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**SO₂ emissions, plume heights and magmatic processes inferred from satellite data:
the 2015 Calbuco eruptions**

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Key points:

- Satellite data from GOME-2 and numerical modelling are used to investigate SO₂ emissions during the 2015 Calbuco eruptions
- Numerically retrieved quantities (SO₂ loading, plume heights and mass eruption rates) are used to investigate magmatic processes such as excess degassing
- Petrological analyses on Calbuco tephra samples validate the hypothesis inferred from space
- **Keywords**
SO₂ emissions; numerical modelling; excess degassing

Abstract

Quantifying time-series of sulfur dioxide (SO₂) emissions during explosive eruptions provides insight into volcanic processes, assists in volcanic hazard mitigation, and permits quantification of the climatic impact of major eruptions. While volcanic SO₂ is routinely detected from space during eruptions, the retrieval of plume injection height and SO₂ flux time-series remains challenging. Here we present a new numerical method based on forward- and backward-trajectory analysis which enable such time-series to be determined.

Using this method applied to GOME-2 satellite imagery we investigate the SO₂ emissions from two sub-Plinian eruptions of Calbuco, Chile, produced in April 2015. Our results show a mean injection height of 15 km for the two eruptions, with overshooting tops reaching 20 km. We calculate a total of 0.295 ± 0.045 Tg of SO₂ emitted, with 0.140 ± 0.033 Tg produced by the first eruption and 0.155 ± 0.031 Tg by the second one. Using standard models we convert plume heights to mass eruption rates (MER). Comparing gas flux and MER we discover quite different volcanic processes driving the two eruptions, with the first eruption producing an SO₂ flux three times higher than the second one, while they both had similar MERs. We propose that this difference reflects different exsolved volatile contents before the onset of the two eruptions, with the first eruption richer in pre-exsolved gas than the second one. This hypothesis is supported by melt inclusion measurements of sulfur concentrations in plagioclase phenocrysts and groundmass glass of tephra samples through electron microprobe analysis. Combining the satellite and petrological analysis, we propose that the overpressure caused by the pre-exsolved volatile phase (not only SO₂, but also probably H₂O and CO₂) may have triggered the eruption.

These results demonstrate that our new methodology produces constraints on SO₂ flux and plume height time-series permitting new insights into sub-surface processes using satellite SO₂ data.

1. Introduction

Understanding the manner and the abundance of sulfur degassing from active volcanoes during explosive eruptions is one key to unravelling eruptive dynamics (Oppenheimer et al., 2011, Wallace and Edmonds, 2011). At a volcanic vent, sulfur gases contribute 2-35 vol% of total gas emissions, with SO_2 and H_2S the dominant sulfur-bearing components, ranging between 1-25 vol% and 1-10 vol% respectively (Textor et al., 2004). Satellite-based instruments operating in the ultraviolet and infrared have detected and quantified volcanic sulfur gases in the atmosphere since 1978 (Carn et al., 2016). Nowadays, this is routinely done for SO_2 (Brenot et al., 2014), while few H_2S satellite retrievals have been performed so far (Clarisse et al., 2011). Satellite-based monitoring of volcanic SO_2 emissions is of value for poorly monitored volcanoes, which make up almost 95% of all volcanoes, but are also useful when well-monitored volcanoes erupt explosively, as local detection system can be saturated or blinded by ash.

Satellite images of volcanic SO_2 plumes contain a lot of information that can be extracted with the appropriate data analysis approach (McCormick et al., 2014; Hayer et al., 2016). The most immediate information is typically vertical column amounts of SO_2 , which can be readily used to determine a total SO_2 mass loading, and this is the most frequently used type of data provided in the literature. We highlight, however, that retrieved SO_2 column amounts are sensitive to plume height, which is not always well-constrained. Valuable time-series information on SO_2 injection height and SO_2 flux time-series are theoretically available, and these allow subtle observations and deductions on the volcanic processes driving eruptions, including magma degassing (Carn et al., 2008; Carn and Prata 2010; Campion 2014) and the role of pre-eruptive gas accumulation (Westrich and Gerlach, 1992). While a lot of work has been done on SO_2 satellite retrievals, a comprehensive, general methodology able to fully characterize both SO_2 flux and plume height

time-series has not been successfully created to date. This is mainly due to the difficulty in retrieving SO₂ vertical profiles for individual SO₂ column amount pixels in an image. All satellite-based SO₂ column amount calculations are dependent on both the measured SO₂ optical depth and the plume height, and so quantification of SO₂ amounts requires an accurate determination of plume height pixel by pixel in an image. Plume heights have been retrieved using infrared and ultraviolet spectra (Yang et al., 2010; Nowlan et al., 2011; Rix et al., 2012; Carboni et al., 2012; Clarisse et al., 2014; Carboni et al., 2016; Grainger et al., 2016) and from numerical models applied to satellite images (Hughes et al., 2012; Moxnes et al., 2014; Heng et al., 2016; Pardini et al., 2017).

SO₂ flux time-series can be calculated from satellite imagery using a variety of methods (a review is presented in Theys et al., (2013). Four methodologies have been applied: the box method (Lopez et al., 2013), the traverse method (Merucci et al., 2011), the delta method (Krueger et al., 1996) and inverse modelling (Eckhardt et al., 2008, Boichu et al., 2013). Depending on the input parameters (plume age at the measurement time, satellite sensor spatial resolution, number of satellite acquisitions in a day, etc...) and expected outcomes (flux time-series, plume height time-series), each method has strengths and weaknesses. The box method is suitable for a first flux evaluation, but it needs constant wind speed and direction together with an *a priori* estimation of plume height. The traverse method has been used to compare fluxes retrieved from satellite-based instruments with those from ground based measurements. This technique allows an almost real time estimate of SO₂ flux, but it needs constant wind direction and plume height as input data. The delta method is independent from wind speed and it produces an estimate of the SO₂ lifetime, however multiple satellite overpasses are needed. Finally, the inverse modelling allows us to compute fluxes at high temporal resolution even for

87 plume presenting a complex vertical profile. The main drawback of this technique is the
88 computational time.

89 In this work we present a trajectory model approach, which we call PlumeTraj, and use it to
90 investigate SO₂ emissions during explosive eruptions with the aim of exhaustively examining the
91 information which can be obtained from satellite imagery. With PlumeTraj we determine both
92 the plume height for each SO₂ pixel in the satellite image and the time at which the SO₂ in each
93 pixel was injected into the atmosphere from the eruption column. PlumeTraj integrates the
94 Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) (Stein et al., 2015)
95 with custom-built Python routines to create a semi-automated numerical procedure from which
96 injection height and flux time-series are quantified with relatively low computational costs (12
97 hours on a 48 node cluster).

