



## Short communication

## High time resolution fluctuations in volcanic carbon dioxide degassing from Mount Etna

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## ABSTRACT

We report here on the first record of carbon dioxide gas emission rates from a volcano, captured at  $\approx 1$  Hz. These data were acquired with a novel technique, based on the integration of UV camera observations (to measure  $\text{SO}_2$  emission rates) and field portable gas analyser readings of plume  $\text{CO}_2/\text{SO}_2$  ratios. Our measurements were performed at the North East crater of Mount Etna, southern Italy, and the data reveal strong variability in  $\text{CO}_2$  emissions over timescales of tens to hundreds of seconds, spanning two orders of magnitude. This carries important implications for attempts to constrain global volcanic  $\text{CO}_2$  release to the atmosphere, and will lead to an increased insight into short term  $\text{CO}_2$  degassing trends. A common oscillation in  $\text{CO}_2$  and  $\text{SO}_2$  emission rates in addition to the  $\text{CO}_2/\text{SO}_2$  ratios was observed at periods of  $\approx 89$  s. Our results are furthermore suggestive of an intriguing temporal lag between oscillations in  $\text{CO}_2$  emissions and seismicity at periods of  $\approx 300$ – $400$  s, with peaks and troughs in the former series leading those in the latter by  $\approx 150$  s. This work opens the way to the acquisition of further datasets with this methodology across a range of basaltic systems to better our understanding of deep magmatic processes and of degassing links to manifest geophysical signals.

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## 1. Introduction

Carbon dioxide ( $\text{CO}_2$ ) is among the most abundant constituents of volcanic gases (Carroll and Holloway, 1994), and exsolves from magmas deeper than other common volatiles such as sulphur dioxide ( $\text{SO}_2$ ) and water vapour ( $\text{H}_2\text{O}$ ) (Giggenbach, 1996). Knowledge of  $\text{CO}_2$  emissions can therefore contribute significantly to our understanding of the movement of magmas in deep volcanic plumbing systems. Hitherto, the measurement of  $\text{CO}_2$  emission rates has been challenging due to the difficulty of resolving volcanogenic  $\text{CO}_2$  above high background atmospheric levels. In consequence, attempts to routinely measure plume  $\text{CO}_2$  emission rates, particularly at high time resolution, have been rather limited (Aiuppa et al., 2006, 2010). Therefore, notwithstanding the significant contributions made in constraining  $\text{CO}_2$  emission rates of volcanic plumes at targets such as Mt. Erebus, Antarctica (Wardell et al., 2004), Ol Doinyo Lengai, Tanzania (Koepnick et al., 1996), White Island, New Zealand (Werner et al., 2008), Ruapehu, New Zealand (Werner et al., 2006), Redoubt, Alaska (Werner et al., 2012a,b), Stromboli, Italy (Aiuppa et al., 2010, 2011), Mt. Etna, Italy (Allard et al., 1991) and Kilauea, USA (Poland et al., 2012), these data remain relatively spartan, and in general lack information regarding

temporal changes. This remains a fundamental weakness in attempts to constrain global volcanogenic  $\text{CO}_2$  emission rate budgets, in view of which there is a pressing demand for the development and application of novel methodologies to improve constraint on spatio-temporal volcanic  $\text{CO}_2$  degassing and our comprehension of volcanic systems.

Recently, the Multi-GAS technique (Aiuppa et al., 2005; Shinohara, 2005) has been pioneered to enable rapid measurements of volcanic plume chemical compositions, including  $\text{CO}_2/\text{SO}_2$  gas ratios, leading to significant advances in our understanding of degassing processes. Furthermore, in the last years, UV camera imagery has been applied in volcanology, enabling acquisition of  $\text{SO}_2$  emission rates with time resolutions of  $\approx 1$  Hz, many orders of magnitude faster than possible in the past (e.g., Mori and Burton, 2006; Tamburello et al., 2011a). Here we report on the first volcanic deployment of a novel technique, by which volcanic  $\text{CO}_2$  emission rates are captured with an acquisition frequency of  $\approx 1$  Hz, based on the integration of the above two approaches. Such a capability will increase the future potential of linking degassing to geophysical data on unprecedented timescales with significant applicability in improving hazard analysis (Gerlach et al., 2002) and eruption forecasting measures (Aiuppa et al., 2007; Poland et al., 2012).

