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Abstract

The impact of small to moderate volcanic eruptions on the regional to global radiative forcing and climate is still largely unknown and thought to be presently underestimated. In this work, daily average shortwave radiative forcing efficiencies at the surface ($\mathrm{RFE}_d^{\mathrm{Surf}}$)*, at top of the atmosphere* ($\mathrm{RFE}_d^{\mathrm{TOA}}$) *and their ratio (f), for upper tropospheric volcanic plumes with different optical characterization, are derived using the radiative transfer model UVSPEC and the LibRadtran suite. The optical parameters of the simulated aerosol layer, i.e., the Ångströem coefficient (α), the single scattering albedo (SSA) and the asymmetry factor (g), have been varied to mimic volcanic ash (bigger and more absorbing particles), sulphate aerosols (smaller and more reflective particles) and intermediate/mixed conditions. The characterization of the plume and its vertical distribution have been set-up to simulate Mount Etna, basing on previous studies. The radiative forcing and in particular the f ratio is strongly affected by the SSA and g, and to a smaller extent by α, especially for sulphates-dominated plumes. The impact of the altitude and thickness of the plume on the radiative forcing, for a fixed optical characterization of the aerosol layer, has been found negligible (less than 1% for* $\mathrm{RFE}_d^{\mathrm{Surf}}$, $\mathrm{RFE}_d^{\mathrm{TOA}}$ *and f). The simultaneous presence of boundary layer/lower tropospheric marine or dust aerosols, like expected in the Mediterranean area, modulates only slightly (up to 12 and 14% for* $\mathrm{RFE}_d^{\mathrm{Surf}}$ *and* $\mathrm{RFE}_d^{\mathrm{TOA}}$*, and 3 to 4% of the f ratio) the radiative effects of the upper tropospheric volcanic layer.*

I. Introduction

Volcanoes can have a direct effect on the atmospheric radiation budget, because of the the absorption and scattering of solar and terrestrial radiation by the emitted primary and secondary aerosols. The most important radiative forcing from volcanic emissions is the shortwave (solar) interaction by long-lived secondary sulphate aerosols, formed by the conversion of sulphur dioxide emissions [Oppenheimer et al., 2011]. Primary ash particles can also significantly modulate the radiation transfer. Volcanic emissions forcing,

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in particular from sulphate aerosols, has been proposed as one of the possible causes of the global warming hiatus observed in the last 15 years [Santer et al., 2014]. A more detailed estimation of the radiative effects of volcanic emissions is hampered by the incomplete knowledge of their physico-chemical processes, especially in the upper troposphere, and in particular the mechanisms of new particle formation leading to sulphate aerosol droplets [Andreae, 2013].

Mount Etna is a hotspot sulphur dioxide source in the Mediterranean basin, accounting for more than $0.7{\cdot}10^6$ t S / yr, 10 times stronger and with injection altitudes significantly higher than anthropogenic sulphur emissions in the same area [Graf et al., 1997]. Despite its potential to form long-lived sulphate aerosols, the radiative forcing of Mount Etna's emission is largely overlooked in aerosols impact studies in the Mediterranean.

In this paper, the surface and top of the atmosphere (TOA) radiative forcing of simulated upper tropospheric volcanic plumes, with different optical characterization, is derived with a forward radiative transfer model. The aerosol layer (optical properties and vertical distribution) and environmental conditions (atmospheric profiles, surface albedo and properties of simultaneous aerosol layers at lower altitudes) are set-up to mimic Mount Etna emissions and the Mediterranean environment. Methods are described in Section II, results are discussed in Section III and conclusions are given in Section IV.

II. Methods

The potential radiative impact of Mount Etna emissions in the Mediterranean basin are estimated by means of the shortwave surface and TOA radiative forcing. These two quantities are calculated using the UVSPEC radiative transfer model and the LibRadtran suite [Mayer and Kylling, 2005]. The general set-up of our simulations is reported in Table 1.