98 PlumeTraj allows us to study both explosive and effusive eruptions, and, for each case study,
99 specific input parameters (such as volcanic location, type of eruption, eruption time) can be set
100 by the user. Our technique requires satellite and wind field datasets, which can be derived from a
101 variety of sources. Indeed, many satellite sensors can detect volcanic SO₂ atmospheric
102 abundance (Carn et al., 2016), and, theoretically, each satellite dataset can be used as input for
103 the model. The same can be done for the wind field data, which, however, must be written in a
104 format that HYSPLIT can read. The main advantage is the possibility to retrieve both SO₂ flux
105 and plume height. Moreover, mass eruption rates and masses of erupted solid material can be
106 derived from the plume height time-series by applying the well-known relationship between
107 plume height and mass eruption rate (Morton et al., 1956). However, we must consider
108 uncertainties and errors due the satellite retrievals of atmospheric SO₂ and the numerical analysis
109 of these data.

We applied our numerical method to GOME-2 satellite images of SO₂ plumes emitted by the two recent sub-Plinian eruptions occurred at Calbuco volcano, Chile, in April 2015. The eruptions have been classified as VEI 4 (Romero et al., 2016) and led to ozone depletion in Antarctica (Solomon et al., 2016, Ivy et al., 2017).

Our retrieved SO₂ flux time-series and masses of erupted material reveal differences between the two eruptions and allow us to infer the presence of a more abundant pre-exsolved gas phase for the first event. To validate and quantify the amount of excess SO₂, we perform microprobe analysis of melt inclusions in plagioclase phenocrysts and ground mass of erupted products. This allows us to compare our numerical results with the SO₂ loading derived from the “Petrological Method” (Devine et al., 1984), which uses information on the mass loading of each eruption and the volatile loss inferred from the difference in sulfur concentration between melt inclusions and groundmass.

The petrological analysis confirms the scenario inferred from space highlighting that the difference between the two eruptions is due to the presence of a higher amount of pre-exsolved SO₂ for the first event in compare with the second one.

2. Case study: the 22-23 April 2015 Calbuco eruptions

On the evening of 22 April 2015, Calbuco volcano started a new cycle of eruptive activity after 54 years of quiescence. Calbuco (41.33° S, 72.61° W) is an active stratovolcano located in the southern region of the Southern Volcanic Zone of the Andes, Chile. It has been volcanically active since the Late Pleistocene to the present, with the formation of 4 principle deposits. The last deposit has a “dome-cone” structure resulting from a series of recent major eruptions which occurred in 1912, 1961, 1971 and 1983-94 (Lopez-Escobar et al., 1992). The new eruptive cycle started on 22 April 2015 and lasted 9 days, until 30 April 2015. An initial sub-Plinian eruption

took place on the evening of 22 April (hereafter Eruption 1), and a second eruption occurred a few hours later in the morning of 23 April (hereafter Eruption 2).

Eruption 1 started suddenly at 20:54 UT. A volcanic column more than 15 km height rose from the main crater and tephra was dispersed in an East-Northeast direction. The overall duration of the event was 1.5 h. After Eruption 1 stopped, moderate seismic events in the form of volcanic tremor were recorded from 00:55 UT. At 04:00 UT, a new eruptive event (Eruption 2) occurred. The eruptive column reached more than 15 km in altitude and tephra was dispersed in a North, Northeast and East direction. At 10:30 UT the eruption was declared over (SERNAGEOMIN, 2015a, 2015b, 2015c).

The eruptions are classified as VEI 4 (Romero et al., 2016) and they produced columns reaching the stratosphere. The stratospheric injection by the volcanic cloud together with the latitude of Calbuco, produced an impact on ozone recovery in Antarctica causing an increase in hole size of 4.4 million km² (Solomon et al., 2106; Ivy et al., 2017). Moreover, extensive damage was caused to the Chilean economy, with agricultural and industrial resources close to Calbuco damaged by ash fall, and air traffic over Chile and Argentina disrupted for some hours (Romero et al., 2016). Considering both the tephra fall and PDC deposits, the deposit volume estimated by Castruccio et al., (2016) is 0.38 km³ assuming a deposit density of 1000 kg m⁻³ (0.15 km³ dense rock equivalent DRE), while Romero et al., (2016) report a tephra fall deposit volume of 0.28 km³ considering a deposit density of 997.3 kg m⁻³ (0.11-0.13 km³ DRE). These values are both within the 0.56±0.28 km³ volume calculated by Van Eaton et al., (2016), which presents a DRE of 0.18±0.09 km³ assuming a magma density of 2500 kg m⁻³.

3. PlumeTraj: Trajectory-based modelling of volcanic plume height and SO₂ flux

PlumeTraj is a development of an approach presented by Pardini et al., (2017). This new approach uses a two-step procedure based on a combination of forward and backward trajectories in order to better constrain plume height and thus SO₂ flux. We also modified the post-processing phase, changing the selection criteria for acceptable trajectories and adding an SO₂ flux calculation. Due to the general implementation of PlumeTraj, it can be easily applied to different volcanic systems to investigate SO₂ emissions during eruptive episodes or produced by quiescent degassing, using either single or multiple satellite images.

To run PlumeTraj we need one or more satellite images of a volcanic SO₂ plume and their associated numerical wind fields. For each pixel in which SO₂ is detected, PlumeTraj calculates three quantities. The first quantity, h , is the height at which the SO₂ is located at satellite measurement time instant (hereafter plume height). The second one, h_{vent} , is the height above volcanic vent at which SO₂ reaches the neutral buoyancy height and the prevailing atmospheric wind starts to disperse the gas into the atmosphere (hereafter injection height). The last one, t_{vent} , is the time when the SO₂ reaches the injection height (hereafter injection time). Knowing these three quantities and SO₂ column amount from satellite images, we can calculate the SO₂ mass loading of the plume and SO₂ flux time-series as it enters the atmosphere from the eruption column.

Plume parameters are computed by PlumeTraj by calculating trajectories backwards and, potentially, forwards in time. The trajectory calculation is performed by using HYSPLIT (Stein et al., 2015) with custom-designed routines written in the Python Programming Language. Below we describe the PlumeTraj procedure using three satellite images capturing a same plume

at different times. Forward and backward trajectories are run from the pixels of the satellite image for which heights and fluxes are computed.

