

## Reply to Anonymous Reviewer #2

We highlight the Reviewer comments in red (R) and our answers in black (A).

### General comments:

**R1.** The method consists first in evaluating, from the known location of the volcanic plume at the time of satellite acquisition and by using HYSPLIT backtrajectory simulations, three variables associated to each pixel of the satellite image : 1- the SO<sub>2</sub> altitude at a given pixel (h), 2- the altitude of SO<sub>2</sub> injection above the vent (h vent ), 3- the associated time of SO<sub>2</sub> injection above the vent (t vent ). To do so, the authors test a broad range of SO<sub>2</sub> altitudes (between 2 and 30 km) at a given pixel and select the pixel altitudes which correspond to backward trajectories that pass over the Calbuco vent (or within a certain radius around the volcano). The set of solutions found for these 3 parameters (h, h vent , t vent ) may be refined in a second step. This refinement consists in subsetting the range of pixel altitudes selected in the first step, to keep only those which initialise HYSPLIT forward trajectories that point toward pixels where the SO<sub>2</sub> plume is detected in a second satellite image acquired 24 h later.

**A1.** We want to point out that our trajectory procedure is not exactly performed as the reviewer describes. We first operate a forward trajectory analysis and we select the altitudes initializing trajectories consistent with the plume position as captured by GOME-2 on April 24. The backward trajectory analysis is the second step. This is done in order to reduce uncertainties due to constant wind field. The plume position is clearly captured from space on April 24, while the determination of a distance of closest approach to the vent is more uncertain and based on mass eruption rate estimates. Thus, the forward trajectory analysis is a reliable way to discharge altitudes not consistent with the plume position. We then operate the backward trajectory analysis and we consider uncertainties on distance of closest approach by doing a sensitivity analysis varying the mass eruption rate, which is the main parameter controlling the radius of the umbrella cloud and thus the distance of closest approach.

However, such a problem has multiple solutions, especially because a strong interdependency often exists between the time and altitude of injection when trying to explain the location of a gas parcel detected in a satellite image.

We completely agree with the reviewer. This is why we operate a two-step procedure and we use the eruption time interval as constrain on the time for the trajectories to be selected. For the Calbuco eruptions, the eruption time is well constrained and it can be found in the numerous papers published on it. All these papers refer to:

SERNAGEOMIN, 2015a. Reporte Especial de Actividad Volcánica (REAV) Región de los Lagos. (REAV) Año 2015 Abril 22 (20:45 HL).

SERNAGEOMIN, 2015b. Reporte Especial de Actividad Volcánica (REAV) Región de los Lagos. Año 2015 Abril 22 (22:30 HL).

SERNAGEOMIN, 2015c. Reporte Especial de Actividad Volcánica (REAV) Región de los Lagos. Año 2015 Abril 23 (10:30 HL)

Here, the authors present a unique solution for the set of variables (h, h vent , t vent ) which they compute from the nmean value (h , h vent , t vent ) ) considering all the possible solutions found for the altitude of the pixel of interest (Fig. 3 and Fig.4) – by the way, the formula is provided in caption of Fig 3, but should be in the text. The validity of this presented solution is highly questionable.

All the equations in caption 3 have been added in the main text as:

“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 ( $\sigma_{j_h}$ ,  $\sigma_{j_{h_{vent}}}$  and  $\sigma_{j_{t_{vent}}}$ ) as:

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

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

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

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

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

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

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 at vent position. “

The strong interdependency between time and altitude of injection is clearly shown by the results of the authors. For example, the comparison between results in Fig. 3 and Fig. 4 indicates that for the exact same pixels in the zone indicated by black arrows in the figure 1 of this review (figure 1 of this review includes subplots taken from Fig. 3 and Fig. 4), an altitude of 14-16 km (associated eruption time retrieved: 23/04/15 at 04 :00 AM) is retrieved on Fig. 4, whereas an altitude of 7-10 km (associated eruption time retrieved: 22/04/15 at 22 :30 AM) is retrieved on Fig. 3.

This example shows the extreme instability of the results which are presented: given the range of time intervals which is assumed for the time period of injection of SO<sub>2</sub>, very different results are obtained for the exact same pixels.

