Response to RC2

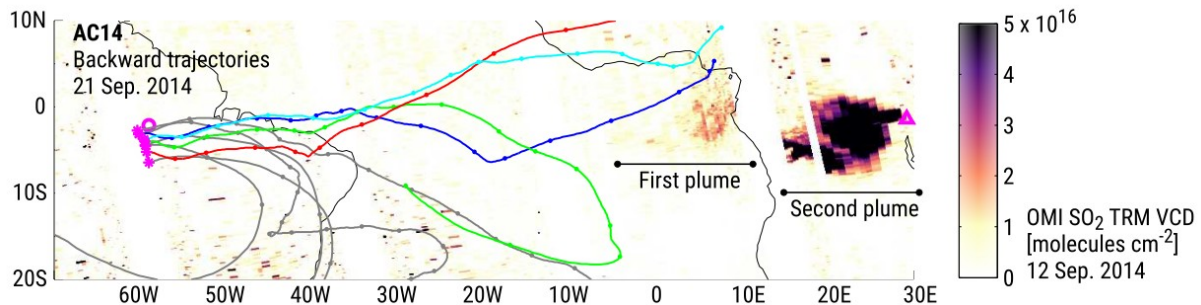
We appreciate the reviewer comments and ideas that helped to improve the manuscript. Our responses are presented below. The text with gray background shows the original comments from the reviewer.

This paper investigates an interesting topic, namely the large scale impact on atmospheric properties induced by tropospheric volcanic eruptions. The authors argue
that two events of volcanic emission from Africa, more precisely from the Nyiragongo-Nyamuragira volcanic system, could modify, after a long-range transport, the properties of the troposphere over the Amazon rain forest region.
To reach this conclusion, the authors present two aerosol datasets collected a few
days apart, from an airborne flight over the Amazon region and from a ground station situated at a location nearby. These observations show anomalies in sulfate mass concentration and sulfate-to-organic aerosols (sulfate-to-OA) mass ratio. To support the volcanic origin of these anomalies, the authors explore sulfur dioxide satellite observations and perform trajectory model simulations.
The paper is well written, figures are clear and interesting aerosol datasets are presented which raise intriguing questions concerning their origin. However, as currently presented, the volcanic origin of the observed rise in sulfate aerosol concentration over the Amazon region is not convincing. As listed and developed in the following, various pending questions need to be answered to clearly proove this volcanic origin.
Backward and forward trajectory model simulations are presented by the authors as a strong evidence of their volcanic origin.

1- However, neither backward trajectory initiated at various points along the flight track (on 21 Sept 2014) reaches the volcanic system (Fig. 6). At best, they reach a distance situated at a distance of ∼2000 km apart. Time is not indicated on the trajectories but would these backward trajectories get closer to Congolese volcanoes if they had been simulated over a longer time duration ?

Trajectory calculations become increasingly uncertain the longer they are run, forwards or backwards, which limits their usefulness for transport over very large distances, such as is the case here, where source and receptor are some 9800 km apart. The trajectories presented in Fig. 6 reach the first plume location around five days after being emitted by the Nyamuragira. Given that the HYSPLIT model has a total error of 15 − 30 % of the travel distance (https://www.arl.noaa.gov/hysplit/hysplit-frequently-asked-questions-faqs/faq-hg11/, last access 19 March 2018), running the backward trajectories back to Nyamuragira

would produce a result with large uncertainties, which will not contribute too much to the discussion. By having the OMI SO2 data, we are able to run shorter trajectories that reach the location of the plume at a given time (12 September 2014) as the source of the measured aerosol during flight AC14. In order to clarify our argument we have edited the figure accordingly, see below.