As reported in Figure 1, the magenta pixels represent the plume as seen at day k , whilst the blue and yellow pixels are the same plume at days $k + 1$ and $k + 2$ respectively. For each pixel j of the day k image and for each potential plume height $h_j(i)$ in a given range, we calculate forward trajectories $traj_j^f(i)$, up to the time of acquisition of the day $k + 2$ image. In this example, from pixel j we run four forward trajectories initialized at four different heights ($traj_j^f(1)$, $traj_j^f(2)$, $traj_j^f(3)$ and $traj_j^f(4)$ in Figure 1). Among these trajectories, only those intersecting with the advected/dispersed plume at days $k + 1$ and $k + 2$ are considered (for example, in Figure 1 only $traj_j^f(1)$ and $traj_j^f(2)$ are acceptable). Then, starting only from $h_j(i)$ of each acceptable forward trajectory, we calculate backward trajectories ($traj_j^b(1)$, $traj_j^b(2)$ in Figure 1). We then select as acceptable trajectories only those approaching the volcanic vent location within the umbrella cloud distance and within the time interval of the eruption (for example, in Figure 1 only $traj_j^b(1)$ is acceptable). We adopt this two-step trajectory analysis to better constrain the height of the plume and show in supplementary materials additional simulations performed with different wind-fields.

Figure 1 Schematic representation of PlumeTraj numerical procedure. Magenta pixels are those associated with the *day k* satellite image, while blue and yellow pixels are those from the *day k+1* and *k+2* images. From pixel *j* (white star), trajectories $\text{traj}_j^f(1)$, $\text{traj}_j^f(2)$, $\text{traj}_j^f(3)$ and $\text{traj}_j^f(4)$ are run forward from different starting altitudes ($h_j(1)$, $h_j(2)$, $h_j(3)$ and $h_j(4)$). While $\text{traj}_j^f(1)$ and $\text{traj}_j^f(2)$ are consistent with the position of the plume at *day k+1* and *day k+2*, $\text{traj}_j^f(3)$ and $\text{traj}_j^f(4)$ are not, thus they are neglected. Starting again from pixel *j*, $\text{traj}_j^b(1)$ and $\text{traj}_j^b(2)$ are initialized from altitudes $h_j(1)$ and $h_j(2)$ and are run backward in time. Only $\text{traj}_j^b(1)$ is acceptable since it approaches the volcanic vent position at a distance less than $r(t)$.

An acceptable back trajectory is one which lies within the radius of the umbrella cloud, $r(t)$, during the eruption. Following Woods and Kienle, (1994), the radius of an umbrella cloud growing with time at the neutral buoyancy height can be expressed as:

$$r(t) = \left(\frac{3\lambda}{2\pi} N \cdot V \right)^{\frac{1}{3}} t^{\frac{2}{3}} \quad (1)$$

where λ is an empirical constant, N is the buoyancy frequency of the atmosphere and V is the volumetric flow rate at buoyancy height. We set λ equal to 0.8 and N equal to 0.017 s^{-1} for stratospheric strong plumes (Sparks et al., 1997). To evaluate the expansion of the umbrella

radius we set the mass eruption rate (MER) as input data. MER and V are related through the following relationship (Morton et al., 1956):

$$MER \approx \left(\frac{VN^{5/8}}{C\sqrt{k_e}} \right)^{\frac{4}{3}} \quad (2)$$

where k_e is an entrainment coefficient equal to 0.1 and C is a proportionality constant equal to $1 \cdot 10^4 \text{ m}^3 \text{ kg}^{-3/4} \text{ s}^{-7/8}$.

Using the two step PlumeTraj approach, for each pixel j forming the day k plume, and for each acceptable backward trajectory $traj_j^b(i)$ starting from the pixel j , we extract the three plume parameters $h_j(i)$, $h_{j_{vent}}(i)$ and $t_{j_{vent}}(i)$. The height $h_j(i)$ is the altitude of the starting point of the backward trajectory $traj_j^b(i)$ of the pixel j . Instead, $h_{j_{vent}}(i)$ and $t_{j_{vent}}(i)$ are respectively the height and the time at which each acceptable backward trajectory $traj_j^b(i)$ approaches the vent. From the plume parameters calculated by our numerical method, we compute, for each pixel j , the mean values ($\bar{h}_j$, $\bar{h}_{j_{vent}}$ and $\bar{t}_{j_{vent}}$) and standard deviations (σ_{j_h} , $\sigma_{j_{h_{vent}}}$ and $\sigma_{j_{t_{vent}}}$) as:

$$\bar{h} = \sum_{i=1}^N \frac{h_j(i)}{N}, \quad (3)$$

$$\bar{h}_{j_{vent}} = \sum_{i=1}^N \frac{h_{j_{vent}}(i)}{N}, \quad (4)$$

$$\bar{t}_{j_{vent}} = \sum_{i=1}^N \frac{t_{j_{vent}}(i)}{N}, \quad (5)$$

$$\sigma_{j_h} = \sqrt{\sum_{i=1}^N \frac{(h_j(i) - \bar{h})^2}{N}}, \quad (6)$$

$$\sigma_{j_{h_{vent}}} = \sqrt{\sum_{i=1}^N \frac{(h_{j_{vent}}(i) - \bar{h}_{j_{vent}})^2}{N}}, \quad (7)$$

$$\sigma_{j_{t_{vent}}} = \sqrt{\sum_{i=1}^N \frac{(t_{j_{vent}}(i) - \bar{t}_{j_{vent}})^2}{N}}, \quad (8)$$

where N is the number of backward trajectories $traj_j^b(i)$ that approach the vent, $h_j(i)$ is the altitude from which trajectories are initialized, while $t_{j_{vent}}(i)$ and $h_{j_{vent}}(i)$ are the time instant and the altitude of approach to the umbrella cloud.

Using these data, we compute the SO_2 loading in the volcanic plume and the mass of the tephra fall deposit. Finally, by associating pixels injection times ($t_{j_{vent}}$) with their SO_2 mass loading, which is calculated from the satellite SO_2 column amount, we calculate SO_2 flux time-series.

Depending on the input images, different combinations between forward and backward trajectories are possible. For example, if the day $k + 1$ image of Figure 1 is used as starting image of the simulation, backward trajectories only can be run using the day k image as a target. In the present work we show the results of two PlumeTraj simulations. Both simulations are performed to retrieve SO_2 injection height and flux time-series for the 2015 Calbuco eruptive event. The difference between the two simulations is the satellite image used as input data for the trajectories calculations.