We agree with the reviewer in considering the results extremely dependent on the time interval under investigation. This is why we use the eruption time as constraint for the trajectories to be chosen and we also try to decrease the uncertainties on plume parameters using the two step procedure. From our results, the plume produced by Eruption 1 appears to be highly stratified with maximum heights of 20 km together with a layer located at lower altitudes (below 10 km). Looking at the pictures taken during the eruptions [Romero et al., 2016], for Eruption 1, a layer located at lower heights appears to be present.

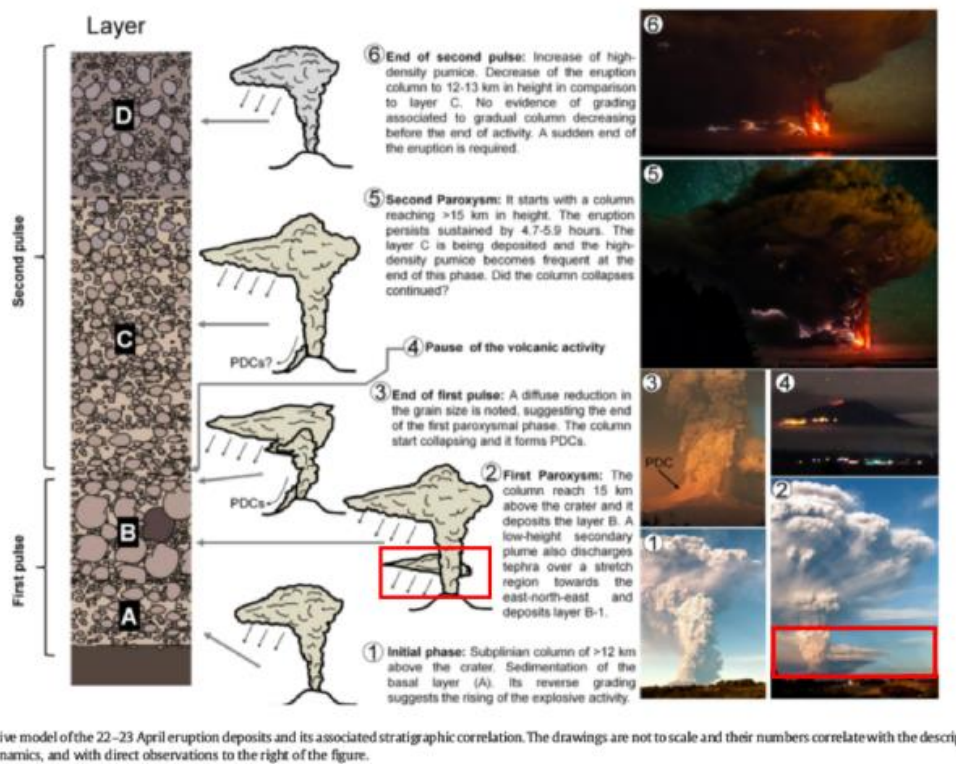


Figure 1: modified from Romero et al., [2016]. In the red box the lower altitude plume can be seen.

We believe this layer is the one than can be seen in Figure 3. Since it is located at lower altitudes, it moved slower than the stratospheric cloud produced later by Eruption 2. Thus, the stratospheric plume emitted during Eruption 2 has been able to reach it. This is why, for a single pixel, two different solutions are presented, one associated to Eruption 1 and one to Eruption 2. We believe that this result reflects the nature of atmospheric advection-dispersal processes, more than high instabilities on numerical results. This has been better explained in the text:

“The presence of this lower altitude layer is confirmed by the pictures of the Calbuco eruptive column taken during Eruption 1 (Romero et al., 2016; Castruccio et al., 2016) and from the analysis

of the tephra deposit (Romero et al., 2016). This highlights the accuracy of our numerical technique in reproducing and unravelling the complex evolution of plume emission and dispersion.”

Another case illustrates these unstable results : neighbour pixels in the subplot taken from Fig. 3 above show a drastic difference in altitude: turquoise blue pixels (5 km altitude) are direct neighbours of red pixels (23 km altitude) emitted at different times of injection.

The fact that neighbour pixels may contain SO<sub>2</sub> located at different heights is due to the nature of the emission. Two SO<sub>2</sub> parcels injected into the atmosphere at different time and height are advected by different wind speeds. This is what is illustrated in Figure 3, where the SO<sub>2</sub> parcels injected at more than 20 km are able to travel faster into the atmosphere than the ones injected at lower heights. This not exclude that in a 2D vision, as the one from space, they may appear occupying neighbour pixels.