Edited Fig. 6 (Fig. 7 in the revised version):

Typically, about 10 days are about as long as trajectories give more or less reliable results, with the accuracy depending on the specific meteorological conditions along the track. The other issue that must be considered here is that the emissions from a point source do not travel as a single parcel, but get spread out horizontally and vertically by diffusive processes. Consequently, the points along the trajectory can be seen as the center of an expanding plume, which is spreading both physically by diffusion and probabilistically by stochastic processes in the atmosphere. In the trajectory calculation mode, HYSPLIT does not consider this diffusion but treats the emission as if it were a neutrally-buoyant balloon released at the source (or time-inverted at the endpoint) following a deterministic path (Stein et al., 2015). The true physical dispersion of the plume is readily seen in Figure 7 (new). In this paper, we mitigate these problems by attaching the trajectories to three fixed points: the location of the suspected source, the know location of the receptor site, and the region in between, where the SO2 from the volcano is detected by remote sensing. Forward trajectories from the volcano confirm that the SO2 seen by OMI indeed come from the volcano, as the trajectories presented in Fig. 6 reach the first plume location around five days after being emitted by the Nyamuragira.

2- Similarly, why are forward trajectories in Fig. 3 initiated on 13 Sept at about 2000 km from the volcanic source ? Why are they not initiated precisely at the volcanic source on 12 September when OMI SO2 satellite observations record the strongest emissions?

As mentioned before, our approach in the HYSPLIT modeling was to run the trajectories as short as possible to avoid large uncertainties. Therefore, we used the OMI data to locate the plume and initialize the trajectory modeling further in time to increase the level of confidence. Since the altitude of the plume is unknown, the trajectories were initialized from several points and heights. The OMI SO_2 data also helped us to decide which starting height was in best agreement with the plume location in the subsequent days. The results shown in Fig. 3 (Fig. 4 in the revised version) correspond to trajectories initialized from the plume location on 13 September 2014 and are an indication that the plume was effectively transported westwards to South America.

We addressed the issue brought up by the reviewer by adding the following text to the revised manuscript in section 3.1:

"This approach of using the OMI data to evaluate the trajectories was used also with the purpose of minimizing uncertainties by calculating shorter trajectories instead of initializing them from the volcano location. It should be noted here that the trajectory calculations by HYSPLIT yield a line, which can be understood as the center line of a propagating plume that widens both by stochastic uncertainty in the model calculations and by diffusive processes in the atmosphere. Consequently, the location of the plume becomes more uncertain the longer the model is run, and the physical size of the plume increases as well. Given the 9000-km distance between the volcanoes and ATTO, the uncertainty in the calculated plume trajectory position would become very large. To mitigate this problem, we use a multi-step approach, where we follow the emissions using the satellite-observed locations of the SO2 plume for the initial days, and then calculate forward trajectories from the observed location of the plume.".

3- From Fig. 4, which explores the origin in sulfate aerosol increase observed at the ATTO ground site from 21 to 29 Sept 2014, most backward trajectories started at the ATTO site do not reach the volcanic region. More information on the few trajectories that reach the volcanic region would be required : what is the date/time of initiation of these specific trajectories (especially, does this date/time correspond to the largest peak in sulfate observed at ATTO on 27 or 28/09/2014)? What is the altitude at the endpoint of these trajectories which reach the volcanic region ? Are these altitudes consistent with this particular volcanic setting (i.e. altitude of volcanoes) and activity (passive versus eruptive degassing) which will both impact the altitude of injection of SO2 into the atmosphere?

Any individual back trajectory, especially when run over such long time periods, provides rather uncertain information, as discussed above. The purpose of Fig. 4 (Fig. 5 in the revised version) is to provide a statistical information, which illustrates that a significant number, although not the majority of trajectories, followed a direction towards Nyamuragira and the area affected by its emission during the period of interest. Given the long period of time between emission and the measurements at the ATTO site, a quantitative relation between the volcanic activity and the observations cannot be expected using the HYSPLIT model (or any other trajectory model).

4- In Figure 7, it appears that a peak in sulfate aerosols is detected at ATTO on 28 or 29 September 2014. This peak is preceded by a smaller one on 25 September. In order to suggest that these peaks may originate from Nyiragongo-Nyamuragira volcanoes, the authors perform forward trajectories (Fig. 3): a. The authors argue that one forward trajectory might be compatible with a scenario whereby the 25 September peak detected at ATTO would come from the Congolese region. This trajectory would fit with the strongest peak of SO2 emissions, released on 12 September as shown by OMI satellite images. However, this forward trajectory reaches the ATTO site at an altitude of 1.8 km while measurements are in fact performed at an altitude of 300 m. How do the authors explain this discrepancy ? b. Surprisingly, the authors do not attempt to perform a similar analysis for the strongest peak in sulfate which is recorded at ATTO ground station on 29 September. Why is it so ?

