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EPL-RADIO (Etna Plume Lab - Radioactive Aerosols and other source parameters for better atmospheric Dispersion and Impact estimatiOns)

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Introduction and motivation

In the last few years, various experiments were set up at Mt. Etna by ENS (Ecole Normale Supérieure, Paris) researchers and in particular the PI, to analyse the properties of volcanic gasses and aerosols and their impact on the atmospheric chemistry and radiative balance/climate from local to regional scale. These activities were conducted under the umbrella of a multidisciplinary research cluster called EtnaPlumeLab (EPL) [1]. In the context of the EPL cluster, **EPL-RADIO project aims at improving the characterization of Mount Etna as atmospheric aerosols source, gathering multi-domain (solid Earth and atmospheric sciences) information.** The target of this research are emission processes, from inner degassing mechanisms to aerosol near-source characterisation.

Scientific objectives

This project brings together a variety of innovative information, exploiting expertise from both atmospheric sciences and volcanology specialists. This information is derived from the observation of: a) coupled direct/remote size-resolved aerosols distribution and composition, b) primary/secondary near-source sulfate aerosols partitioning and c) radioactive disequilibria of radon daughters. These observations have systematically been coupled with the detailed characterisation of the environmental conditions, in terms of the plume's gasses composition and its thermodynamics. The results relative to points a) and b) allow a **detailed size-resolved physico-chemical characterisation of the emitted aerosols to constrain the regional climatic impacts of Mt. Etna in the Mediterranean area** (from local to regional scale). With regards to the latter, it should be mentioned that it has been recently demonstrated that Mt. Etna's emissions have the potential to significantly modulate the atmospheric composition, the optical properties of the distal aerosol layer [2] and the radiative balance in the Mediterranean area [3], thus producing regional climate forcing, which depends on the chemical and micro-physical characterisation of the emitted and produced (in-plume) aerosols [4]. The results of point c) provide **new information on inner degassing dynamics**.

Campaign C1
Campaign C2 - 2016

Day	LiDAR	MicroTops	Direct sampling	Gas observations
30/06	Raman+Elastic		@BN crater	UVS (@BN crater) FTIR (@BN crater)
01/07	Raman+Elastic			
14/07		1 distal		
18/07		3 plume section		
19/07	Elastic	1 proximal 1 distal	@BN crater	UVS (@TDF) FTIR (@BN crater)
20/07		1 proximal 1distal		UVS (@SLN) FTIR (@SLN, TDF)
22/07			@V, NE crater	

Campaign C3 - 2017

Day	LiDAR	MicroTops	Direct sampling	OPC	Gas observations
18/07		1 distal (8 km) 1 very distal (18 km)			FTIR (distal – sync with MII)
19/07	Raman+Elastic	1distal (5 km)			UVS (distal) FTIR (distal – sync with MII)
20/07		2 distal (12 km)	@BN,V crater	@BN,V crater	UVS (@BN,V crater) FTIR (@BN,V crater)
21/07		1 distal (12 km) 1 proximal (0.5 km) - lateral			FTIR (distal – sync with MII)

Table 1: Experimental set-up for EPL-RADIO campaigns C1, C2 and C3.

Methodology and experimental set-up

In the context of EPL-RADIO, **three measurement campaigns have been carried out at Mount Etna active craters and the surrounding area, during summers 2016 (Campaigns C1 and C2) and 2017 (Campaign C3)**. The experimental set-up for the three campaigns is summarized in **Table 1**. The volcanic aerosol source has been

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characterised by determining the size-resolved aerosol emissions, with cascade impactors, an optical particle counter (OPC) and two Microtops-II (MII) sun-photometers (one operating in the UV/NIR – MIIOM model – and one in the VIS/NIR – MIISP model – spectral ranges), and the primary fraction of the emitted sulfate aerosols (with respect to the secondary sulfate aerosols produced by in-plume conversion of SO2 emissions) by Fourier Transform InfraRed spectroscopy. The extreme portability of MII was exploited to provide quasi-simultaneous characterisation of the optical properties of the plume in an extended area around the active craters (from craters edge to ~20 km downwind). Complementary three-dimensional aerosols information have been obtained by means of a multi-wavelength polarimetric scanning elastic/Raman LiDAR (Light Detection And Ranging) system at a fixed station (Serra La Nave observatory, 7 km from the active craters). The radioactivity characterisation of the plume was carried out through direct sampling and subsequent laboratory analyses at LMV Clermont-Ferrand.

