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."

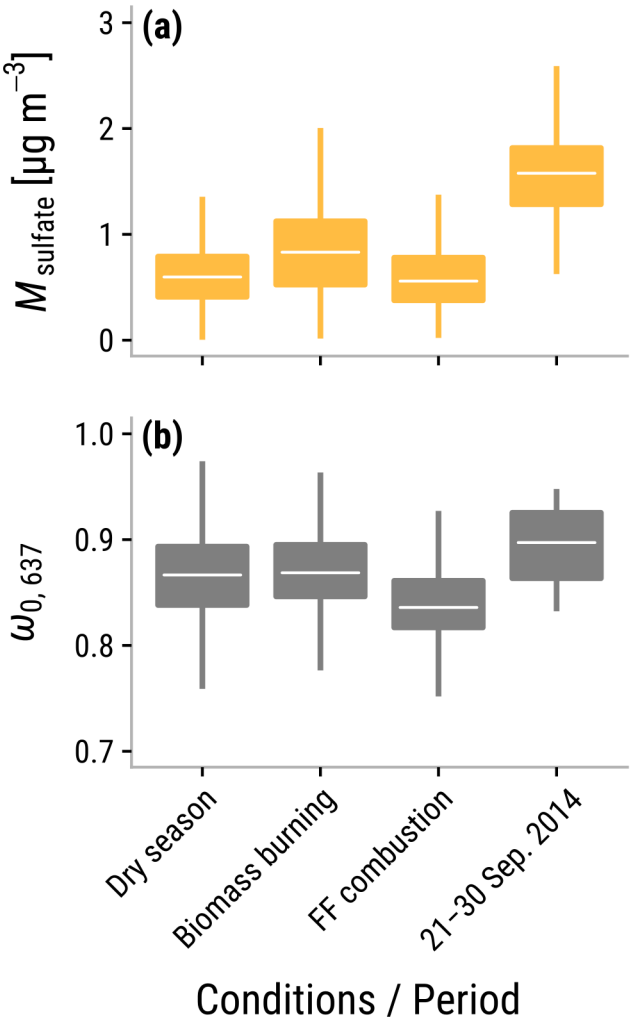


Figure 2. Box and whisker plots of (a) sulfate mass concentration, $M_{sulfate}$, and (b) single scattering albedo at 637 nm, $\omega_{0,637}$, during different periods and conditions including dry season average 2014 – 2016, biomass burning and fossil-fuel combustion influenced conditions and the period 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.

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.

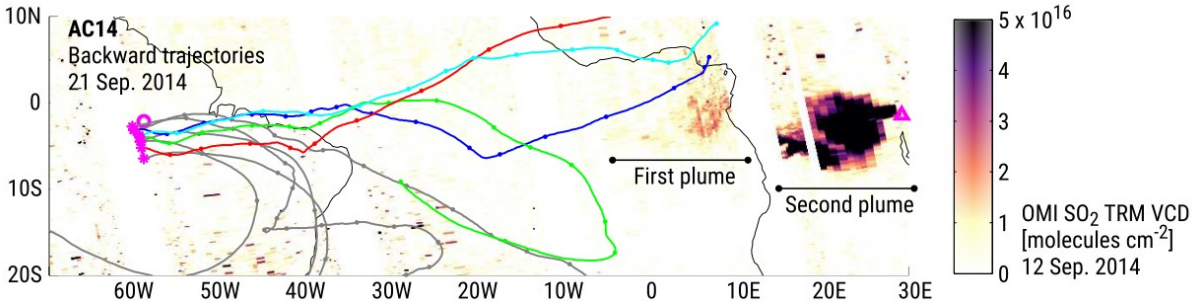
This paper investigates an interesting topic, namely the large scale impact on atmospheric properties induced by tropospheric volcanic eruptions. The authors argue that two events of volcanic emission from Africa, more precisely from the Nyiragongo-Nyamuragira volcanic system, could modify, after a long-range 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.