According to this instability of the results, one can hardly envisage an uncertainty on the SO<sub>2</sub> height (at distance or above the vent) of less than 1 km, as presented in Fig.3- d/Fig.3-e. The authors report a mean uncertainty of 0.5 km (Line 30 page 7). In the present state of the paper, this uncertainty appears to be greatly underestimated. Indeed, the authors only illustrate a single solution for the set of variables ( $h$ ,  $h_{vent}$ ,  $t_{vent}$ ). Unfortunately, they do not provide any additional figures that could show intermediate results on the panel of solutions which has been selected so as to allow the reader to apprehend the range of variability of these solutions before the procedure of averaging.

We computed  $h$ ,  $h_{vent}$  and  $t_{vent}$  applying Eq. (2)-(7) as presented in the main text. The fact that we obtain relatively low values for the standard deviations derives from our two step procedure and from the constraint on the eruption time. This allows us to detect a well-defined range of possible plume altitudes ( $\bar{h} \pm \sigma$  and  $\bar{h}_{vent} \pm \sigma_{vent}$ ) both at vent and at satellite overpass. This is particularly true for pixels of Eruption 1 which have travelled further to the vent position. However, this range appears to be wider when looking at the pixels located near the volcanic vent position (Figure 4). In this case our uncertainties are of about 5 km. This reflects the difficulty to retrieve plume heights when the cloud is still close to the vent. In order to show better our uncertainties on plume height, we rescale the colorbars of panels (d) and (e) of Figure 3 and 4. Now uncertainties are displayed in the range 0-5 km and not 0-9 km as did before.

One might wonder if the very small uncertainty on altitude could be due to the rather narrow range of injection time prescribed in input by the authors (the reason for choosing such a narrow range is not explicited in the paper). In other words, the posterior uncertainty reported by the authors seems artificially underestimated by severely restricting the size of the manifold of a priori parameters. If emissions were allowed to occur beyond the start/stop times prescribed by the authors, then a much broader range of  $h$ ,  $h_{vent}$  and  $t_{vent}$  would almost certainly be obtained.

We specified the use of the eruption time in the main text as:

“After having reduced the number of possible trajectories going forward in time up to the time acquisition of the 24 April image, 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. 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 Eruption2. It is the inclusion of accurate information on the timing of the eruption from independent observations which allows our approach to reveal details of the eruption evolution, and represents one of the main innovations of this work.”

The use of the well-constrained eruption time is a key point in order to obtain results which are not highly uncertain. In our opinion, there is no need to vary the eruption time since it is well documented and constrained, see above. Moreover, we do not restrict any a priori parameter. First, we operate our trajectory analysis on the GOME-2 image retrieved at 2.5 km. This means that no initial conjectures are done on plume altitude and that the SO<sub>2</sub> abundance is overestimated. In this way more pixels are included in our retrieval procedure and thus a wider range of possible scenario is examined. The other parameter which plays a key role in our plume height retrieval is the distance of approach used in order to select the acceptable trajectories. This parameter reflects the radius of the umbrella cloud which is a function of the mass flow rate. Since we do not want to operate any restriction on it, we operate a sensitivity analysis using mass eruption rates inferred from previous papers using a wide range of techniques (from deposit analysis to satellite detection).

To conclude, we believe that the point raised by the reviewer about the difficulty of obtaining stable results on plume altitudes from satellite images is true. However, in order to deal with such issue, we used well documented information on Calbuco volcanic activity (eruption time) and in case of data not univocally fixed (mass flow rate) we operated a sensitivity analysis.

**R2.** Results on altitude presented in Fig. 3 and 4 are also suspicious. The authors find approximately the same altitude for both the SO<sub>2</sub> injection at the vent and the pixel of the satellite image at distance from the volcano. This result means that a parcel of SO<sub>2</sub> emitted at a specific altitude is detected by the satellite at the exact same altitude, even after a travel of several hundred (up to a thousand) kilometers. In other words, the plume travels at constant altitude at continental scale. This implicitly indicates an extremely stratified and stable atmospheric environment all over the region of study, which includes the Andes Mountains. This is really surprising and doubtful.