Preliminary results and conclusions

First results of the analysis of EPL-RADIO observations can be grouped into three general categories: 1) new methodological developments, 2) small-scale volcanic aerosols variability and atmospheric processes, and 3) radioactive characterization of the plume and inner degassing dynamics. These are briefly discussed in the following.

New methodological developments

A number of novel observation methodologies have been produced or are under development. A new methodology for the determination of ozone-corrected UV (320 nm) volcanic AOD and UV/NIR Angström parameters, using MIIOM portable sun-photometer has been developed [3]. This has allowed the generation of the first dataset of UV optical properties of volcanic aerosols, possibly accessing more specific information on small-sized particles like secondary aerosols from volcanic gases condensation. Co-located proximal and distal MIIOM and UVS observations will allow the development of SO2-correction procedures for UV volcanic AOD photometric observations. During C3, MIIOM (UV+NIR) and MIISP (VIS+NIR) observations have been systematically coupled and an algorithm to couple, for the first time, extinction information of volcanic aerosol in such a large spectral range (UV+VIS+NIR) is under development. New inversion schemes to extract the size distribution information from this extended spectral extinction information are under study, as well. A new inversion algorithm is under development to extract, for the first time, aerosol information, in combination with gases information, from FTIR high-spectral-resolution observations.

Small-scale volcanic aerosols variability and atmospheric processes

The EPL-RADIO project allowed, for the fist time, the investigation of the small-scale variability of Mount Etna volcanic aerosols properties (within a few km from the volcanic source) and the inherent processes. The MIIOM method described in [3] has been used to characterise Mt. Etna distal and proximal plumes. Results of quasi-simultaneous characterisation of proximal and distal plume (7 km from craters), for 20 July 2016 have shown that, even if these plumes were observed within approximately only one hour, their average size varied significantly, thus indicating quick sedimentation of ash particles. In general, **the ensemble of EPL-RADIO observations shows that small-scale chemical/microphysical processes play a major role in the determination of wider-scale aerosol optical properties**. The series of MII observation at different sites provided the first three-

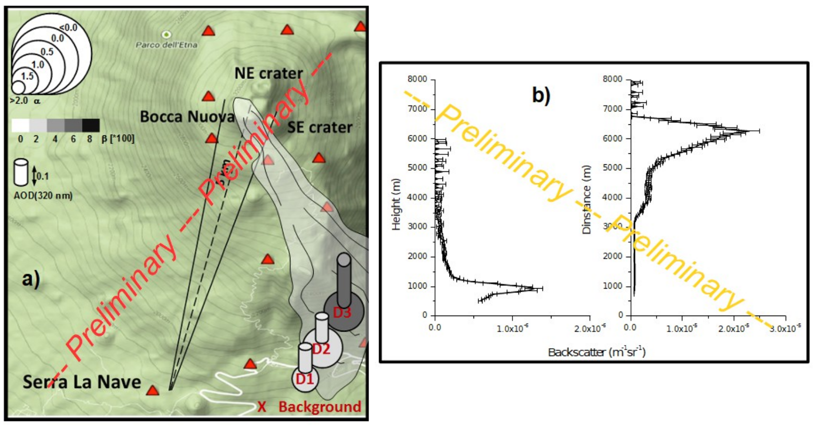

Figure 1: **a)** an example of optical properties of volcanic aerosol observed during a plume section with MIIOM on 18/07/2016 (C2); **b)** an example of zenith-vertical and crater-looking LiDAR observations series on on 19/07/2016 (C2).