3.1. Application of PlumeTraj to the Calbuco 2015 Eruptions

To investigate SO_2 plumes emitted during the Calbuco 2015 eruptions we use satellite data from the GOME-2 sensor (Rix et al., 2008). GOME-2 is an ultraviolet spectrometer (290-790 nm) aboard the polar-orbiting satellites MetOp-A (launched in 2006) and MetOp-B (launched in 2012) taking global measurements of atmospheric composition on daily basis. The two satellites operate in tandem with a temporal shift between acquisitions of 48 minutes and provide nadir-view scans with ground pixel size resolution equal to 40x40 km (swath of 960 km) in case of MetOp-A and 80x40 km (swath of 1920 km) in case of MetOp-B. Despite GOME-2 has a lower spatial resolution compare to the Ozone Monitoring Instrument (OMI), we used data from

GOME-2 since the OMI dataset covering the Calbuco eruptions is affected by the so called row-anomalies and thus the SO₂ plume is not fully seen by OMI. For GOME-2 retrievals, the Differential Optical Absorption Spectroscopy (DOAS) (Platt and Stutz, 2008) technique is applied to retrieve SO₂ vertical column amount by measuring the portion of the sunlight backscattered in the atmosphere. The overall error in SO₂ vertical column estimates is in the range 20-70% (Rix et al., 2012). This range of uncertainty accounts for both random and systematic errors. Random errors are mainly due to instrument noise and they are typically of 5-20%. The main contribution to systematic errors comes from the difficulty in assessing the plume height at measurement time and it is estimated to be in the range 10-60% (Rix et al., 2012). Plume height is a central parameter when converting SO₂ slant column density (i.e. the gas concentration along the entire light path) into vertical column density (i.e. the gas concentration right above the satellite footprint). When using an SO₂ retrieval made assuming the plume located at a certain height, errors up to 50% on vertical column amount can arise if the actual plume height is not the one used for the SO₂ retrieval. To deal with the missing information on plume height at measurement time, for GOME-2 retrievals, three different SO₂ estimates are provided for three hypothetical plume altitudes equal to 2.5 km, 6 km, 15 km. If we use the 2.5 km retrieval, this means that the maximum accuracy in SO₂ estimation is achieved if the actual plume is located at 2.5 km. In case of different plume height (higher than 2.5 km), SO₂ column amount for a single pixel can be overestimated up to 70-80% (Carn et al., 2013). On the contrary, when using the 6 and 15 km retrievals for a plume which is located at lower heights, the SO₂ column amount is underestimated and thus information on plume spatial distribution can be lost.

The first GOME-2 image of the Calbuco SO₂ plume was collected at ~13:00 on 23 April 2015, after the end of the two eruptive events. Then, plume advection/dispersion paths can be followed for about one month until they are diluted under the satellite detection limit (GOME-2 images can be displayed and datasets downloaded from the Support to Aviation Control Service (SACS) website <http://sacs.aeronomie.be/>). Due to GOME-2 MetOpA and B different pixel resolution, the original images are re-gridded into a new one presenting a spatial resolution of 30x30 km. For each day we use data from GOME-2 MetOpA, MetOpB or from a combination of the two sensors depending on the possibility to reconstruct the whole SO₂ cloud.

In Figure 2, we report the atmospheric SO₂ loading in Dobson Unit ($1 \text{ DU} = 2.7 \cdot 10^{16} \text{ molecules cm}^{-2}$) retrieved assuming the plume located at 2.5 km, 6 km and 15 km on the 23, 24 and 25 April 2015. To isolate volcanic plumes from background noise, we select pixels with a vertical column higher than a certain threshold calculated applied the Normalized Cloud-mass technique presented in Carn et al., (2008).

Figure 2 Upper row: Calbuco SO₂ plume as seen by GOME-2 on 23 April 2015. The three panels show the SO₂ vertical column retrieved assuming a plume altitude of 2.5 km (a), 6 km (b) and 15 km (c). In order to isolate the plume from the background noise, thresholds equal to 3.6 DU, 1.95 DU and 0.8 DU have been computed for the 2.5 km, 6 km and 15 km images respectively. Middle row: Calbuco SO₂ plume as seen by GOME-2 on 24 April 2015. The three panels show the SO₂ vertical column retrieved assuming a plume altitude of 2.5 km (d), 6 km (e) and 15 km (f). Lower row: Calbuco SO₂ plume as seen by GOME-2 on 25 April 2015.

To deal with the uncertainties and the errors associated with both the satellite retrieval and the numerical analysis, we perform two different simulations. In the first we initialize trajectories from the 23 April image and use the images collected on the 24 and 25 April as a target for the forward trajectory analysis (Simulation 1). SO₂ injection height and flux time-series are retrieved from the pixels of the 23 April image, which is the one capturing the plume a few hours after the

end of both eruptions. For the second simulation the trajectories are initialized from the 24 April image, while the 23 April is used as a target for the backward trajectories (Simulation 2). In this case the 24 April image is used for the retrieval of plume heights and SO₂ fluxes.

In both Simulation 1 and 2 the centre positions of the pixels forming the volcanic plume are used as starting points on the horizontal plane for the trajectory analysis.

We do not utilize a-priori assumptions on SO₂ plume altitude, and this information cannot be directly extrapolated from the satellite data, so we initialize trajectories exhaustively from 1 km to 30 km asl (upper stratosphere). Assuming an interval of 500 m between each starting height, we produce a total of 59 trajectories for each pixel. We set the centre position of each pixel as the starting point on the horizontal plane of each trajectory. The time at which the trajectories are initialized is coincident with the time at which the SO₂ vertical column was measured for each pixel.

For our test case, the numerical wind data comes from the global ECMWF atmospheric reanalysis ERA Interim dataset with a 0.75° grid.

After having performed the forward-backward trajectory selection for Simulation 1 and 2, we accept backward trajectories approaching Calbuco vent location (41.33° S, 72.61° W) using Eq.(1) with the additional constraint from eruption time interval. This means that we consider as acceptable only backward trajectories approaching the vent at a time instant which is consistent with the eruption time interval. This part of the numerical analysis is the same for the two approaches.

For the Calbuco eruptions, the eruption time is well constrained by visual-, satellite- and ground-based observations (SERNAGEOMIN 2015a, 2015b, 2015c; Van Eaton et al., 2016). Thus, for the study of Eruption 1, we use 21:00 and 22:30 UT (22 April) as beginning and end of the

