of the same kind are available for a direct comparison, this measurement is consistent with lidar ratio values reported for urban and industrial aerosol where there is presence of sulfates and soot [*Ferrare et al.*, 2001; *Franke et al.*, 2001; *Matthias and Bösenberg*, 2002]. However, since soot is highly absorbing and results in very high lidar ratios, the observed *LR* value of 55 sr is consistent with sulfate particles with a low amount of soot. On the other hand, the estimated value for δ of 2.4 is consistent with submicron particles [*Bösenberg et al.*, 2003], that is reasonable for young sulfates particles directly transported from the volcano.

[12] In conclusion, we can assert that, the measured lidar ratio of 55 sr together with the estimation of the wavelength dependence of the aerosol backscatter coefficient of 2.4 indicate the presence, in the observed volcanic aerosol layer, of young sub-micron sulfate particles, a low soot content, and the absence of large ash particles.

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