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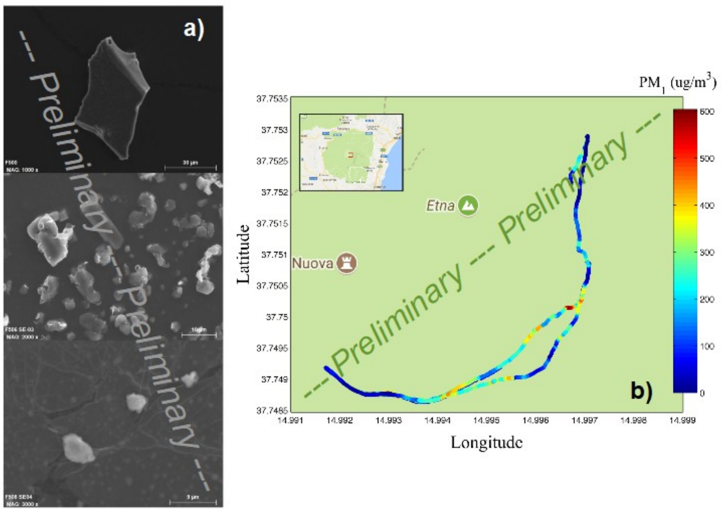

Figure 2: **a)** Scanning electron microscope pictures of typical grains of volcanic aerosols collected during the EPL-RADIO campaigns (C1, C2 and C3). From top to bottom: single silicate glass fragment, pluri-micron Cu-alkali sulfates from the aphitalite group, micron alkali chlorides with infra-micron SO_4 droplets. **b)** Small-sized particle matter concentration (PM1) from OPC at central craters during a plume traverse on 20/07/2017 (C3).

dimensional short-term (1 week for C2 and C3) map of volcanic aerosols properties (mean size, burden/composition) with portable photometry. First analyses of these data show: 1) very strong variability of aerosol properties at small spatial scales (model sub-grid/satellite observations sub-pixel), 2) bigger particles with bigger burden at proximal locations, important role of sedimentation processes, 3) big day-to-day variability, and 4) particle mean size and burden gradients between central/lateral plume sections (see an example for a plume section in **Fig. 1a**). In addition to MII, LiDAR observations (zenith and crater-pointing, see an example of quasi-simultaneous observations with the two geometries in **Fig. 1b**)) provided further vertically-resolved information. During the C2 and C3 campaigns, two periods of time characterised by non-eruptive volcanic activity, LiDAR observations revealed, in general, an aerosol layer: 1) at low altitude (from near-ground to ~3km) over the Serra La Nave station, 2) in many cases, covering the whole line of sight, from station to crater, 3) weakly depolarising, 4) probably absorbing. This point at big ash particles at the source, very important sedimentation processes, possible adsorption of smaller ash particles into rapidly forming sulphate droplets, resulting in layers of sulphate aerosol droplets with an absorbing core, with varying size but covering large areas between the crater area and Serra La Nave. Finally, summit area direct sampling revealed a complex composition at the source (an example of scanning electron microscope pictures is in **Fig. 2a**) and OPC observations suggest very high particle matter concentration at the summit with the plume associated with a consistently high fine-to-coarse particle ratio (an example of PM1 map is in **Fig. 2b**).