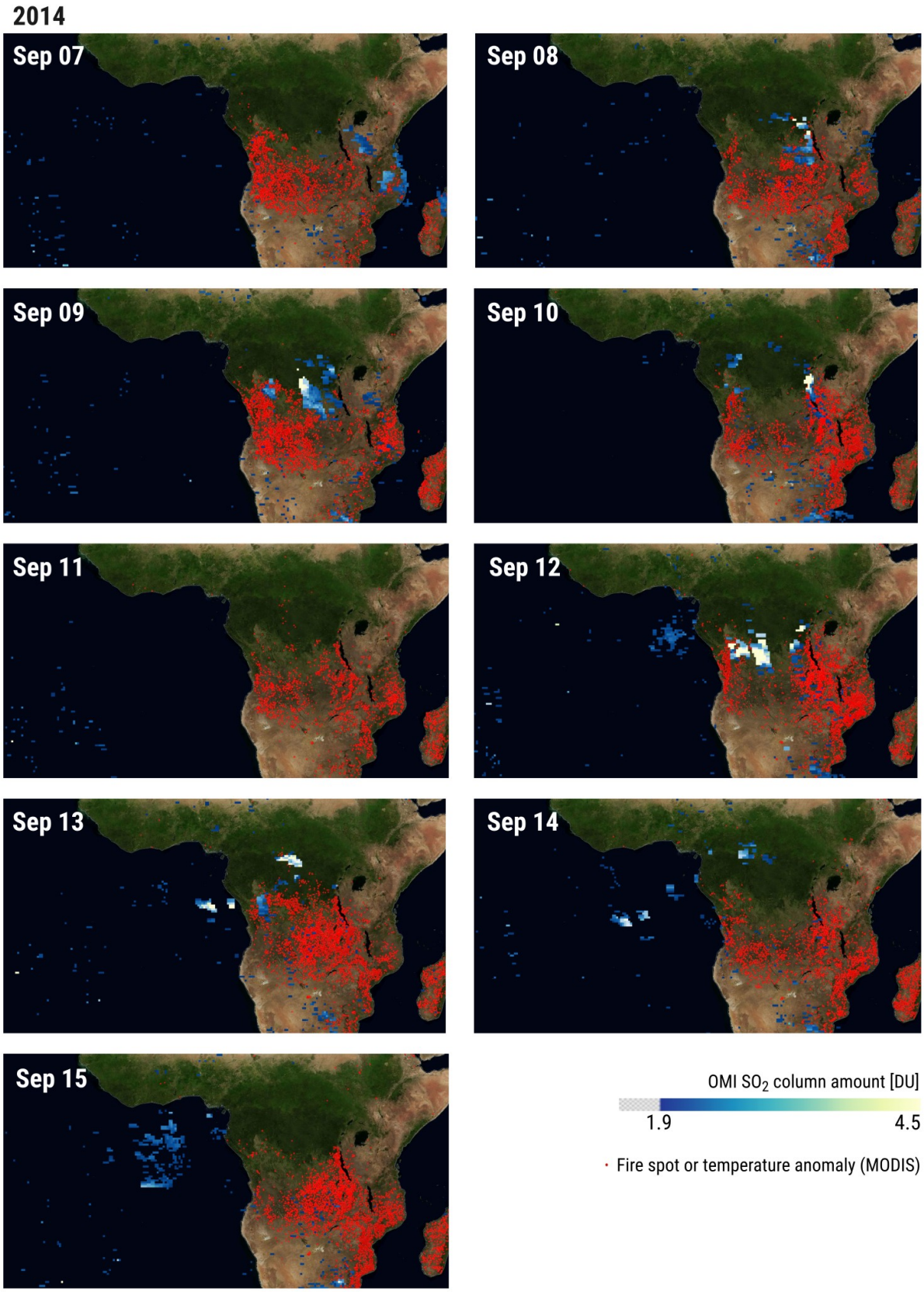
3- More generally, the Nyamuragira-Nyiragongo volcanic system is recognized as a worldwide major emitter, constantly producing large emissions of SO2, as shown by Figure 2. Carrying out a more systematic study over the 2011-2016 period illustrated in Fig. 2, instead of just focusing on two isolated events 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:



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.

