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Etna Plume Imaging and Chemical Composition (EPICC)

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1. Introduction and motivation

A recent study (Surl et al. 2015), combining in situ and remote measurements of O_3, BrO, and SO_2 in the plume of Etna, confirmed the expected depletion of O_3 by bromine species. However, large discrepancies between measurements and models were noticed, largely due to the uncertainty on the presence of significant amounts of NO_x (=NO+NO_2) in the plume. Significant levels of NO_2 in the Etna plume have never been observed. However, classical remote sensing techniques such as MAX-DOAS and, a fortiori, in situ samplings do not capture the large spatio-temporal inhomogeneities of the plumes. Therefore, transient, localized NO_2 emissions may have been missed. Few instruments are capable of providing quantitative information about the chemical composition of dynamic targets such as volcanic plumes. So far, only SO_2 cameras have been capable of observing large portions of the SO_2 field above volcanoes. Recently, a NO_2 camera has been prototyped and successfully tested on industrial exhaust gases (Dekemper 2016). While the technology is completely different than what is used in SO_2 cameras, their scientific products are similar: slant column densities (SCDs) of the absorber are mapped over the plume image with a much higher spatio-temporal resolution than classically achieved with DOAS instruments, making them better suited for studying dynamic scenes (e.g. smokestacks, volcanic plumes). One of the motivations for this campaign was to take advantage of the broad field of view and high temporal resolution offered by this NO_2 camera in order to seek for transient traces of NO_2 in the plume of Mount Etna. Furthermore, the synergistic exploitation of simultaneously acquired images of a same plume can yield a better 3-dimensional representation of the plume. Such a knowledge would improve the determination of emission fluxes time series for instance, a key quantity for both volcanology, and air quality.

2. Scientific objectives

The scientific objectives of the EPICC campaign are summarized below:

1. Operate for the first time a NO_2 camera pointing at a volcanic plume in order to detect potential traces of NO_2, transient or not. Such a detection would contribute to solving some pending questions in the volcanic plume chemistry.
2. Perform simultaneous acquisitions by SO_2 cameras and the NO_2 camera from two or more locations in order to obtain a dataset enabling progresses in the field of 3-D modelization of plumes.

3. Methodology and experimental set-up

The campaign consisted in installing SO_2 cameras in two or more locations around Mount Etna, at a relatively short distance of the craters owing to the limited visual range reached by these UV instruments. In agreement with the local team, two sites were selected: Pizzi Deneri, and La Montagnola. The former was offering a shelter for the scientists and their instruments and was therefore more convenient. The latter was only accessible during the day, and needed daily transportation of the equipment and the team. The list of instruments deployed during the campaign is given below

Instruments	Location	Owner	Capabilities
NO_2 camera	Pizzi Deneri	BIRA-IASB	NO_2 maps (slant columns)
SO_2 camera #1	Pizzi Deneri	NILU	SO_2 maps (slant columns, UV)
SO_2 camera #2	Pizzi Deneri	NILU	SO_2 maps (slant columns, IR)
MAX-DOAS	Pizzi Deneri	Heidelberg	plume transects, slant column NO_2, SO_2, BrO, OClO
SO_2 camera #3	La Montagnola, Crater 2002-2003	BIRA-IASB	SO_2 maps (slant columns, UV)
SO_2 camera #4	La Montagnola	NILU	SO_2, ash maps (slant columns, UV)
MAX-DOAS	La Montagnola	NILU, op. by BIRA-IASB	plume transects, slant column NO_2, SO_2, BrO, OClO

In total, four SO_2 cameras were used: two at each site, and on the 27th of July, SO_2 camera #3 was operated from the crater of the 2002-2003 flank eruption. Pizzi Deneri also hosted the NO_2 camera, and the MAX-DOAS. The measurement plan was simple: operate the instruments as much as possible during the campaign, and make sure that the main plume remains in the instrument field of view. The clocks of each computer were synchronized before reaching the summit.