The highest concentration of sulfate shown in Fig. 7 (Fig. 8 in the revised version) was observed on 26 September 2014, not on 29 September as pointed out by the reviewer. This date fits with the forward trajectories presented in Fig. 3 (Fig. 4 in the revised version). Indeed, the trajectory model results in altitudes of 1.8 km when these air masses arrive at the ATTO site.

As discussed above, the locations along the trajectory should be interpreted as the centers of a plume, which had dispersed both physically and probabilistically. This dispersion is not considered in the trajectory mode of HYSPLIT (Stein et al., 2015), but of course takes place in reality both by turbulent and convective processes. Consequently, a trajectory "parcel" moving at 1.8 km altitude will certainly be dispersed to the surface in the highly convective Amazonian atmosphere. In fact, what is most striking is that the plume is even detectable at all, given the dispersion that must take place along this long transport path.

We addressed the issue brought up by the referee by replacing the original section in page 9, line 229-232:

> "The southernmost trajectories reach South America and come within several hundreds of kilometers of ATTO within 15 days. One of them reached ATTO on 25 September at 1.8 km altitude, whereas the other one passed at an altitude of 1.5 km at the point nearest to ATTO on 24 September."

by the following update version:

> "The southernmost trajectories reach South America and come within several hundreds of kilometers of ATTO within 15 days, which is well within the HYSPLIT uncertainty, estimated at 15-30 % of the trajectory length (https://www.arl.noaa.gov/hysplit/hysplit-frequently-asked-questions-faqs/faq-hg11/, last access: 19 March 2018). One of them reached ATTO on 25 September at 1.8 km altitude, whereas the other one passed at an altitude of 1.5 km at the point nearest to ATTO on 24 September. As discussed above, the locations along the trajectory should be interpreted as the centers of a plume, which had dispersed both vertically and horizontally. This dispersion is not considered in the trajectory mode of HYSPLIT (Stein et al., 2015), but of course takes place in reality by turbulent and convective processes. Consequently, a trajectory "parcel" moving at 1.8 km altitude will certainly be dispersed to the surface in the highly convective Amazonian lower troposphere."

5- In forward trajectories of Figure 3, could you please mention the altitude of the endpoint of the trajectory crossing the ATTO site ? We expect this altitude to be close to the ground (or in the first hundred of meters) to justify the increase in sulfate recorded by the ground station.

The ending altitude of the trajectories shown in Fig. 3 (Fig. 4 in the revised version) is around 1-2 km height but, as mentioned before, the trajectory line is just the centerline of a dispersing plume that will be dispersed to the surface.

Given the questions listed above, the sentence Line 270-272 appears as an overstatement.

The statement has been modified in the new version.
Original section in page 10, line 270-272:

> "Nevertheless, the fact that forward and backward trajectories calculated from various starting points and times agree on the sulfate source is a strong indication that the sulfate plumes observed at and near ATTO originate from the Nyamuragira volcano".

Updated version:

> "Nevertheless, the fact that forward and backward trajectories calculated from various starting points and times agree on the sulfate source is a further indication that the sulfate plumes observed at and near ATTO originate from the Nyamuragira volcano".

Concerning aerosol datasets :
1- How do the authors explain that : a. Little increase in sulfate is recorded on the ground at ATTO on 21/09/2014 while a strong anomaly in sulfate-to-OA is observed onboard the flight ? b. Similarly, a strong peak in sulfate is observed on the ground on 28 or 29/09/2014 while no anomaly was recorded onboard flights. Does this reflect a very contrasted atmospheric behaviour of the volcanic cloud?

The flight tracks were not planned to look for or intersect the plume, as the campaign had very different objectives and the flights went into different regions of the Amazon each flight day. The aircraft thus intercepted the plume only by coincidence. In fact, nothing was known even about the possibility of the existence of such a plume during the campaign. It would have been extremely difficult to look for the plume, even if we had known about its existence, since there are no tools to remotely locate the plume once it has been oxidized to sulfate (not detectable by OMI anymore). However, we can use the "volcanic signature" to determine which flights were affected by the volcanic emission and we found this was only clear in flights AC14 and AC17. The fact that the observation on 26 September 2014 at the ATTO site was the strongest one agrees with the stronger emission event by the Nyamuragira on 12 September 2014 compared to the previous one on 7 September 2014. Given the broad area covered by the aircraft measurements, a direct comparison to ATTO measurements is not possible.