The  $\text{CO}_2$  emission rate data were captured during a field campaign on Mt. Etna ( $37.734^\circ\text{N}$ ,  $15.004^\circ\text{E}$ ), an alkaline strato-volcano whose  $\text{CO}_2$ -rich magmas (Spilliaert et al., 2006) result in the volcano being the largest time averaged contributor to global volcanic emissions of  $\text{CO}_2$  (Allard et al., 1991; Gerlach, 1991). Etna currently has four

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degassing summit areas: the South-East crater (SEC), the Central Craters (Bocca Nuova and Voragine), and the North-East crater (NEC) (Fig. 1). Our study is based on passive emissions from the NEC, in recent times one of the most actively degassing vents on Etna (Aiuppa et al., 2008) and the site of recurrent eruptive activity in the last few decades (Allard et al., 2006).

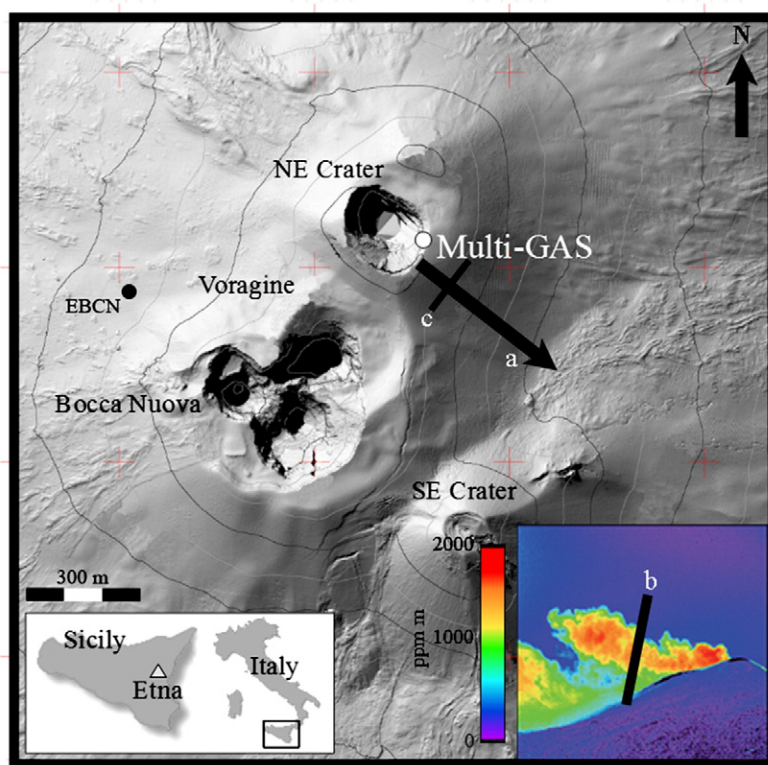
## 2. Methodology

The SO<sub>2</sub> emission rates were captured using two Apogee Alta U260 cameras, fitted with 16 bit 512 × 512 pixel Kodak KAF-0261E thermoelectrically cooled CCD array detectors. A Pentax B2528-UV lens of  $f = 25$  mm was attached to the front of each camera, providing  $\approx 24^\circ$  field of view. The lenses were fitted with filters of 10 nm FWHM (Asahi Bunko Inc.), one centred around 310 nm, where plume SO<sub>2</sub> absorbs incident UV radiation, and the other at 330 nm, where no such absorption occurs. Qualitative plume absorbances captured in the camera plume images were converted to column amounts via a calibration procedure involving four quartz cells containing known SO<sub>2</sub> column amounts: 100, 200, 1000, 2000 ppm m; SO<sub>2</sub> values within the plume were always within this range. The calibrations were performed at the time of measurement, by viewing clear sky adjacent to the plume, resulting in  $R^2$  values  $>0.99$  for the linear fitting. As the measurement conditions were favourable: e.g., the plume was transparent, the background sky was cloudless and the plume was  $<4$  km distant, additional DOAS based calibrations were not performed, as there is an excellent match between DOAS and cell based calibrations under such conditions (Lübcke et al., 2013). Under such circumstances we speculate that the measurement error was low, however, as radiative transfer has yet to become a routinely considered element of UV camera retrievals it is hard to provide an exact error budget in this case (e.g., Kern et al., 2009). For full details on all data capture, retrieval and calibration

procedures please see Kantzas et al. (2010). All of these protocols were executed using the Vulcamera code (Tamburello et al., 2011b).