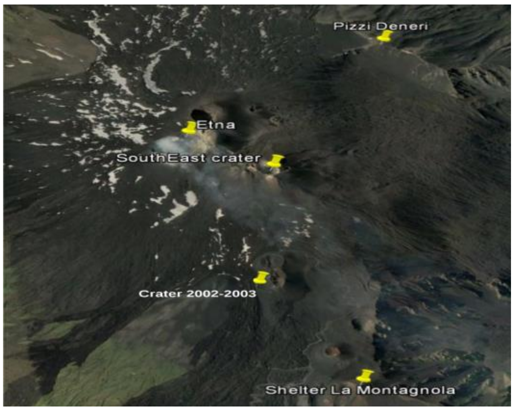
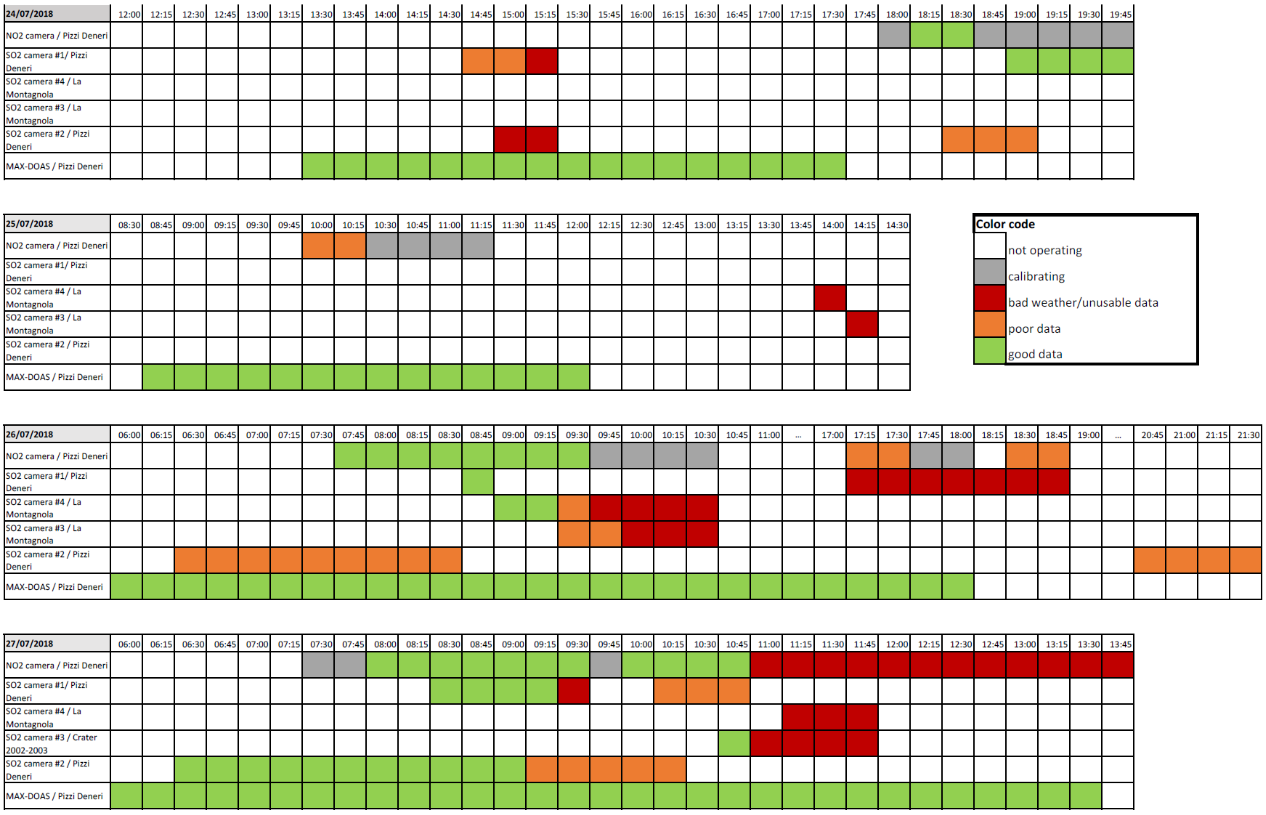


Figure 1: Left: The NO_2 camera at Pizzi Deneri. Right: a map providing the location of the three measurement sites (Pizzi Deneri, La Montagnola and Crater 2002-2003.