Radioactive characterization of the plume and inner degassing dynamics

A special emphasis of summit sampling activities was given to the analysis of the natural radioactivity of aerosols, which mainly arises from the presence of chemical compounds of U-series isotopes. Historically, Mt. Etna is the place where short-lived radioactive disequilibrium measurements in volcanic gases were initiated more than 40 years ago; in the framework of EPL-RADIO, this activity were revived more than two decades after the last field campaign and already led to the publication of a first paper on this topic [5]. ^{226}Ra daughters (namely ^{222}Rn, ^{210}Pb, ^{210}Bi and ^{210}Po in their successive order of appearance in the ^{238}U decay chain) are significantly enriched in volcanic gases since they form gaseous compounds at magma temperature. Once released in the cold atmosphere, such gaseous species are quenched into solid particles (see an example of these particles pictures in **Fig. 2a**), mixed with air and diluted accordingly, thus allowing their sampling by filtration of the diluted plume on a raw membrane. Aerosol and gas sampling was carried out during campaign C1 and C3 and subsequent analyses were performed at LMV Clermont-Ferrand. Preliminary analyses shown that the majority of samples present significant enrichment in ^{210}Pb, ^{210}Bi, ^{210}Po compared to the local atmosphere, which confirms that **Mt. Etna is a major source of natural radioactivity injected into the atmosphere**. In addition, the calculated average (^{210}Po/^{210}Pb) and (^{210}Bi/^{210}Pb) activity ratios of the magmatic vapor are significantly higher than those determined in 2015 during a preliminary survey prior to EPL-RADIO characterized by active volcanic activity [5]. These preliminary results suggest that **gases released during quiescent degassing periods (2016 and 2017) come from a shallower part of the plumbing system of the volcano than those released during Strombolian to paroxysmal eruptions (2015)** .

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Multidisciplinary approach

The EPL-RADIO effort is intrinsically and deeply multidisciplinary. The overarching goal of the general EPL research cluster is the systematic understanding of Mt Etna impact on the atmospheric composition and regional climate in the Mediterranean region, thus bridging the gap between small-scale volcanology/geochemistry and large-scale atmospheric dynamics, chemistry and physics. This synoptic volcanological/atmospheric sciences view is particularly exploited in the EPL-RADIO component, that targets specifically the volcanic source and first plume processing before large-scale impacts. The EPL-RADIO experience was particularly fruitful in terms of methodological synthesis and exchanges between experts in atmospheric observations (e.g., atmospheric spectroscopic and remote sensing) and volcanology/geochemistry measurements (e.g., summit samplings and laboratory analyses). **A new cross-domain collaboration (LMD-ENS Paris – experts in atmospheric sciences, INGV and LMV – experts in volcanology and geochemistry) is initiated and is expected to provide a new integrated European core of expertise capable to aggregate other institutions and experts around this multidisciplinary topics**. In addition, new didactic activities are initiated during this project.

Outcome and future studies

The EPL-RADIO biennial (2016-2017) transnational access to Mt. Etna observatory has allowed the detailed characterisation of this volcanic aerosol source by means of novel campaign protocols and coupling different observations (direct measurements + remote sensing – including multi-spectral, surface + column + vertically-resolved information, plume's gases + aerosols characterisation from the crater to several km downwind, etc). Thanks to the synergy of observations from: 1) a multi-spectral polarimetric scanning elastic/Raman LiDAR, 2) a multi-spectral combination of two portable sun-photometers, 3) an optical aerosol counter, 4) direct sampling of particles and gases at the craters, and 5) gas observations with Fourier transform infrared spectrometry and ultraviolet spectrometry, **an unprecedented information has been gathered on the three-dimensional distribution of near-source volcanic aerosols and their emissions dynamics, their space-time variability and small-scale evolution processes**. Studies are ongoing to fully exploit this dataset, which are discussed above (in **Preliminary results and conclusions**). These future studies will provide new knowledge like: 1) the first analysis of short-term variability of aerosol optical properties around the crater and in an extended nearby area (including maps), 2) the analysis of this variability as a function of the plume and atmospheric chemical and thermodynamic characterisation, 3) the local radiative balance perturbation of volcanic aerosols and the interaction with transports from other sources, e.g. Saharan dust outbreaks. **This new knowledge will provide constrains of volcanic degassing both for downwind impacts on atmospheric composition and climate studies, on one side, and for better understand inner magma extraction dynamics, on the other**. A photographic reportage of campaign C1 has been published on the big-audience magazine GEO (French edition) [6].

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