**A2.** The almost constant height between emission and satellite overpass is not due to our procedure, but to wind field data. We used the ECMWF - ERA Interim, Daily dataset. Wind vertical velocity at stratospheric heights, as those experimented by the Calbuco plume, is really low in comparison to the horizontal one. Differences in plume heights between the injection time and the satellite overpass are of few hundred meters. In the following, we show

some of the data used to produce Figure 3 and 4. A difference in the plume altitude between the satellite measurement time (#h) and the injection time (#hvent) can be seen for the investigated pixels (#lat, #lon).

#lat	#lon	#h	#hvent
-34.500	-77.000	10250.000	11689.283
-34.200	-77.000	10250.000	11713.822
-34.500	-76.700	10250.000	11711.356
-39.300	-69.800	5718.839	6511.175
-39.000	-69.800	5567.065	5924.320

**R3.** Authors report a 10 min time resolution for their retrieved time series of flux and altitude, while they also mention ' an uncertainty on injection time in 0 – 110 min with a mean value of 45 min ' (caption of Fig. 4). Isn't it contradictory? (by the way, more information on the calculation of the uncertainty on injection time should be provided)

**A3.** Results of Figure 4 are obtained using mean value for the time instant. This is why we computed fluxes and injection height time series with a time resolution of 10 minutes.

**R4.** For all the issues mentioned above, the validity of the evaluation of the variables (altitude of satellite image pixels, associated altitude of injection, associated time of injection) is questionable. As the time series of flux and injection altitude are deduced from (1), their validity is also questionable. Unfortunately, a weak validation of the results is presented with only comparison with mean values of altitudes roughly evaluated from tephra deposit studies and remote sensing methods. However, the authors could compare their results on pixel altitude with for example the altitude of the SO<sub>2</sub> plume retrieved from IASI satellite images.

See the paper of Begue et al. 2017 in discussion in ACPD, which provide IASI SO<sub>2</sub> altitudes on 24/04 in Fig 3:

<https://www.atmos-chem-phys-discuss.net/acp-2017-544/>

One might wonder if the problem posed here is not underdetermined. At least to check the validity of the results, I recommend using a longer time series of GOME-2 satellite images (which exists for this eruption, as mentioned by the authors). It would at least allow for checking that the trajectory is correctly modeled by comparison with GOME-2 images which are acquired later.

**A4** According to the replies provided above, we do not agree with the reviewer. Clearly our results have some uncertainty, and we have investigated it in detail. However, this does not mean that our results are not valid. According to the suggestion of the reviewer, we have tested the validity of our results comparing them with GOME-2 images acquired later. We produced a new figure which has been inserted in the text where we show the comparison

between the plume as seen by GOME-2 on 24, 25 and 26 April and as simulated by our trajectory analysis. A good match emerges as discussed in the text.

The paper by Begue et al. [2017] was not cited in the original version of the manuscript since it had been submitted just few days before our work and we were not aware of it. This contribution is now cited in the text.

**R5.** No reference of studies published on the same topic.

Another major concern is that the authors do not properly acknowledge in the introduction section papers that have already been published on the exact same subject, such as:

1- papers that develop algorithms that allow for retrieving both the volcanic SO<sub>2</sub> column amount and plume height at a given pixel of a satellite image collected from various UV and thermal IR sensors (OMI : Yang et al., 2010; Nowlan et al., 2011; Rix et al., 2012; IASI : Carboni et al., 2012; Clarisse et al., 2014; Carboni et al., 2015).

2- papers which presented robust methods for retrieving both the flux and injection altitude of volcanic SO<sub>2</sub> emissions from satellite imagery (e.g. Moxnes et al. 2014, Boichu et al. 2015, Heng et al. 2016).

**A5.** Carboni et al., 2012, Clarisse et al., 2014 and Rix et al., 2012 were cited and referenced in the original paper we submitted. The contribution of Boichu was mentioned with the paper:

Boichu, M., Menut, L., Khvorostyanov, D., Clarisse, L., Clerbaux, C., Turquety, S. and Coheur, P.-F.: Inverting for volcanic SO<sub>2</sub> flux at high temporal resolution using spaceborne plume imagery and chemistry-transport modelling: the 2010 Eyjafjallajökull eruption case-study, *Atmospheric Chemistry and Physics*, 13(17), 8569–8584, doi:10.5194/acp-13-8569-2013, 2013.

The other references have been added, thanks for the suggestions.