2- Surprisingly, another peak in sulfate-to-OA (»1) is also observed on 27/09/2014 according to Fig. S5. The authors argue that such a high ratio is representative of a volcanic pollution to interpret the peak observed on 21/09/2014. Why do the authors chose not to study this event on 27/09/14 ? Would it be also of volcanic origin ?

Presumably, the aircraft observations on 27 September 2014 were also affected by the volcanic plume as pointed out by the referee and we are mentioning this fact in the revised version of the manuscript. We focused only on the observations collected on 21 September 2014 because the volcanic signature was more evident during that flight and we consider it enough illustration of the vertical profile of the volcanic plume. However, the rest of the information from the flight campaign was included in the supplementary material as a reference to the reader.

3- More generally, the Nyamuragira-Nyiragongo volcanic system is recognized as a worldwide major emitter, constantly producing large emissions of SO2, as shown by Figure 2. Carrying out a more systematic study over the 2011-2016 period illustrated in Fig. 2, instead of just focusing on two isolated events of aerosol detections, would certainly provide more convincing arguments in favor of a volcanic origin of the anomalies detected at one (or, better, at several) ground stations in the Amazon region. The authors quickly discard the potential contribution of biomass burning fires to the detected anomalies in sulfate-to-OA mass ratio. This would require more explanations and references. Furthermore, even if local sources were to be excluded, the authors could bring to the reader's attention the fact that large biomass burning fires have also been documented in the Congo basin. In fact, fires in this African region were the second highest after Brazil for the period 2005-2009 (de Sherbinin et al., Env Res. Lett. 2014). Since the geographic location of these fires roughly corresponds to that of the Nyiamuragira-Nyiragongo volcanoes, it is not enough to rely on trajectory analyses to distinguish them. A clear and independent argument has to be put forward to back up the hypothesis of a volcanic origin. For these reasons, it is very important that the authors provide a more in-depth analysis of the significance of the sulfate-to-OA mass ratio.

We understand the concerns of the reviewer regarding biomass burning (BB) as a potential sulfate source, especially because in this period of the year fire events occur often in the south of the Amazon and also close to the Nyamuragira volcano in Congo. We dismissed this possibility based on certain aerosol properties observed during the volcanic event (Nya2014), like (a) comparatively low black carbon mass concentration, (b) increased single scattering albedo, and (c) increased sulfate-to-OA mass ratio (BB emits important amounts of organics). The first two aspects are discussed in our reply to RC1, where we have included the modifications to the manuscript, and a new figure that was included in the