eruption, while 04:00 and 10:00 UT (23 April) are the values referred to Eruption 2. The
 umbrella cloud radius is evaluated using the mass eruption rate (*MER*) for both Eruption 1 and
 Eruption 2. Since we do not have a precise estimation of mass eruption rates, we perform a
 sensitivity analysis on it and we investigate the range $0.3 \cdot 10^7 \text{ kg s}^{-1} - 2.7 \cdot 10^7 \text{ kg s}^{-1}$. These values
 are chosen accordingly to the minimum and maximum *MER* calculated for the 2015 Calbuco
 eruptions by previous works (Romero et al., 2016, Castruccio et al., 2016, Van Eaton et al.,
 2016). The sensitivity analysis is performed using the Design and Analysis toolKit for
 Optimization and Terascale (DAKOTA) (Adams et al., 2009), selecting a Latin Hypercube
 approach on a total number of 15 samples (i.e. 15 different values of *MER*). Our numerical
 results show that using the calculated umbrella cloud radius as a test for acceptable back
 trajectories works well for Eruption 2 but not for Eruption 1. We believe that the short duration
 of Eruption 1 and high gas content (see below), together with numerical uncertainties, meant that
 we need to investigate a wider range of potential radii than $r(t)$ to cover all pixels where SO_2
 was detected. For Eruption 1, we use an approaching radius varying from 0 to 500 km, whilst
 for Eruption 2 we calculate $r(t)$ using Eq. (1). In this case, the radius of the umbrella cloud
 ranges between 0 (beginning of the eruption) and 200 km (end of the eruption) for *MER* equal to
 $0.3 \cdot 10^7 \text{ kg s}^{-1}$ and between 0 and 360 km for *MER* equal $2.7 \cdot 10^7 \text{ kg s}^{-1}$. The sensitivity analysis
 performed on *MER* produces 15 sets of acceptable trajectories for each pixel (one set for each
MER). For a given *MER*, the number of the acceptable trajectories can vary from 0 (i.e. no
 acceptable trajectories starting from the considered pixel) to 59 (i.e. all the trajectories starting
 from the considered pixel are acceptable). Depending on the satellite image, from 50.000 to
 100.000 trajectories are computed during a single PlumeTraj simulation and about 15.000
 trajectories are then selected as acceptable (i.e. the 20% of the initial number).

4. Numerical Results

Using the previously described technique, we calculate the plume height, the injection height and the injection time for each pixel of the computational domain where SO_2 is detected. Following the results are presented for the two simulations. For the first simulation (Simulation 1), Figures 3 and 4 report both the mean values ($\bar{h}$, $\bar{h}_{vent}$ and $\bar{t}_{vent}$) and standard deviations (σ_h , $\sigma_{h_{vent}}$ and $\sigma_{t_{vent}}$) of plume parameters calculated for each pixel. Figure 3 shows the SO_2 cloud emitted from Eruption 1, whilst in Figure 4 we plot the one emitted from Eruption 2. We do not separate a-priori the plume of Eruption 1 from that of Eruption 2, but it is the model that, according to the approaching time and radius, distinguishes the two plumes. Figures 3 (a), (d) and 4 (a), (d) show respectively $\bar{h}$ and σ_h computed for each pixel of the computational domain. Similarly, Figures 3 (b), (e) and 4 (b), (e) report $\bar{h}_{vent}$ and $\sigma_{h_{vent}}$ respectively, whereas Figures 3 (c), (f) and 4 (c), (f) illustrate $\bar{t}_{vent}$ and $\sigma_{t_{vent}}$.

As we can see from Figure 3 and 4, the whole SO_2 plume is split into two clouds, both transported in the same direction (North North-East). The SO_2 injected into the atmosphere at the beginning of the eruptive phases travelled furthest from the vent, while pixels closer to vent location contain SO_2 emitted at the end of the two eruptions.

Due to low vertical velocity at stratospheric heights, $\bar{h}$ and $\bar{h}_{vent}$ are almost coincident (Figure 3 (a), (b) and 4 (a), (b)) and similar result is obtained for $\sigma_{h_{vent}}$ and σ_h (Figure 3 (d), (e) and 4 (d), (e)). For Eruption 1, a mean plume height of 15.7 ± 0.4 km both at vent and at satellite overpass is computed, with peaks of 21 km. The mean plume height computed for Eruption 2 is 15 ± 1.2 km, with some pixels presenting a maximum height of 20 km. Finally, uncertainties on injection time

365 ($\sigma_{t_{vent}}$) range between 0 and 40 min for Eruption 1 and between 0 and 100 min for Eruption 2
 366 with mean values of 12 min and 30 min respectively.

367
 368 **Figure 3** Results PlumeTraj Simulation 1 considering trajectories approaching the vent from 21:00 to 22:30 on 22
 369 April 2015 (Eruption 1). In panels (a), (b), and (c) mean plume height ($\bar{h}$), injection height ($\bar{h}_{vent}$) and injection
 370 time ($\bar{t}_{vent}$) are shown, whereas panels (d), (e) and (f) show the relative standard deviations (σ_h , $\sigma_{h_{vent}}$ and $\sigma_{t_{vent}}$).

Figure 4 Results PlumeTraj Simulation 1 considering trajectories approaching the vent from 04:00 to 10:00 on 23 April 2015 (Eruption 2). In panels (a), (b), and (c) mean plume height ($\bar{h}$), injection height ($\bar{h}_{vent}$) and injection time ($\bar{t}_{vent}$) are shown, whereas panels (d), (e) and (f) show the relative standard deviations (σ_h , $\sigma_{h_{vent}}$ and $\sigma_{t_{vent}}$).

Figure 5 and 6 show the results of the simulation performed using the 24 April image as input data (Simulation 2). We compute a mean height of 17.5 ± 0.2 km and 15 ± 0.5 km for the plume emitted during Eruption 1 and Eruption 2 respectively (both at vent and satellite overpass), with top plume height of 24 km. The uncertainties on the injection time range from 0 to 45 min for Eruption 1 and from 0 to 145 min for Eruption 2, with mean values of 6 and 60 minutes for the two eruptions.

Figure 5 Results PlumeTraj Simulation 2 considering trajectories approaching the vent from 21:00 to 22:30 on 22 April 2015 (Eruption 1). In panels (a), (b), and (c) mean plume height ($\bar{h}$), injection height ($\bar{h}_{vent}$) and injection time ($\bar{t}_{vent}$) are shown, whereas panels (d), (e) and (f) show the relative standard deviations (σ_h , $\sigma_{h_{vent}}$ and $\sigma_{t_{vent}}$).

Figure 6 Results PlumeTraj Simulation 2 considering trajectories approaching the vent from 04:00 to 10:00 on 23 April 2015 (Eruption 2). In panels (a), (b), and (c) mean plume height ($\bar{h}$), injection height ($\bar{h}_{vent}$) and injection time ($\bar{t}_{vent}$) are shown, whereas panels (d), (e) and (f) show the relative standard deviations (σ_h , $\sigma_{h_{vent}}$ and $\sigma_{t_{vent}}$).