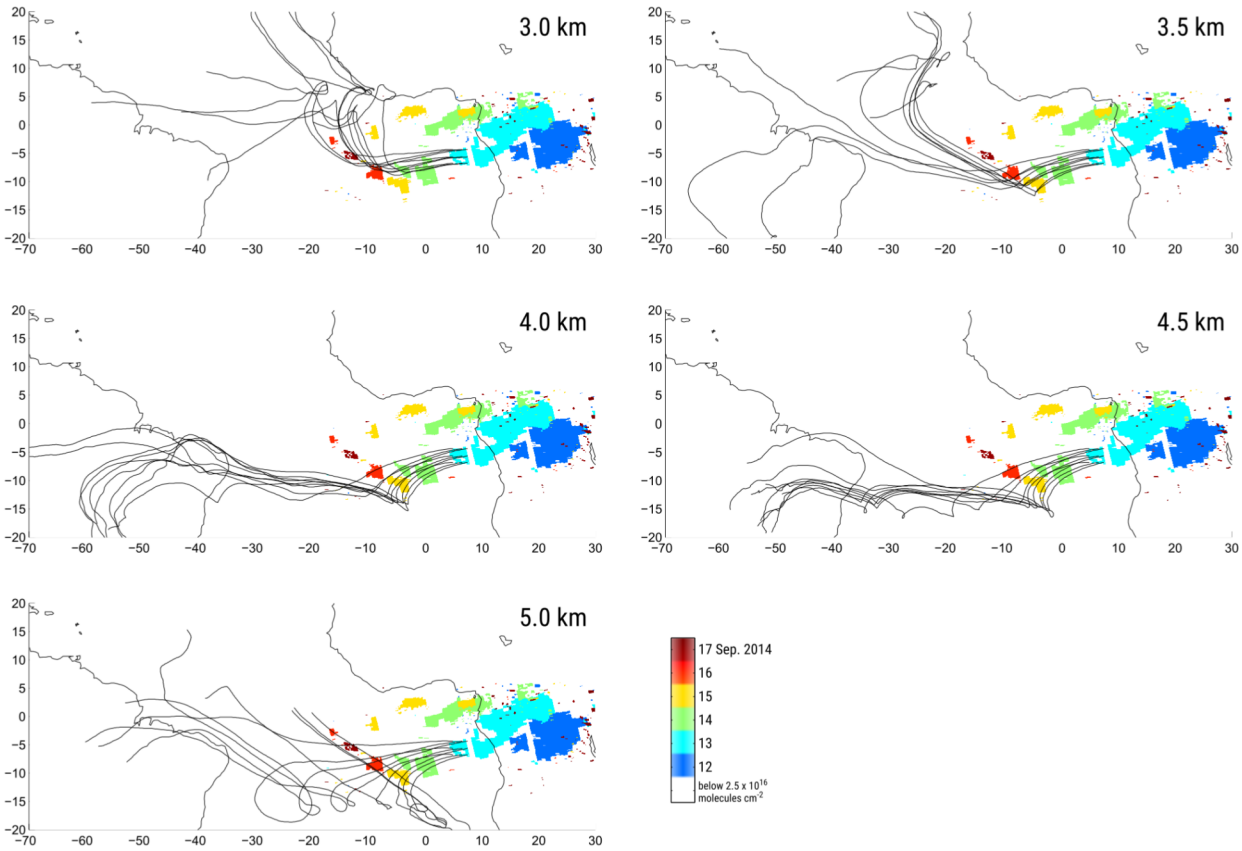


Figure S4. Map of SO_2 plumes with VCD > 2.5×10^{16} molecules cm^{-2} color-coded by date of observation. Forward trajectories started at different heights above mean sea level, a.m.s.l., as indicated in the upper right corner of each figure.

- 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.

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African volcanic emissions influencing atmospheric aerosol ~~particle~~s over the Amazon rain forest

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Abstract.