The UV camera was located at the Pizzi Deneri observatory which provided a clear vantage point of the NEC plume, at a distance of  $\approx 2$  km (Fig. 1); the data were acquired between 08:45 and 09:45 GMT on the 12th of September 2012. Integrated column amount (ICA) values were determined by summing SO<sub>2</sub> concentrations over the plume profile, perpendicular to its transport vector (Fig. 1). The emission rates ( $\text{kg s}^{-1}$ ) were then found by multiplying ICAs by the plume transport speed, with the latter arising from cross-correlation analysis of the propagation of the plume across the field of view over a sequence of camera images (e.g. see McGonigle et al., 2005; Williams-Jones et al., 2006). The plume speed varied very little over the acquisition period ( $\approx 13.4 \text{ m s}^{-1}$  throughout). The camera capture rate ranged between 0.5 and 1 Hz depending upon incident light levels, hence linear interpolation was applied, where necessary, to produce a uniform 1 Hz SO<sub>2</sub> emission rates dataset.

The CO<sub>2</sub>/SO<sub>2</sub> degassing ratios of the NEC were measured with a field portable Multi-GAS unit (Aiuppa et al., 2005; Shinohara, 2005) located at  $\approx 100$  m downwind of the crater's vent, at a site chosen to avoid signal contamination from low-temperature fumarolic discharges (Shinohara et al., 2008). This unit extractively sampled the plume gases, providing CO<sub>2</sub> and SO<sub>2</sub> concentration readings at  $\approx 0.5$  Hz measurement frequency. The SO<sub>2</sub> concentrations were measured with an electrochemical sensor (City Technology, sensor type 3ST/F), of calibration range of 0–200 ppm, and the manufacturer quoted accuracy of  $\pm 2\%$ , repeatability of 1% and a resolution of 0.5 ppm v. The CO<sub>2</sub> concentrations were measured with an infrared sensor (Edinburgh Instruments, Gascard II), of 0–3000 ppm v range, and with an accuracy of  $\pm 2\%$  and a resolution of 0.8 ppm v. Prior to the campaign, the Multi-GAS sensors were calibrated in the laboratory using standard gas cylinders of concentrations within the sensor ranges (e.g., 10 and 100 ppm SO<sub>2</sub> and 3000 ppm CO<sub>2</sub>; all in nitrogen matrixes) and gas mixtures



**Fig. 1.** Map of the summit of Mount Etna showing the craters (NE—North East crater, Voragine, Bocca Nuova, SE—South East Crater), EBCN seismic station, the Multi-GAS location and (a) the plume direction; the left inset shows the volcano location in Sicily; the right inset shows the NE crater plume on the acquisition day as viewed with the UV camera from Pizzi Deneri, with the colour scale indicating ppm m column amounts of SO<sub>2</sub> over the image pixels; (b) shows the plume cross-section used to determine the Integrated Column Amount (ICA) within this inset; and (c), within the main image, the viewing vector corresponding to this profile with respect to the Multi-GAS location.

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