4. Preliminary results and conclusions

In this section, we provide some details about the instruments used during the campaign, and a sample of the data acquired. The time table of the instrument operations is given below.



NO_2 camera

Instrument concept and measurement principle: The NO_2 camera is a spectral imager based on an acousto-optical tunable filter (AOTF). The instrument has been optimized for operating in the region 430-470 nm, a

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classical one for NO_2 remote sensing. The rest of the instrument consist of lenses, polarizers, and a CMOS detector. A full description can be found in (Dekemper 2012) and (Dekemper 2016). In standard operations, the camera takes spectral images of the scene (20°x20° field of view, 512x512 pixels) at a number of wavelengths sequentially. A DOAS fitting algorithm is then applied to the spectrum recorded by each pixel in order to retrieve the NO_2 differential slant column density (dSCD). Like other UV-VIS instruments, optimal measurement conditions are achieved when an optically thin volcanic plume is observed with a blue sky in the background.

Sample data and results: Over the few days of the campaign, the measurement conditions were only occasionally fine. The best conditions were found in the morning of the 27/07/2018. Etna was quiet, and its plume was thin while the sky was cloud-free. About 2h of good data could be acquired. Figure 2 shows the results of the retrieved NO_2 field for a slot of 30 minutes. Besides a clear offset (caused by the use of reference spectra not free from NO_2), no clear pattern could be observed.

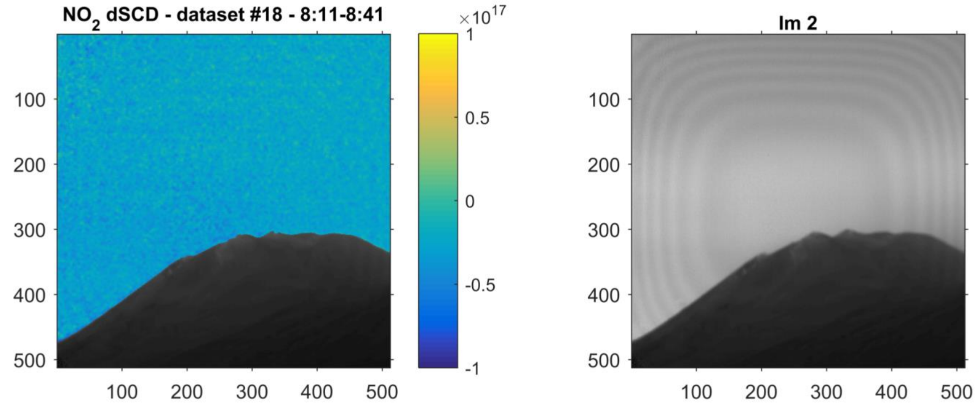

Figure 2: Left: Mean NO_2 dSCD map based on measurements taken between 8:11 and 8:41 on the 27/07/2018. Right: The average spectral image at 449nm indicating that the sky was clear.

SO_2 cameras

Instrument concept and measurement principle: Three SO_2 cameras were based on a UV sensor, and the use of filters allowing to identify the signature of SO_2 in the 310-330 nm region (Mori and Burton, 2006; Bluth et al., 2007). The BIRA-IASB instrument (SO_2 camera #3) was an EnviCam 3 (Nicarnica Aviation), while those from NILU were EnviCams 2 (for specifications see Kern et al., 2015). By comparing the intensities observed in two regions of the UV spectrum, one can first determine the absorbance of the scene, then the SO_2 dSCD. The infrared (IR) SO_2 camera consists of an uncooled microbolometer array, which detects emitted and absorbed radiation in the region 8-14 μm at multiple wavelength channels. The instrument is a successor to Cyclops as described by Prata and Bernardo 2009, 2014).

Sample data and results: As shown in the acquisition time sheet above, the two SO_2 cameras had very few occasions to observe the plume in good conditions. The one located in Pizzi Deneri had a bit more chance and some SO_2 data will be retrievable. The images below are absorbance data acquired by the SO_2 cameras.