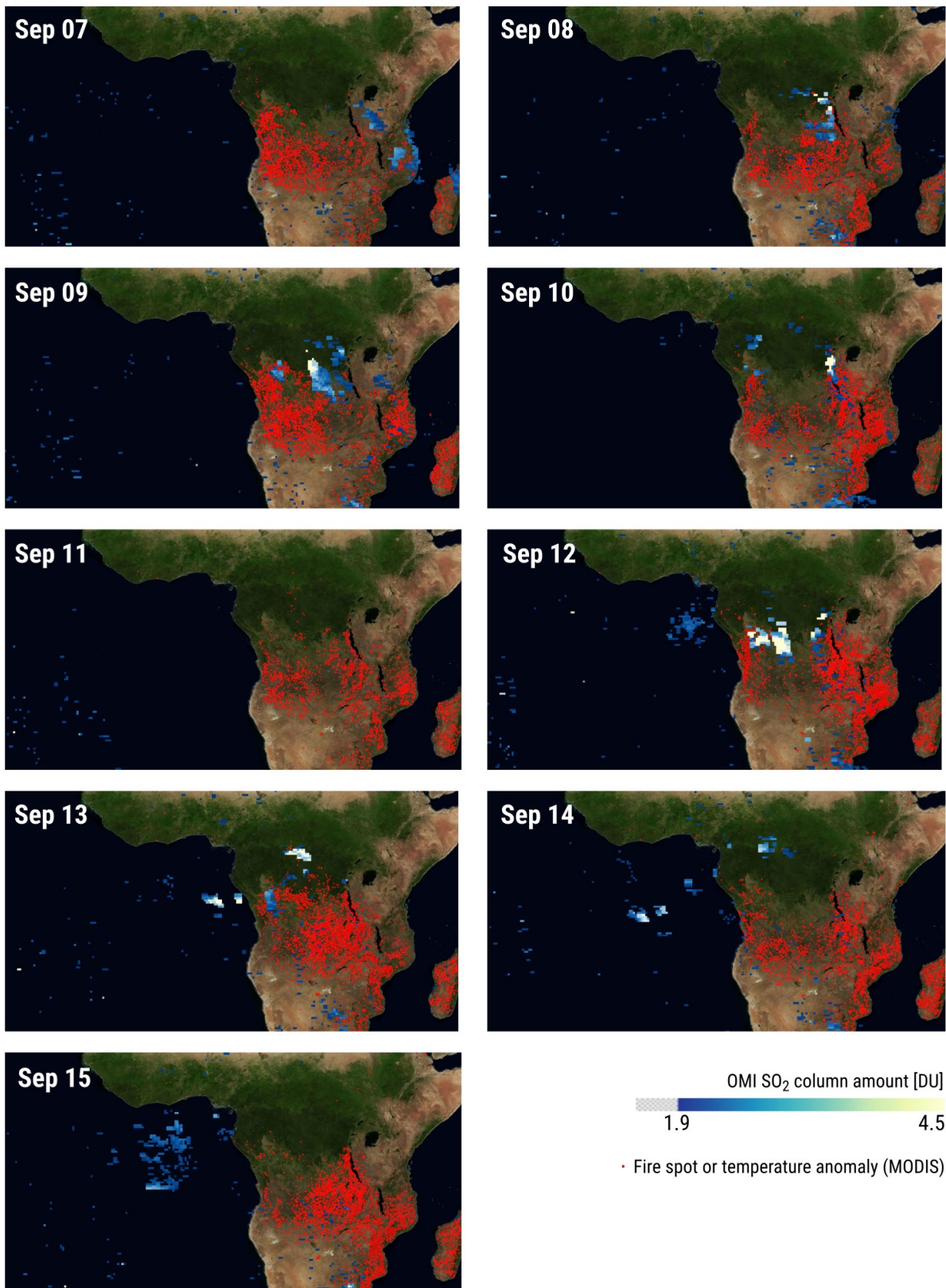
revised version (Fig. 2). Regarding the third aspect we have included the following text to the introduction:

> "Observations in the Amazon rain forest have shown that sulfate and organic aerosol (OA) mass concentrations can increase up to ten times from the wet to the dry season with rather stable sulfate-to-OA mass ratio throughout the year (Andreae et al., 2015; Fuzzi et al., 2007; Martin et al., 2010)."

Additionally, using MODIS data, we observed that the occurrence of fire events in Africa was not related to the SO_2 emissions as can be seen in the following figure:

2014

Sep 07 · Sep 08 · Sep 09 · Sep 10 · Sep 11 · Sep 12 · Sep 13 · Sep 14 · Sep 15

OMI SO$_2$ column amount [DU]
1.9   4.5
· Fire spot or temperature anomaly (MODIS)

We are aware of the large volcanic degassing from the Nyamuragira since 2012, as discussed and supported by Fig. 2 (Fig. 3 in the revised version). However, chemically-speciated aerosol observations at the ATTO site only started by middle 2014. A systematic

study of aerosol source apportionment at the ATTO site is difficult given the complexity of the Amazonian atmospheric aerosol composition and the number of sources that provide different kind of particles. The observations presented in this study show a special case when all conditions were given to make possible the identification of volcanogenic aerosol at ground level over the rest of the characteristic signatures of additional sources. These special conditions include: (i) the strongest Nyamuragira degassing event observed by remote sensors in the period 2012 to 2017, (ii) and air masses originated in Congo that were transported over the Atlantic Ocean towards central Amazonia. This was not always the case during 2014 (see Fig. S2).

