Response to E. Ilyinskaya (RC1)

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.

General comments
The paper presents an interesting novel dataset of atmospheric measurements from the Amazon rain forest, combining ground-based measurements from a long-term monitoring station and airborne measurements from a research aircraft campaign. The paper also presents results from the OMI satellite and air mass trajectory modelling.
The main conclusion is that the enhanced sulphate observed over the Amazon rain forest is sourced from two active volcanoes in Central Africa ~10.000 km away. In my opinion, while the dataset is good, the main conclusion is not sufficiently well supported by it. The reasons for this are outlined in Specific comments. The manuscript is well written, easy to follow, and the figures are generally well made with few relatively minor exceptions (see Technical comments)

We agree with most of the comments presented by the reviewer. After a major revision of the manuscript, we consider we have addressed all the reviewer concerns.

Specific comments
1. Source of SO2: The Nyamuragira & Nyiragongo volcanoes are concluded to be the source of the observed sulphate due to their supposedly high SO2 flux. However, it isn't mentioned anywhere in the manuscript how much SO2 they actually emit. It is only mentioned that the emission is either 'high' or 'enhanced' during certain time periods. Therefore it is very hard to judge whether it is credible that these volcanoes emit enough SO2 for the plume to be detectable ~10.000 km away. This is a problem that is easily fixed but it undermined the credibility of the conclusions, as it did not demonstrate that the authors investigated a very basic source term.

We appreciate this comment and understand that SO_2 fluxes are an important aspect that is related to our study. In order to shed some light on this aspect we are adding different references to the new version of the manuscript, which provide information of in-situ emission fluxes (Bobrowski et al., 2017) and also satellite-based emission data (Barrière et al., 2017; Fioletov et al., 2016).

2. Trajectory modelling: There are no direct observations to show that the plume from the Nyamuragira & Nyiragongo volcanoes reaches the monitoring station in the Amazon. The SO2 plume can only be traced by OMI as far as the mid-Atlantic. This is understandable because SO2 eventually becomes too dilute or completely converted into sulphate and therefore undetectable by OMI. Therefore, the conclusion that the volcanic emissions from these two volcanoes can reach the Amazon monitoring station is based on forward- and backward trajectory modelling by HYSPLIT. The following comment is made with the caveat that I am not a modelling specialist, but to me the trajectories do not look sufficiently convincing. For example, the modelled source for the most accurate of the backward trajectories is ~2000 km away from the actual position of the volcanoes (Fig S6). Therefore I was left unconvinced that Nyamuragira & Nyiragongo emissions can reach the Amazon. Could the authors strengthen their results with e.g. other types of models, or by improving the performance of HYSPLIT?

Direct observations over the South Atlantic Ocean are scarce because they require dedicated ship or aircraft campaigns. Our conclusion that the volcanic emissions from the Nyamuragira reached the Amazon is based on multiple lines of evidence (i.e., aerosol ground-based observations, satellite data, and air mass trajectories). The aerosol observations we use to support the volcanic emission reaching the Amazon Basin is based on the characterization of a "volcanic signature", that shows a high sulfate mass concentration, increased single scattering albedo, decreased rBC mass concentration (aircraft observations) and increased sulfate to organic aerosol (OA) mass ratio.

The forward and backward trajectory analysis together with OMI SO_2 data are used to confirm that air masses occurring during the volcanogenic aerosol observation were indeed originated from the volcano and the plume location detected by remote sensors. Actually, HYSPLIT model air mass trajectories should be used with care since the model uncertainties increase with time. In our case, the forward trajectories shown in Fig. 3 (Fig. 4 in the revised version) were initialized on 13 September in order to reduce the total error of the model and by using OMI data we were able to find the trajectories that matched the plume location in the following days. More details on the changes to the manuscript regarding the modeling issues can be found in our reply to RC2.