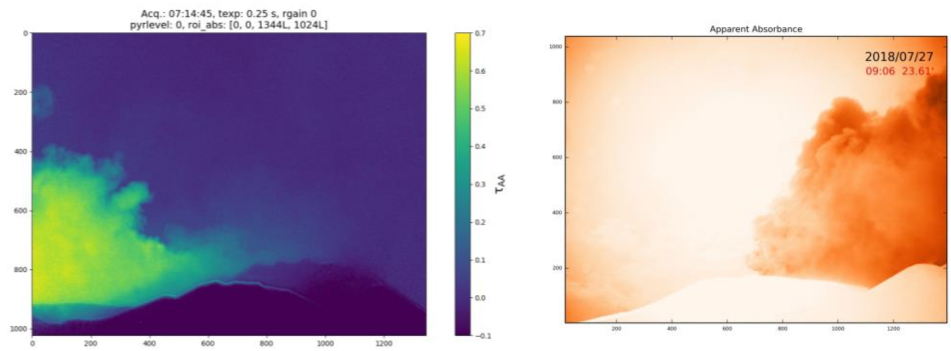

Figure 3: Absorbance maps derived from the SO_2 cameras at Pizzi Deneri (left), and from the site of the crater of the flank eruption of 2002-2003 (right).

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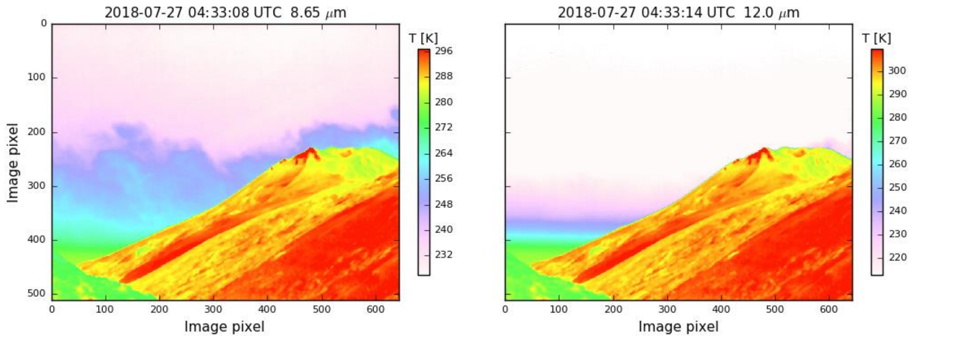

Figure 4: Temperatures from the IR SO_2 cameras at Pizzi Deneri. SO_2 is absorbing in the 8.6 μm wavelength region.

MAX-DOAS

Instrument concept and measurement principle: The instrument was built in the Institute of Environmental Physics in Heidelberg, Germany. It consists of an Avantes spectrograph covering the wavelength range between ~295 nm and ~450 nm with a ~0.6 nm spectral resolution. The spectrograph is actively temperature stabilized and connected to a rotation scanning telescope via a quartz fiber.

Sample data and results: We can measure the differential absorption of SO_2, BrO, OClO and NO_2 in the volcanic plume with respect to a clear sky reference and thereby determine the gas ratios/emissions. **Figure 4** shows a sample dataset for the 26.07.2018 recorded at Pizzi Deneri with a viewing azimuth of 182°N. The four panels show a time series differential column densities for SO_2, BrO, OClO and NO_2, respectively, for the recorded elevation scans. SO_2 is relatively inert in the early plume and can therefore be used as plume tracer. BrO and OClO are reactive intermediates of halogen chemical cycles mostly driven by solar radiation and background ozone. In this study, no NO_2 could be detected within the volcanic plume. The detection limit was around 4E15 molec cm^{-2}.