To test the consistency of our results we use our injection height time-series to initialize a dispersal simulation using the HYSPLIT dispersion module. Puffs of gas are released from the Calbuco vent location at each retrieved $\bar{h}_{vent}$ and $\bar{t}_{vent}$ from an area which is consistent with the growth of the umbrella radius at that height and time during the eruption. The position of the plume is captured in agreement with the satellite measurement on 24, 25, 26, 27 and 28 April 2015 as shown in Figure 7. Panels from (a) to (f) show GOME-2 images of the Calbuco plume from 23 to 28 April 2015, while in panels from (g) to (n) our dispersal simulation is presented. A good match between the plume as captured by GOME-2 and as retrieved from our simulation can be observed especially for the part of the cloud located at the highest heights (from 15 to 25 km) where the bulk of the SO_2 appears to be located.

Figure 7 Panels from (a) to (f) show the Calbuco SO₂ plume as measured by GOME-2 from 23 to 28 April 2015. Panels from (g) to (n) present the results of the dispersal simulation performed using the retrieved injection time and height as input parameters. The altitudes at which the plume is located are shown with different colours from 0 to 25 km.

Plume height retrievals from PlumeTraj are consistent with those derived from analysis of both the tephra deposit and remote sensing techniques. Following the method of maximum clast diameters (Carey and Sparks, 1986), Romero et al., (2016) computed maximum column heights of 15.4 ± 3.08 km during Eruption 1 and the first phase of Eruption 2, while a decrease during the last phase of Eruption 2 emerges with heights of 12-13 km. Similar values are reported by Castruccio et al., (2016) with the only difference of proposing an increase in column height at the end the Eruption 2. These values are also in agreement with that presented by Van Eaton et al., (2016) considering the growth of the umbrella cloud (14.5-15.5 km for Eruption 1 and 16.9-17.3 km for Eruption 2). The main difference we notice from these deposit estimates of plume height compared with those performed by our numerical simulation is the absence of heights higher than 20 km. However, Vidal et al., (2017) show, using a dual polarized weather radar, the

main column located between 7 and 15 km for Eruption 2, with a maximum value of 23 km asl, in agreement with our estimations for plume heights. Finally, using spaceborne observations in the microwave, thermal infrared and visible wavelength Marzano et al., (2018) show a plume height between 15 km and 20 km with a maximum value of 21 km asl.

4.1 Masses estimation from numerical results and SO₂ flux time-series

Our retrieval of plume height time-series opens the possibility of quantifying mass eruption rate time-series, and to compare these data with field data. For the Calbuco eruptions, separation of volcanic ash and SO₂ gas has not been observed, so retrieved SO₂ injection heights are representative of column height evolution during the eruptions. Column height is primary controlled by the thermal buoyancy of the erupted material, which is a function of the mass flux supplied during the eruption (Sparks 1997). We use our mean injection height time-series to calculate a mean mass eruption rate (*MER*) and we use it to evaluate the mass of erupted solid material (*M*). From Simulation 1, *MER* and *M* are equal to $1.9 \pm 0.55 \cdot 10^7 \text{ kg s}^{-1}$ and $10.6 \pm 3 \cdot 10^4 \text{ kt}$ for Eruption 1 and $1.73 \pm 0.34 \cdot 10^7 \text{ kg s}^{-1}$ and $37.4 \pm 7 \cdot 10^4 \text{ kt}$ for Eruption 2. From Simulation 2, they are equal to $3.05 \pm 0.21 \cdot 10^7 \text{ kg s}^{-1}$ and $16.4 \pm 1.1 \cdot 10^4 \text{ kt}$ for Eruption 1 and $1.51 \pm 0.42 \cdot 10^7 \text{ kg s}^{-1}$ and $32.6 \pm 9.1 \cdot 10^4 \text{ kt}$ for Eruption 2. From these results, we compute a mean *MER* of $2.51 \pm 0.60 \cdot 10^7 \text{ kg s}^{-1}$ for Eruption 1 and of $1.62 \pm 0.54 \cdot 10^7 \text{ kg s}^{-1}$ for Eruption 2 and we infer $13.5 \pm 3.2 \cdot 10^4 \text{ kt}$ of solid material emitted during Eruption 1 and $35.0 \pm 11.7 \cdot 10^4 \text{ kt}$ during Eruption 2.

These values are in good agreement with those from Castruccio et al., (2016) which report $8 \cdot 10^4 \text{ kt}$ for Eruption 1 and $32 \cdot 10^4 \text{ kt}$ for Eruption 2. Our satellite-based interpretation seems to confirm the eruptive scenario proposed by Castruccio et al., (2016) which assign the first of the four

layers of the deposit to the first eruption and the other three to the second one. Differently, Romero et al., (2016) assign the first two layers to Eruption 1 and the other two to Eruption 2. Despite this, the two authors agree on the general stratigraphy.

Assuming a magma density of $\sim 2500 \text{ kg m}^{-3}$, we compute a deposit dense rock equivalent (DRE) of $0.198 \pm 0.050 \text{ km}^3$, with $0.055 \pm 0.013 \text{ km}^3$ resulting from Eruption 1 and $0.143 \pm 0.048 \text{ km}^3$ from Eruption 2.

Finally, we use our retrieved plume heights to correct the SO_2 vertical columns as computed from space. Indeed, SO_2 vertical columns retrieved from satellite data depend on several factors, such as plume height, SO_2 lifetime and satellite sensors signal saturation. In our test case, the effect related to SO_2 lifetime can be neglected. Indeed, lifetime of volcanic SO_2 injected into the stratosphere depends primarily on injection altitude and can vary from 8-9 days for 11 km height plumes (Krotkov et al., 2010) to 25 days for higher injection altitudes (Guo et al., 2004). For the Calbuco SO_2 cloud an e-folding time of 11 days has been computed by Bègue et al., (2017).

SO_2 retrievals from GOME-2 report vertical columns for each pixel at 3 hypothetical plume heights of 2.5 km, 6 km, 15 km (Figure 2(a)-(c)). We interpolate SO_2 column amounts between these three heights and use our calculated mean SO_2 height ($\bar{h}$) to correct the column amount. From Simulation 1, we compute 0.085 Tg and 0.090 Tg of SO_2 released during Eruption 1 and Eruption 2 respectively. From Simulation 2 we evaluate 0.195 Tg of SO_2 emitted during Eruption 1 and 0.219 Tg during Eruption 2. We compute a total SO_2 mass loading of $0.295 \pm 0.045 \text{ Tg}$, with $0.140 \pm 0.033 \text{ Tg}$ produced by Eruption 1 and $0.155 \pm 0.031 \text{ Tg}$ by Eruption 2. A major source of uncertainty for this result is represented by the possible underestimation of the SO_2 loading computed from the GOME-2 data. Signal saturation due to high SO_2 column amounts and presence of volcanic ash can affect the satellite retrieval. Thus,

our ~ 0.3 Tg of SO_2 emitted can be seen as a minimum value. However, Bègue et al., (2017) show that the atmospheric SO_2 loading computed from the IASI data varies from 0.1 Tg on 23 April 2015 to a maximum amount of 0.4 Tg on 25 April, in agreement with our estimate. Finally, by associating the injection time at the vent (t_{vent}) for each pixel SO_2 mass loading, we calculate SO_2 flux time-series, Figure 8.

