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The impact of Mount Etna sulfur emissions on the atmospheric composition and aerosol properties in the central Mediterranean: A statistical analysis over the period 2000–2013 based on observations and Lagrangian modelling

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HIGHLIGHTS

- Etna's decadal impact in the central Mediterranean is estimated for the first time.
- Transport of Etnean airmasses perturb SO_2 content and aerosols mean size at Lampedusa.
- 40% (SO_2) and 20% (alpha) exceedances at Lampedusa are attributed to Etna's forcing.

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ABSTRACT

The emission of gases and aerosols due to volcanic activity may impact significantly atmospheric composition, cloud occurrence and properties, and the regional and global climate. While the effects of strong explosive (stratospheric) eruptions are relatively well known, limited information on the impacts of small to moderate volcanic activities, including passive degassing, is available. In this paper, the downwind impact of Mount Etna's sulfur emissions on the central Mediterranean is investigated on a statistical basis over the period 2000–2013 using: (a) daily sulfur dioxide emission rates measured near crater at Mount Etna with ground-based ultraviolet spectrophotometers, (b) Lagrangian trajectories and simulated plume dispersion obtained with the FLEXPART (FLEXible PARTicle dispersion) model, and (c) long-term observations of column SO_2 concentration and aerosol Ångström exponent α at Lampedusa (35.5° N, 12.6° E). This statistical analysis has allowed, for the first time, the characterization of decadal impact of Mount Etna's sulfur emissions on the sulfur dioxide and the aerosol microphysical/optical properties in the central Mediterranean. On average, statistically significant higher SO_2 concentrations and smaller aerosol sizes are present when air masses from Mount Etna overpass Lampedusa. Despite being upwind of Lampedusa for only 5% of the time, Mount Etna is potentially responsible for up to 40% and 20% of the SO_2 and α extreme values (exceedances of a fixed threshold), respectively, at this location. The most important factor determining this perturbation is the prevailing dynamics, while the magnitude of the SO_2 emission rates from Mount Etna appears to be likely important only for relatively strong emissions. The observed perturbations to the aerosol size distribution are expected to produce a direct regional radiative effect in this area.

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1. Introduction

The gaseous and particulate matter emissions due to volcanic activity may produce an important impact on the tropospheric and stratospheric composition (von Glasow et al., 2009), the distribution and optical properties of low [e.g., (Gassó, 2008)] and high clouds [e.g., (Lohmann et al., 2003)], the Earth radiation budget from the regional to the global scale, and climate (Robock and Oppenheimer, 2003). The effect on the stratospheric composition and the global radiative balance of strong explosive eruptions (strong enough for the effluents to reach the stratosphere) is dominated by the highly reflective sulfate aerosols formed by the gas-to-particle conversion, involving volcanic sulfur dioxide (SO_2) emissions (McCormick et al., 1995). Due to the long lifetime of sulfate aerosols in the stratosphere (up to a few years for very strong eruptions) and the stratospheric dynamics (dominated by the poleward Brewer-Dobson circulation), the cooling effect produced by these secondary sulfate aerosols can act at the global scale (Hamill et al., 1997). The stratospheric aerosol perturbation and the direct radiative forcing of strong explosive eruptions is relatively well known, and has been observed for several recent eruptions, see, e.g. the cases of Mount Pinatubo (1991) [e.g., (Robock, 2002)], Kasatochi (2008) [e.g., (Wang et al., 2013)], Sarychev (2009) [e.g., (Jégou et al., 2013)] and Nabro (2011) volcanoes [e.g., (Penning de Vries et al., 2014)]. On the contrary, the influence of the more frequent weak volcanic activity, including passive degassing, on the tropospheric aerosol properties and on the regional radiation budget is still largely unknown. As for stratospheric eruptions, most of the radiative effects of moderate eruptions are associated with changes of the aerosol size distribution, composition, and shape. Emission of primary particles, mainly ash, and secondary aerosols through gas-to-particle conversion of volatile sulfur compounds affect the aerosol properties.