Secondary comments:

- Introduction Line 58-65: the authors mention how volcanic eruptions can have a large-scale impact on the atmosphere. However, the authors should explicitely distinguish the impacts of tropospheric vs. stratospheric eruptions, which are different and presently mixed in the current draft.

We addressed the issue brought up by the referee by replacing the original section in page 2, line 59-61:

> "Two prominent examples are the Pinatubo eruption in 1991 (Kirchner et al., 1999) and the 2014 − 2015 eruption of the Holuhraun volcano in Iceland (Ilyinskaya et al., 2017)".

by the following update version:

> "Two prominent examples are the Pinatubo eruption in 1991 that reached the stratosphere (> 10 km) (Kirchner et al., 1999) and the 2014 − 2015 tropospheric effusive eruption of the Holuhraun volcano in Iceland (Ilyinskaya et al., 2017)".

- As it is the topic of their paper, the authors should precisely focus on the atmospheric impact of tropospheric eruptions and develop more on the studies that have already been carried out in this field (just one reference is cited, which is insufficient in view of the number of studies already published on this topic)

We are including a new reference to the Nyamuragira-Nyiragongo emissions in the period 2014 − 2017, which is of relevance to our study.

Reference: Barrière, J., Oth, A., Theys, N., D'Oreye, N. and Kervyn, F.: Long-term monitoring of long-period seismicity and space-based SO2 observations at African lava lake volcanoes Nyiragongo and Nyamulagira (DR Congo), Geophys. Res. Lett., 44(12), 6020–6029, doi:10.1002/2017GL073348, 2017.

- If available, the authors should provide more information on the volcanic activity of the Nyiragongo-Nyamuragira system and mention especially the type of degassing activity (passive vs eruptive) with time, which will impact the altitude of injection of SO2 into the atmosphere and its lifetime.

Volcanic activity reported by Barrière et al. (2017) has been included in section 2.4 of the revised manuscript as follows:

> Nyamuragira produced frequent intensive SO_2 emission events in 2014 with a mean emission of 14.4×10^6 kg SO_2 day^{-1} (Barrière et al., 2017). According to Barrière et al. (2017), the emissions from June to October 2014 where mostly due to lava fountaining activity in the Nyamuragira, characterized by strong tremors.

- Line 265 : Fig. S3 illustrating SO2 emissions detected by OMI satellite on 7 September should be included in the manuscript (and not the supplementary material) as these emissions would explain the anomaly in sulfate-to-OA mass ratio observed during AC17 flight according to the authors.

SO_2 emissions detected by OMI are already presented in Fig. 3 and 6 (Fig. 4 and 7 in the revised manuscript) and we consider including one more figure with these data would be redundant. The supplementary Fig. S3 has been added as a reference of the different specific dates but we consider not necessary to include it in the manuscript.

- Line 218-221 :"Forward trajectories were started at the time of satellite overpass at seven altitudes very consistent patterns were found". The authors should provide in the supplementary material the figure which illustrates these results.

A new figure has been included in the supplementary material to illustrate this point.