3. Direct observations: In addition to the trajectory modelling the main conclusion is based on ground- and aircraft measurements that show enhanced sulphate over the Amazon. While I think the data show convincingly that the sulphate was indeed enhanced, the conclusion that it comes from Nyamuragira & Nyiragongo needs more data behind it. The

conclusion is based on a very short time period, approx. 5 September – 10 October 2014, within which there is apparently only one volcanic-sulphate event (21 Sept – 1 Oct). This is essentially one data point. Considering the extremely large distances between source and measurement locations, and the high degree of uncertainty in the trajectory modelling it would have been better to consider a much longer time series so that we can be convinced that these sulphate-enhancement events can be repeatedly traced to Nyamuragira & Nyiragongo volcanoes. It wasn't clear to me why this wasn't done already, as the monitoring station has been in operation since 2012. Nyamuragira has been degassing strongly since 2012 (e.g. Campion, R. (2014), New lava lake at Nyamuragira volcano revealed by combined ASTER and OMI SO2 measurements, Geophys. Res. Lett., 41, 7485–7492, doi:10.1002/2014GL061808) so I suggest the authors consider looking further back in time and try to identify more than one volcanic-sulphate event.

Even though ATTO observations started in 2012, sulfate mass measurements only started in middle 2014. We are aware of the large volcanic degassing from the Nyamuragira since 2012. We discuss this idea in the main text and support it using Fig. 2 (Fig. 3 in the revised version). Regarding the ground- or aircraft-based observations, we consider that given the complexity of the Amazonian atmospheric aerosol composition and the number of sources that provide different kind of particles, identifying a volcanic event was only possible given the exceptionally high emissions of September 2014. The "volcanic signature" could be present at other times but unfortunately they are not identifiable given the presence of biomass burning emissions that could be masking the volcanic signature.

The observations presented in this study show a special case when all conditions were given to make possible the identification of likely volcanogenic aerosol over the Amazon forest. These conditions include: (i) the strongest Nyamuragira degassing event observed by remote sensors in the period 2012 to 2017, and (ii) air masses originated in Congo were transported over the Atlantic Ocean towards central Amazonia. Please note the latter one is not always the case (see Fig. S2). Furthermore, we benefited from the extremely lucky circumstance that this event occurred during the only time ever that a research aircraft equipped to detect such a sulfate plume was present over the Amazon.

Most likely, we actually measured two different sulfur plumes emitted from the Nyamuragira (7 and 12 September 2014). The first one being measured by aircraft instruments on 21 September 2014 at ~4.5 km altitude. The second one, which reached ground level at the ATTO area, with the largest sulfate concentration measured on 26 September 2014. After carefully studying our data, we did not find any other events where the volcanogenic footprint can be distinguished from the rest of the sources.

4. I would like to see more in-depth discussion about why the observed sulphate is conclusively of volcanic origin.

We have included a new paragraph and a new figure at the beginning of section 3 to present an introduction to the aerosol properties and why we conclude they were influenced by volcanic emissions in September 2014.

The new paragraph is the following:

> "The Amazonian dry season (August – November) $M_{sulfate}$ median over 3 years of measurements at the ATTO site was 0.60 μg m^{-3} (0.41 – 0.79, inter-quartile range, IQR) as shown in Fig. 2a. This value slightly increased under the influence of BB (median: 0.83 μg m^{-3}) and did not change significantly during FF combustion influence (median: 0.56 μg m^{-3}). During African dust advection periods, when mineral dust particles are usually mixed with BB emissions and sea-salt aerosol particles, Moran-Zuloaga et al. (2017) measured sulfate mass concentrations of ~0.44 μg m^{-3} using energy-dispersive X-ray analysis and Pöhlker et al. (2017) reported a $M_{sulfate}$ mean of 0.25 ± 0.19 μg m^{-3} measured by ACSM, both studies at the ATTO site. Furthermore, sulfate measurements over the South Atlantic Ocean (Huang et al., 2018) were well below the values measured at ATTO between 21 and 30 September 2014 (median: 1.60 μg m^{-3}, see Fig. 2a). Regarding the single scattering albedo, $\omega_{0,637}$, shown in Fig. 2b, the lowest average was observed during FF influenced periods, indicating the presence of dark aerosol particles, rich in BC, which contrasts with the higher $\omega_{0,637}$ observed in September 2014. As can be seen in Fig. 2, the ATTO observations between 21 and 30 September 2014 are remarkably different from strong BB and FF combustion influence periods in terms of $M_{sulfate}$ and $\omega_{0,637}$. Given this, the elevated $M_{sulfate}$ observed in September 2014 could not be explained by combustion sources. Therefore, the possibility of an additional sulfate source, like volcanogenic sulfate aerosol, was considered. In the following section, satellite data is used to study the possibility of a volcanic plume reaching the Amazon rain forest during the period of interest. The last two sections are dedicated to the discussion of the aerosol physicochemical properties measured by aircraft- and ground-based instruments."