Simulation 1

Simulation 2

Simulation 1&2

Figure 8 Upper row: SO₂ fluxes and injection height time-series resulting from Simulation 1. Middle row: SO₂ fluxes and injection height time-series resulting from Simulation 2. Lower row: SO₂ fluxes injection height time-series resulting from the combination of the two simulations.

The similar amount of SO₂ released during the two eruptions and the different duration of the events (1.5 h and 6 h) are reflected in the average SO₂ fluxes. Eruption 1 produced an intense gas emission with mean flux of 2200 ± 520 kt day⁻¹, while a smoother gas released can be observed for Eruption 2 together with a lower mean flux of 620 ± 120 kt day⁻¹. Comparing SO₂ flux and injection height time-series, two different volcanic processes driving Eruption 1 and 2 emerge. This is reflected in the similar total amount of SO₂ (~150 kt) released in different time scale (1.5 h and 6 h) but in a similar range of altitudes (14-18 km).

5. Estimate of SO₂ in Magma: method and results

To quantify the atmospheric SO₂ yield from the Calbuco eruptions, we adopt the well-established petrological method (Devine et al., 1984). This consists of measuring the sulfur concentration in glassy melt inclusions, which represent the undegassed melt, and in matrix glasses, which instead represent the degassed melt. The mass of sulfur released is then calculated by the difference of the two concentrations multiplied by the mass of erupted material. A correction considering the mass fraction of syn- or post-eruptive crystals can also be considered. To compare our satellite estimates of SO₂ mass loadings with elemental sulfur concentrations, it is useful to convert petrological estimates of sulfur mass loadings into SO₂ mass loadings. This is readily achieved by multiplying by two, following the molecular weight of SO₂ (64 g mol⁻¹) and atomic weight of

sulfur (32 g mol^{-1}). Thus, according to the petrological method, the SO_2 loading released into the atmosphere during an individual eruption ($m(\text{SO}_2)_{\text{PETR}}$) can be expressed as:

$$m(\text{SO}_2)_{\text{PETR}} = M \cdot (S_{\text{MI}} - S_{\text{MG}}) \cdot \frac{MW(\text{SO}_2)}{MW(\text{S})} \cdot (1 - CF), \quad (9)$$

where M is the mass of erupted material, S_{MI} and S_{MG} are the sulfur concentrations measured in melt inclusions and matrix glass, $MW(\text{SO}_2)$ and $MW(\text{S})$ are the molecular weights of SO_2 and S and CF is a coefficient accounting for the volume of syn- or post-eruptive crystals in the melt.

This method has been largely applied often in conjunction with satellite estimates of atmospheric SO_2 yield to investigate sulfur degassing mechanisms (Sharma et al., 2004; Sigmarsson et al., 2013). Depending on magma type, volcanic setting and eruption style, the atmospheric SO_2 yield retrieved from space can exceed the one resulting from the Petrological Method by up to one order of magnitude (Wallace 2001, Danyushevsky et al., 2002). For explosive eruptions, this behavior, which is known as “excess SO_2 ”, has been explained with the presence of a pre-eruptive exsolved SO_2 gas phase present at depth before the beginning of the eruption. From these analyses we can examine if excess SO_2 was produced during the Calbuco eruptions.

The tephra deposit formed by the Calbuco eruptions is composed by four layers named A, B, C and D. According to Castruccio et al., (2016), we consider scoriae of the first layer (layer A) as produced by Eruption 1 and those from the other three layers (layers B, C and D) from Eruption 2. Sulfur concentrations in the MIs of Eruption 1 (scoriae of layer A) vary from 240 to 520 ppm (see Table S1 in the supplementary material), and sulfur contents in the MIs of Eruption 2 (scoriae of layers B, C and D) range between 270 and 590 ppm. Low values of sulfur are measured in the matrix glasses of the scoriae erupted from both eruptions, ranging between 30 and 150 ppm (these values are close to the detection limit of the electron microprobe). From these analyses, the amount of sulfur dissolved in the melt is equal to 351 ± 98 ppm for Eruption 1

and 412 ± 101 ppm for Eruption 2. For the residual melt (matrix glass) we evaluate 93 ± 6 ppm for Eruption 1 and 92 ± 40 ppm for Eruption 2.

In order to evaluate the atmospheric SO_2 yield as shown in Eq. (9), we consider CF equal to 0.5 (i.e. we account for a 50 vol.% of crystals in the melt, from personal communication with Fabio Arzilli). For Eruption 1 M , S_{MI} and S_{gm} are equal to $13.5 \pm 3.2 \cdot 10^4$ kt, 351 ± 98 ppm and 93 ± 6 ppm, while for Eruption 2 they are equal $35.0 \pm 11.7 \cdot 10^4$ kt, 412 ± 101 ppm and 92 ± 40 ppm. From Eq.(9), the SO_2 yield emitted during Eruption 1 is 35 ± 16 kt, while the one emitted during Eruption 2 is 112 ± 53 kt.

6. Discussion

Excess SO_2 has been invoked to explain a large body of evidence where satellite detection of volcanic SO_2 plumes demonstrated that the amount of SO_2 released into the atmosphere during volcanic eruptions, both explosive and effusive, can exceed that resulting from syneruptive volatile exsolution (Devine et al., 1984).

The process which produces such excess SO_2 degassing depends on tectonic setting, magma type, magma evolution and eruption style and it appears to be particularly characteristic of explosive eruptions of intermediate and silicic magma in subduction zone settings (Andres et al., 1991), such as Calbuco. For explosive eruptions, excess degassing is caused by the presence of pre-eruptive exsolved bubbles accumulated at the top of the magma chamber where the erupted magma is also located. Pre-eruptive bubbles can be supplied to erupted magma by the intrusion of bubble-bearing magma into a magma chamber, by crystallization of part of the magma and subsequent bubble formation or by the presence of a sulfur-rich basaltic magma located beneath the silicic one (Shinohara, 2008). Once pre-eruptive bubbles are formed they migrate at the top

of the chamber due to buoyancy forces and form a gas rich foam which, once erupted, produces the observed excess degassing. We highlight that excess SO_2 degassing is likely to be associated with excess degassing of other volcanic gas species, such as the more abundant H_2O and CO_2 (Edmonds et al., 2010), and that this may represent a large but unconstrained contribution to the global geological carbon cycle.