Table 1: *General set-up of UVSPEC. (SDISORT=[Pseudo-]Spherical Discrete Ordinate Radiative Transfer, LOWTRAN=Low Resolution Atmospheric Radiance and Transmittance Model, AFGL=Air Force Geophysics Laboratory)*

Radiative transfer solver	SDISORT [Dahlback and Stamnes, 1991]
Solar spectrum	[Kurucz, 1994]
Gas absorption model	LOWTRAN [Pierluissi and Peng, 1985]
Atmospheric state	AFGL mid-latitude summer standard
Surface albedo	0.09, as in [Meloni et al., 2005]
Spectral range	300.0-3000.0 nm
Spectral resolution	0.1 nm

The volcanic aerosol layer is modelled by means of its (wavelength-independent) optical properties and vertical distribution. The optical characterization of the aerosols has been defined by means of the single scattering albedo (SSA), Ångström exponent (α) and asymmetry parameter (g). These three parameters describe the absorption properties, the size distribution of the aerosols, and the angular distribution of the radiation field after the interaction of the layer, respectively. Different simulations are performed with varying parameters values; the explored intervals are summarized in Table 2. These simulations are discussed in Section III.1. To gather indications on the impact of Mount Etna aerosols and gaseous precursors emissions in the Mediterranean, this work is focused on aged volcanic plumes, i.e., plumes where the gas-to-particle conversion of SO_2 emissions has enriched the plume of sub-micrometric sulphate droplets and/or the coarse ash component is significantly reduced by gravitational settling. At these conditions, typical values of the mentioned optical parameters vary from 1.0 [Hervo et al., 2012, e.g.] to more than 1.6 [Watson and Oppenheimer, 2001, specific determination for a non-ash-bearing plume of Mount Etna] (α), from 0.8-0.9 [Derimian et al., 2012] to 1.0 (SSA) and from 0.7-0.8 [Derimian et al., 2012] to 0.7 (g), for ash and sulphate aerosols, respectively. In general ash is characterized by bigger (smaller α), more absorbing (smaller SSA) particles. The variability of g is of less immediate interpretation because it depends on both the size and the shape of the aerosol particles. In any case, slightly smaller values are expected for sulphate aerosol layers, due to the very small mean sizes of these particles. The variability of the aerosol optical properties of these simulations are then tought to mimic plumes dominated by sulphate aerosols, ash and mixed layers of these two compositions. Finally, a total of more than 1000 (11x13x7 optical properties combinations) aerosol scenarios are tested.

Table 2: *Variability of the aerosol properties. (Par.=parameter, Min=minimum, Max=maximum, Incr.=increment, N=number of different values)*

Par.	Min	Max	Incr.	N
SSA	0.80	1.00	0.02	11
α	0.6	1.8	0.1	13
g	0.60	0.90	0.05	7

The vertical distribution of the plume has been modeled by setting its aerosol optical depth (AOD) profile. In our simulations, the AOD has a maximum value at 11 km and linearly decreasing to zero at higher (up to 13 km) and lower altitudes (down to 8 km), see the distribution in red in Figure 1. The thickness of the plume is then 6 km. Different altitudes of the plume, i.e., with a rigid shift of the distribution 2 km upwards and downwards (distributions in orange and brown in Figure 1) are also tested to estimate the impact of the plume altitude on the radiative forcing. The impact of different thicknesses of the plume have also been investigated by compressing the vertical distributions in a 3 km vertical interval, with constant AOD (distributions not shown in Figure 1). The results of these simulations are discussed in Section III.2. Finally, the impact of the the presence of different aerosol typologies in the lower troposphere (LT, vertical distribution in blue in Figure 1) is studied and the results are discussed in Section III.3.