Mount Etna is one of the most important emitters of gases and particles on Earth, accounting for about 10% of the global average volcanic emissions of carbon dioxide and SO_2 (Burton et al., 2013). Its continuous degassing and episodic explosive eruptions are important sources of particles and gases for the Mediterranean atmosphere. The total mass of gaseous sulfur compounds emitted by Mount Etna is estimated to be 0.7×10^6 tonnes of sulfur per year, which corresponds to about ten times the anthropogenic emissions in the same area (Graf et al., 1997). Please note that the emissions estimations by Graf et al. (Graf et al., 1997). are based on data which date back to the end of the 1980s. Since then, the anthropogenic SO_2 emissions in Europe have been drastically reduced (up to about 75% from the year 1990 to the year 2010, see, i.e. http://www.eea.europa.eu/data-and-maps/indicators/eea-32-sulphur-dioxide-so2-emissions-1/assessment-3). Thus, the volcanic source of sulfur compounds has become more and more important, relatively to the anthropogenic source, during the last few decades. In addition, volcanic sulfur emissions are more efficient climate forcers than anthropogenic (pollution) emissions because they are released at higher altitudes, and the produced sulfate aerosols have longer lifetimes (Stevenson et al., 2003). The impact of Mount Etna on the atmospheric composition, the aerosol chemical, microphysical, and optical properties, the cloud occurrence and properties, the radiative balance and the regional climate in the Mediterranean are not sufficiently known and probably underestimated. Recently, using an individual case study, it has been shown that Mount Etna emissions may produce a detectable effect on the aerosol microphysical and optical properties in the central Mediterranean, during periods when this region is exposed to volcanic air masses originating from Mount Etna (Sellitto et al., 2016). This perturbation to the local and regional aerosol layer has the potential to produce non-negligible radiative perturbations. Using radiative transfer simulations, the radiative forcing efficiency (radiative forcing per unit aerosol optical depth) of modelled Etnean volcanic plumes has been found comparable with the efficiency of the frequent Saharan dust or pollution transport events in the Mediterranea area (Sellitto and Briole, 2015). The analysis of Sellitto et al. (Sellitto et al., 2016) was based on a single case study, and a more extended, long-term characterization of this impact is still lacking.

In this paper, the downwind impact of Mount Etna's SO_2 emissions in the central Mediterranean is estimated over the period 2000–2013 using the long-term observations of SO_2 column and aerosol Ångström exponent α at the ENEA (Italian National Agency for New Technologies, Energy, and Sustainable Economic Development) Station for Climate Observations on the small island of Lampedusa (35.5° N, 12.6° E).

It must be pointed out that in the Mediterranean SO_2 may originate from different sources, both natural and anthropogenic (e.g., in addition to the volcanic source, combustion, ships, biogenic and marine sources [e.g., (Becagli et al., 2012; Calzolai et al., 2015)]). Similarly, fine particles are associated with polluted air masses and secondary aerosols, with biomass burning particles [e.g., (Pace et al., 2005, 2006)], and with new particle nucleation cases, e.g. [e.g., (Pace et al., 2015)]. Moreover, observed column optical properties may be the result of the overlapping of different types of particles at different altitudes. Thus, the determination of the volcanic source based only on values of SO_2 column and aerosol Ångström exponent is intrinsically equivocal, and a more complex approach is necessary. Therefore, the determination of a possible Etna influence on the SO_2 column and aerosol column optical properties must be made on a long-term statistical basis, combining available information on the source and on the transport processes.

The observations at Lampedusa are thus linked to the information on the volcanic source, in terms of 1) the dynamical processes, using a long series of trajectories and plume dispersion calculations obtained with Lagrangian modelling, and 2) Mount Etna SO_2 emissions derived from the long-term series of daily SO_2 emission rates near-source measurements. This statistical analysis allows, for the first time, the characterization of the impact of Mount Etna's sulfur emissions to the SO_2 concentrations and the aerosol microphysical/optical properties in the central Mediterranean, and then possibly on the radiative budget.

The paper is structured as follows. Data and methods used in this work are introduced in Sect. 2; the results of our analysis are given in Sect. 3; conclusions are drawn in Sect. 4.

2. Data and methods

2.1. Near source and downwind observations

2.1.1. Sulfur dioxide emission rate measurements at Mount Etna

Measurements of bulk SO_2 emission rates from the volcanic plume of Mount Etna have been conducted routinely between years 2000 and 2014 using remote ground-based ultraviolet (UV) spectroscopy techniques. Three different spectrometers and methods have been employed throughout the 15-year period: discrete sampling by COrrelation SPECtrometer (COSPEC) (Stoiber et al., 1983; Caltabiano et al., 1994), CCD (charge-coupled device)-based ultraviolet–visible devices [e.g (Galle et al., 2003; McGonigle et al., 2003).], and automatic observation using an array of permanent ultraviolet scanning spectrometers (FLAME - FLux Automatic Measurement) (Salerno et al., 2009a). The different instruments were replaced gradually over the course of years, after comparing and validating the results obtained by the three techniques (Salerno et al., 2009b).

SO_2 measurements have been carried out with COSPEC and