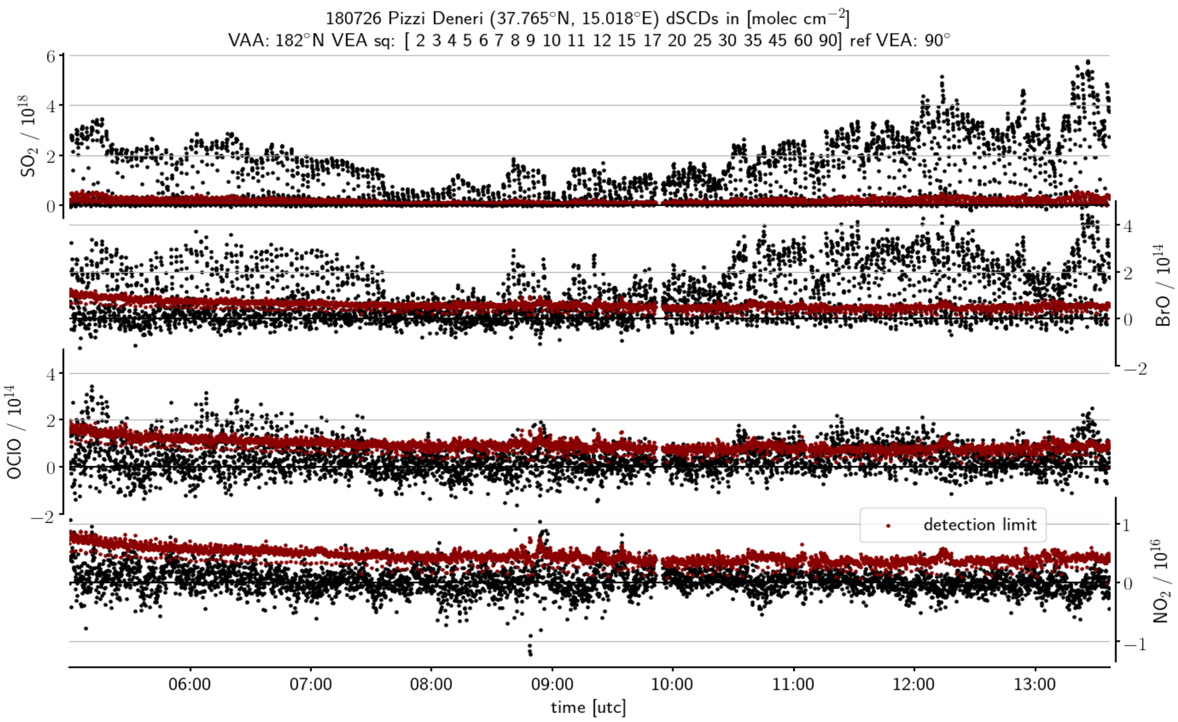

Figure 5: Sample data acquired by the MAX-DOAS at Pizzi Deneri.

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Preliminary conclusions

- Four SO_2 cameras, one NO_2 camera, and 2 DOAS spectrometers were operated successfully during 2.5-3 days on two measurement sites.
- For Pizzi Deneri, about 10% of the total presence time coincided with good measurement conditions. Over this time, all instruments operated nominally. The conditions were even less favorable for the team in La Montagnola. Almost no good data could be taken from there. This was also caused by unfavorable logistical conditions, which prevented the team to arrive early at the site.
- The analysis of the NO_2 camera data revealed that no clear signal of NO_2 could be detected in the plume. This is in agreement with the dSCD time series of the MAX-DOAS instrument.
- The absence of good measurements from La Montagnola makes the second scientific objective unachievable (3-D modelization of the plume). In fact, on 27 July, we expected to be the golden day. SO_2 camera #3 was operating data acquisition from 10h30 to 12h at the 3rd site (crater 2002-2003), and at this time (from 10h30 to 11h) it was the only site with good condition (the 2 other sites were cloudy).

5. Multidisciplinary approach

The EPICC campaign was multidisciplinary in the sense that it was aiming at using an instrument developed for the air quality monitoring theme to a volcanic site in search for traces of NO_2. Furthermore, the simultaneous observation of the volcanic plume with several SO_2 cameras, and the NO_2 camera created the possibility of making progresses in the field of 3-D plume modelization. Such an achievement is desirable both in geophysics and air quality because it would allow for a better estimation of emissions by point sources, a key quantity to these two research areas.

6. Outcome and future studies

The simultaneous observation of the SO_2 contained in volcanic plumes from different point of view is the key to a better estimation of the SO_2 emission fluxes. Campaigns allowing to set up SO_2 cameras in at least three sites for a longer time (weeks) have the best chance to succeed. For EPICC, an air quality instrument (the NO_2 camera) was operated on a volcanic site with SO_2 cameras. Maybe in a future campaign, one should consider to proceed the other way around: bringing the SO_2 cameras to an industrial site such as a coal-firing power plant in order to take advantage of the strong SO_2 and NO_2 plumes.

7. References

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