Our PlumeTraj results show that the SO_2 flux produced during Eruption 1 was three times higher than that of Eruption 2 (2200 ktday^{-1} and 620 ktday^{-1} respectively), indicating a much more gas-rich magma in the first eruption. Using the PlumeTraj mass eruption rates we can infer the original SO_2 contents of magmas required to produce the observations: 0.10 wt% for Eruption 1 and 0.04 wt% for Eruption 2. Such a contrast in original SO_2 contents of magmas erupted so close in time suggests that the most likely explanation is not different dissolved volatile contents, but a larger role for magma chamber vapour segregation contributing to Eruption 1 than Eruption 2. We highlight that this inference on the subsurface process is produced purely through application of PlumeTraj to satellite imagery.

In order to quantify the pre-accumulated gas phase in Calbuco's 2015 eruptions, we compare the SO_2 yield derived from our numerical satellite-based technique, with the one resulting from petrological estimates, and we calculate the excess SO_2 as the difference between the mass of SO_2 inferred from space and the one inferred from the petrological analysis. For Eruption 1, we find $105 \pm 36 \text{ kt}$ of excess SO_2 , whereas for Eruption 2 $43 \pm 62 \text{ kt}$, see Table 1.

	M	S_{MI}	S_{gm}	$m(\text{SO}_2)_{PETR}$	$m(\text{SO}_2)_{SAT}$	$m(\text{SO}_2)_{ex}$
E1	$13.5 \pm 3.2 \cdot 10^4 \text{ kt}$	$351 \pm 98 \text{ ppm}$	$93 \pm 6 \text{ ppm}$	$35 \pm 16 \text{ kt}$	$140 \pm 33 \text{ kt}$	$105 \pm 36 \text{ kt}$

E2   35.0±11.7·10⁴kt   412±101ppm   92±40 ppm   112±53 kt   155±31 kt   43±62 kt

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M , mass of erupted solid material as estimated from our numerical method.

S_{MI} , S_{gm} , concentration of sulfur in melt inclusions and glassy matrix respectively.

$m(SO_2)_{PETR} = M \cdot (S_{MI} - S_{gm}) \cdot \frac{MW(SO_2)}{MW(S)} \cdot (1 - CF)$, where M is the mass of erupted material, $MW(SO_2)$ and

$MW(S)$ are the molar weights of SO_2 and S equal to 64 g mol⁻¹ and 32 g mol⁻¹ and CF is a coefficient accounting for 50 vol.% of crystals.

$m(SO_2)_{SAT}$, atmospheric sulfur yield as retrieved from satellite data.

$m(SO_2)_{ex} = m(SO_2)_{SAT} - m(SO_2)_{PETR}$, mass of pre-eruptive exsolved SO_2 .

Table 1 Calculation of SO_2 budget from Calbuco eruptions

This demonstrates that 75±26% of the SO_2 emitted during Eruption 1 and 28±40% of SO_2 emitted during Eruption 2 were already present as part of the pre-eruptive exsolved gas phase. From the petrological results, the weight percent of SO_2 lost from the erupted magma is ~0.03 wt% for both the eruptions. Comparing this value with those calculated from PlumeTraj (0.10 wt% for Eruption 1 and 0.04 wt% for Eruption 2), we find an excess of 0.07 wt% of SO_2 for Eruption 1 and 0.01 wt% for Eruption 2. Within the uncertainties, we conclude that excess SO_2 is present for Eruption 1, while Eruption 2 ranges between no excess SO_2 and an amount of already degassed SO_2 which is in, any case, lower in compare with Eruption 1.

To consolidate this result, additional PlumeTraj simulations have been performed using different wind-fields (see the Supplementary Material). We found an overall agreement between all the analyses, and the presence of excess SO_2 appear to be confirmed. This result is not affected by the possible underestimation of the SO_2 loading as computed from the GOME-2 dataset. In fact, a higher SO_2 loading computed from space would result in a higher weight percent of SO_2 computed by PlumeTraj, while the outcomes of the petrological analysis would be the same.

Thus, for atmospheric SO₂ yield higher than that retrieved by GOME-2 (~0.3 Tg) the SO₂ excess would be confirmed for the two eruptions.

From the combined satellite and petrological analysis, we conclude that Eruption 1 was triggered by the overpressure caused by the accumulation of pre-eruptive bubbles at the top of the magma chamber. This resulted in a gas-rich impulsive eruption. The subsequent pressure release allowed the exsolution of new bubbles which, together with the pre-eruptive ones (less abundant than those triggering Eruption 1), powered Eruption 2 during which a larger amount of magma was released in compare to Eruption 1.

7. Conclusions

We have developed the PlumeTraj technique to retrieve SO₂ flux and eruption plume height time-series of explosive eruptions. PlumeTraj combines satellite imagery of volcanic SO₂ plumes with independent observations of the timing of the eruption and forward and backward trajectory simulations performed through the HYSPLIT software, can be generally applied, and used to investigate SO₂ emissions during any type of volcanic eruption. The algorithm is computationally efficient and can be run in an automated manner in <12 hours. Retrieved plume heights are used to correct the assumption that the whole plume is located at the same hypothetical altitude, thus producing corrected SO₂ vertical columns.

Here, we quantified SO₂ emissions from the recent April 2015 Calbuco eruptions using imagery from the GOME-2 satellite sensor. It is worth to notice that in 2017 the TROPOspheric Monitoring Instrument (TROPOMI) has been launched aboard the Copernicus Sentinel-5 Precursor (S-5P) platform. TROPOMI will open the possibilities of SO₂ retrievals at higher spatial resolution than before (Theys et al., 2017) and the investigation of volcanic processes

through satellite data can be further improved with the use of more accurate SO₂ retrievals coming from this instrument.

In the present paper, we retrieved SO₂ injection height and flux time-series together with masses of erupted material and we used them to unravel triggering mechanisms and volcanic processes of the Calbuco eruptions. Comparing our results in terms of atmospheric SO₂ yield and masses of solid material released during the eruptions, we inferred the presence of different exsolved volatile phase at depth between the two eruptions. Electron microprobe analyses performed on Calbuco tephra samples confirmed our conclusions, validating our hypothesis made just from our numerical technique. One of the main results of this study is the evidence that exsolved SO₂ was present before the onset of the eruptions, highlighting that other volatiles (H₂O, CO₂ and Cl) were already exsolved at the same pre-eruptive conditions (Wallace 2005). This lead us to conclude that the overpressure due to pre-eruptive exsolved volatiles (not only SO₂, but probably also H₂O and CO₂) may have played a role in the triggering mechanisms of both the sub-Plinian eruptions.

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