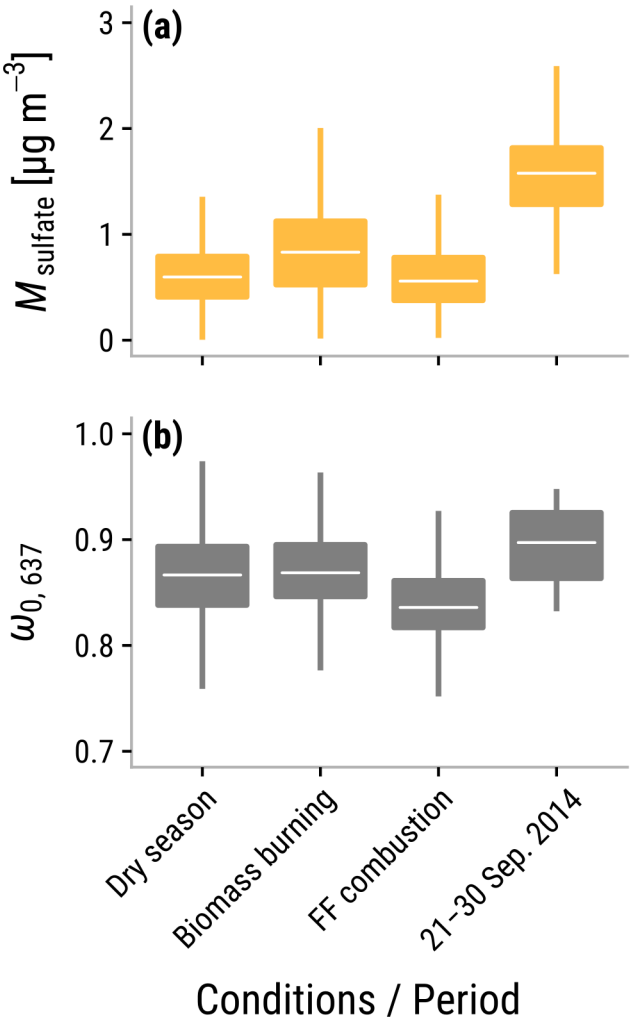

Figure 2. Box and whisker plots of (a) sulfate mass concentration, $M_{sulfate}$, and (b) single scattering albedo at 637 nm, $\omega_{0,637}$, during different periods and conditions including dry season average 2014 – 2016, biomass burning and fossil-fuel combustion influenced conditions and the period of interest from 21 to 30 September 2014. The white segment inside the box represents the median. Lower and upper box edges represent the first and the third quartiles, respectively. The whiskers represent the lowest and highest observations within the 99.3 % confidence interval.

Additionally, the following statement was added to section 3.2:

"The concentrations of rBC in the region between 4 and 5 km were also very low (9 ng m^{-3}), compared to the values below 3 km altitude (270 ng m-3), ruling out a combustion source of the sulfate. In the light of all the observations (i.e., enhanced sulfate layer above 4 km height, high sulfate-to-OA, very low rBC) there is no other

plausible explanation for the source of this sulfate plume than the LRT of volcanogenic aerosols."

Technical comments
Added as notes to the pdf file.

We would like to thank the reviewer for the detailed technical notes to the manuscript. Most of the comments were addressed in the revised version.

References

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.

Fioletov, V. E., McLinden, C. A., Krotkov, N., Li, C., Joiner, J., Theys, N., Carn, S. and Moran, M. D.: A global catalogue of large SO2 sources and emissions derived from the Ozone Monitoring Instrument, Atmos. Chem. Phys., 16(18), 11497–11519, doi:10.5194/acp-16-11497-2016, 2016.