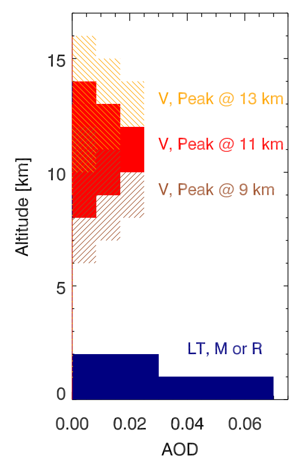

Figure 1: *Vertical AOD profiles used in the simulations. (V=volcanic, LT=lower troposphere, M=maritime, R=rural)*

A further baseline simulation is carried out, with the same atmospheric state but without aerosols. Starting from the high spectral resolution spectra, the instantaneous shortwave radiative forcing has been calculated as the difference between the net flux with and without aerosols, integrated in the whole spectral range. The radiative forcing per unit of AOD, also called radiative forcing efficiency (RFE), is discussed in the following rather than the absolute radiative forcing. This is done to obtain more general results and because it is difficult to single out the optical depth of volcanic-only aerosols in *real world* mixed aerosol layers. The simulations are made at different solar zenith angle values (from 0 to 90°, with 15° steps) and the daily mean value of the radiative forcing efficiency at the surface and at the TOA (RFE_d^{Surf} and RFE_d^{TOA}), with the different aerosol characterization, is finally calculated (equinox simulations). The ratio f between RFE_d^{Surf} and RFE_d^{TOA} is important to determine the reflective/absorbing properties of the layer and is also discussed in the present work.

III. Results and discussion

III.1 Dependence on volcanic aerosol optical properties

The radiative forcing of the simulated volcanic plume varies significantly with the three varying aerosol optical parameters. RFE_d^{Surf} and RFE_d^{TOA} vary between about -12 and -118 $W/m^2/AOD$ and about 0 and -66 $W/m^2/AOD$, respectively (results not shown here). The variability of the ratio f is shown in Figure 2. F varies between about 5 (very absorbing layer; higher values of f for small SSA and big g; slightly facilitated for high α) and 0.8 (very reflective layer; smaller values of f for bigger SSA and small g; at these conditions f is weakly sensitive to α). The most sensitive parameter determining f in our simulations is SSA, while g can also be important. The red cross in Figure 2 indicates the measured values of SSA, α and g for the test case of [Sellitto et al., 2015]. Plume dispersion simulations and observations for that case study suggest that the plume is composed of sulphate aerosol; an f of about 1.0 was found. The present simulations suggest that, in this area of the SSA-α-g space, f does not vary rapidly with varying parameters. We conclude that sulphate aerosols plumes tend to have stable radiative forcings (reflective) for moderate oscillations of the optical properties (e.g., disregarding the micro-physics and composition of the sulphates or in presence of an ash component). Ashy plumes, on the contrary, can be very absorbing (comparable with dust or urban tropospheric aerosols in the Mediterranean, see e.g. [Di Biagio et al., 2010]) and their radiative forcing has the potential to vary more abruptly with varying optical properties.

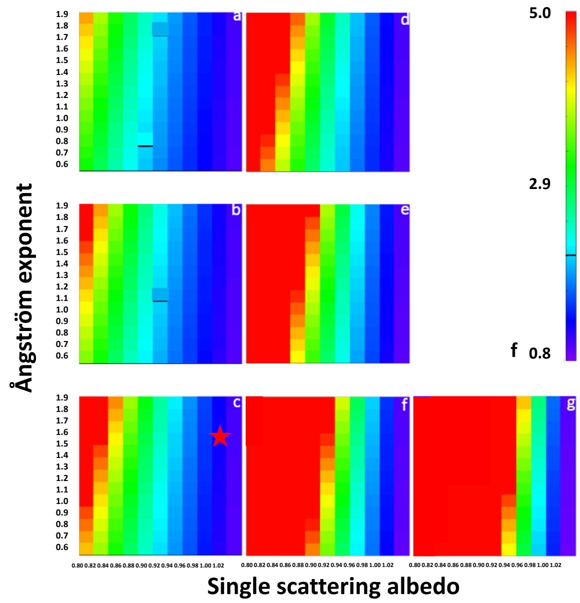

Figure 2: *F ratio as a function of single scattering albedo SSA and Ångström coefficient α, for asymmetry factor g varying between 0.6 (a) to 0.9 (g). The red star indicates the SSA, α and g values for the case study discussed in [Sellitto et al., 2015].*