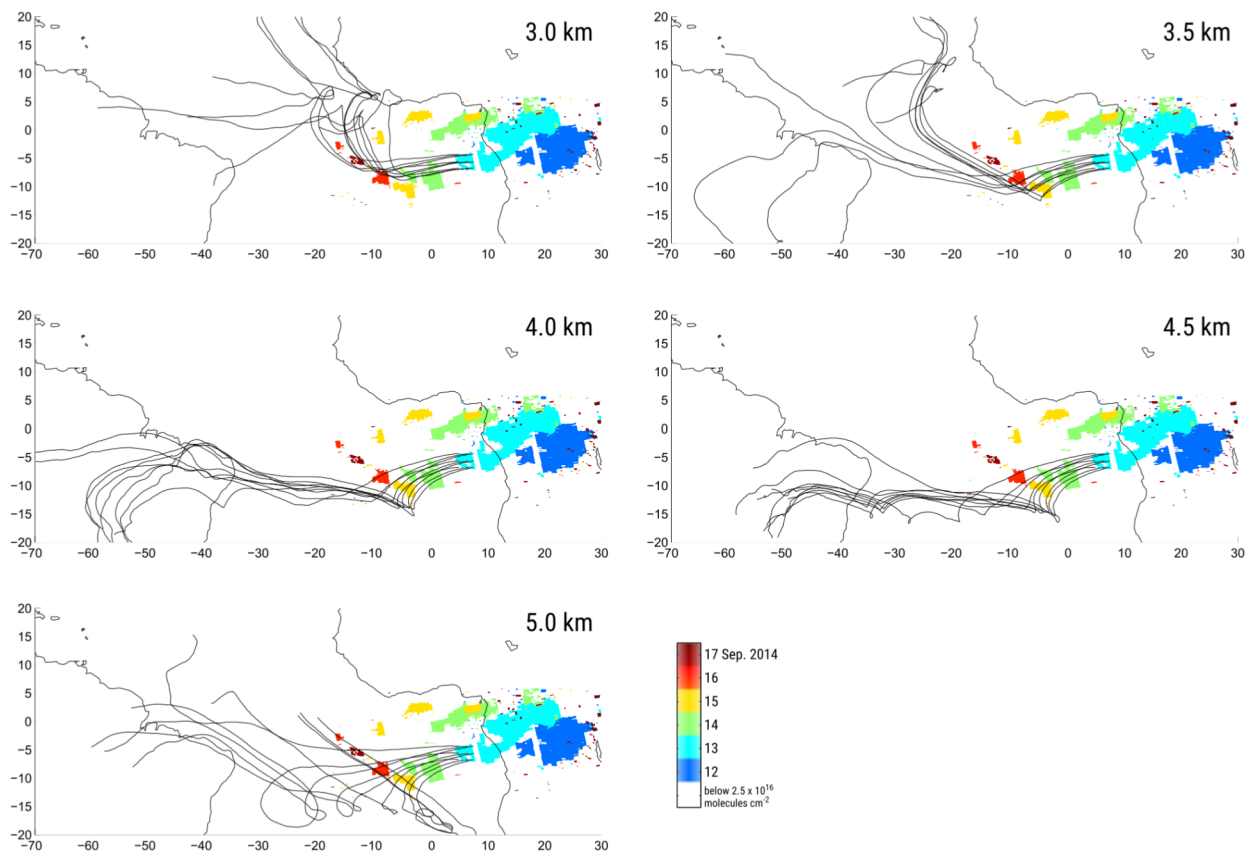



Figure S4. Map of SO$_2$ plumes with VCD > 2.5 × 10^{16} molecules cm^{-2} color-coded by date of observation. Forward trajectories started at different heights above mean sea level, a.m.s.l., as indicated in the upper right corner of each figure.

- Line 223 : "Trajectories started within the leading edge of the plume are in good agreement with the OMI data". How do the authors explain that trajectories initiated in the core of the volcanic plume, i.e the most concentrated part of the plume, would not be in agreement with OMI data ?

In principle, we agree with the reviewer's comment. But again, trajectories cannot be seen as deterministic line connecting source and receptor. At best, they can show that there is a certain degree of plausibility that a parcel emitted at the position of the source can arrive in the vicinity of a receptor site, or that a substance detected at the receptor can have originated in the region of the source.

- Concerning the comparisons of the measured concentrations of sulfate aerosols in section 3.3 : are the listed values all hourly mean values ?

Yes, we used hourly mean values. We have added a comment in section 2.1 of the revised manuscript.

- Hygroscopicity of volcanic aerosols (from Line 322) : the authors should provide more background information on the studies already carried out on volcanic material. Are the results obtained in agreement with previous studies ?

The changes in hygroscopicity were mainly driven by the high sulfate fraction. We have included a reference to a volcanic plume measurement study at Jungfraujoch, Switzerland where the Aitken mode hygroscopicity parameter increased from 0.15 to 0.4 (Bukowiecki et al., 2011). A detailed discussion about the hygroscopicity observations during the Nya2014 event can be found elsewhere (Pöhlker et al., 2017).

- Fig.2 : please indicate on the time series the two degassing events that are studied in the paper.

We have modified the figure accordingly (Fig. 3 in the revised manuscript). It was not possible to indicate both events in the figure since they are separated by 5 days and the time series spans for 6 years but the most important emission event on 12 September 2014 was indicated in the revised version. Additionally, the figure has been updated for a mistake that affected the absolute values shown in the previous version.

References

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