III.2 Dependence on volcanic plume altitude and depth

The impact of different altitudes (orange and brown distributions in Figure 1) and thicknesses of the plume are investigated on selected optical properties, i.e., sulphates-typical values of 0.98, 1.6, and 0.7 for SSA, α and g, respectively. The AOD has been kept constant in these simulations. The RFE_d^{Surf}, RFE_d^{TOA} and f varied for less than 1% with these different configurations, indicating that the altitude and the vertical extent of the volcanic plume has a negligible effect on its radiative forcing. These results are consistent with earlier observations of the small impact of the aerosol vertical distribution on radiative forcing [Meloni et al., 2005].

III.3 Impact of the presence of other aerosols types in the lower troposphere

The impact of the presence of a LT aerosol layer, at lower altitudes than the volcanic plume, is also investigated. To this aim, simulations are performed with: 1) a volcanic-only case (distribution in red in Figure 1), 2) a volcanic plus maritime LT layer (distributions in red + blue in Figure 1), 3) a volcanic plus rural LT layer (distributions in red + blue in Figure 1). The optical characterization of the different layers is derived from [Shettle, 1985]. For the volcanic plume, moderate volcanic conditions are selected. Maritime and rural optical properties are selected to mimic sea salt-preponderant aerosols (maritime) and the dust component (rural), which are two typical conditions for the LT aerosols in the Mediterranean. Background conditions are then chosen as: 1) no aerosol (for the volcanic-only case), 2) LT maritime-only aerosols (for the volcanic plus maritime case), 2) LT rural-only aerosols (for the volcanic plus rural case). The RFE_d^{Surf}, RFE_d^{TOA} and f for the 3 cases are reported in Table 3. The interference of simultaneous LT aerosol layers on the volcanic plume radiative forcing are found relatively small. While the individual surface and TOA components can reach up to 6 W/m^2/AOD smaller values, with respect to the volcanic-only case, the ratio f is only slightly perturbed, indicating a very small additional absorption component. Please note that more consistent variations of the radiative forcing, due to the simultaneous presence of LT aerosols, are found for other aerosols types, like dust, in the Mediterranean [Gómez-Amo et al., 2010]. One marked difference with respect to our simulations is the smaller altitude of the dust plume (1.5-4.0 km).

Table 3: *Radiative forcing efficiencies at the surface and the TOA, and their ratio f, for 3 cases: volcanic-only (V-only), volcanic + maritime (V+M), volcanic + rural (V+R).*

Conf.	RFE_d^{TOA}	RFE_d^{Surf}	f
V-only	-47.80	-47.83	1.00
V+M	-41.37	-42.85	1.04
V+R	-41.04	-42.08	1.03

IV. Conclusion

Simulations have been carried out to estimate daily average shortwave radiative forcing efficiencies (RFE_d^{Surf}) and (RFE_d^{TOA}), and their ratio f, for upper tropospheric volcanic plumes with different optical characterization. We have shown how the optical characterization of the layer affects the reflective/absorbing nature of the plume. The dominating factors are SSA and g, with α being less important, especially with modelled sulphate-dominated layers. The altitude and thickness of the plume has a negligible impact on the radiative forcing, for a fixed optical characterization. The simultaneous presence of boundary layer/LT Mediterranean aerosols (marine or dust aerosols) affects for only up to 12 and 14% (RFE_d^{Surf} and RFE_d^{TOA}), and 3-4% (f) the radiative effects of the upper tropospheric volcanic layer. More detailed simulations and dedicated observations are required to better constrain the volcanic radiative forcing in *real world* conditions, especially for a volcano like Mount Etna, which has a particularly complex and varied activity.

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