The long-range transport (LRT) of trace gases and aerosol particles plays an important role for the composition of the Amazonian rain forest atmosphere. Sulfate aerosols originate to a substantial extent from LRT sources and play an important role in the Amazonian atmosphere as strongly light-scattering particles and effective cloud condensation nuclei. The transatlantic transport of volcanic sulfur emissions from Africa has been considered as a source of particulate sulfate in the Amazon, however, direct observations have been lacking so far. This study provides experimental evidence for the influence of volcanic emissions on the Amazonian aerosol properties and atmospheric composition during September 2014, based on comprehensive ground-based and airborne aerosol measurements

together with satellite observations. Under the volcanic influence, hourly mean sulfate mass concentrations in the sub-micron size range reached up to 3.6 μg m^{-3} at the Amazon Tall Tower Observatory, the highest value ever reported in the Amazon region. The substantial sulfate injection increased the aerosol hygroscopicity with κ values up to 0.36, thus, altering aerosol-cloud interactions over the rain forest. Airborne measurements and satellite data indicate that the transatlantic transport of the volcanogenic aerosols occurred in two major volcanic plumes with a sulfate-enhanced layer between 4 and 5 km altitude. This study demonstrates how remote aerosol sources in Africa, such as the volcanic sulfur emissions, can substantially affect the aerosol cycling and atmospheric processes over the Amazon rain forest.

~~Long-range transport (LRT) plays an important role in the Amazon rain forest by bringing in different primary and secondary aerosol particles from distant sources. The atmospheric oxidation of dimethyl sulfide (DMS), emitted from marine plankton, is considered an important sulfate source over the Amazon rain forest, with a lesser contribution from terrestrial soil and vegetation sulfur emissions. Volcanic sulfur emissions from Africa could be a source of particulate sulfate to the Amazonian atmosphere upon transatlantic transport but no observations have been published. By using satellite observations, together with ground-based and airborne aerosol particle observations, this paper provides evidence of the influence that volcanic emissions have on the aerosol properties that have been observed in central Amazonia. Under the volcanic influence, sulfate mass concentrations reached up to 3.6 μg m^{-3} (hourly mean) at ground level, the highest value ever reported in the Amazon region. The hygroscopicity parameter was higher than the characteristic dry-season average, reaching a maximum of 0.36 for accumulation mode aerosol particles. Airborne measurements and satellite data indicated the transport of two different volcanic plumes reaching the Amazon Basin in September 2014 with a sulfate-enhanced layer at an altitude between 4 and 5 km. These observations show that remote volcanic sources can episodically affect the aerosol cycling over the Amazon rain forest and perturb the background conditions. Further studies should address the long-term effect of volcanogenic aerosol particles over the Amazon Basin by running long-term and intensive field measurements in the Amazon region and by monitoring African emissions and their transatlantic transport.~~

1 Introduction

Sulfate aerosol particles are produced in the atmosphere by oxidation of sulfur dioxide (SO_2) emitted by fossil fuel (FF) combustion, volcanic emissions, and by oxidation of reduced sulfur compounds, such as DMS (Andreae and Rosenfeld, 2008). These particles scatter solar radiation and act as efficient cloud condensation nuclei (CCN) (Stevens and Feingold, 2009). Anthropogenic SO_2 emissions have increased over the 20th century to a maximum around the year 1980 and declined somewhat thereafter to around 100 Tg SO_2 per year, but they are still the most important source of sulfur to the atmosphere (Boucher et al., 2013; http://edgar.jrc.ec.europa.eu/overview.php?v=431, last access: 6 Sep 2017). Volcanic emissions are the predominant natural source of SO_2 and account for about 5 % of total annual SO_2 emissions (Textor et al., 2004; Yang et al., 2017). The emitted sulfur dioxide is oxidized in the atmosphere to gaseous sulfuric acid, which is quickly converted to sulfate aerosol particles (Chin et al., 1996; Reiner and Arnold, 1994). According to Chin and Jacob (1996), volcanic sulfur emissions can account for 20 – 40 % of sulfate aerosol particle mass concentrations in the troposphere and might therefore be able to substantially alter the atmospheric composition. Experimental evidence of drastic changes hundreds of kilometers downwind of volcanic eruptions affecting physical and chemical aerosol properties can be found in Mather et al. (2003) and Robock (2000, 2007). 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), where the emissions affected the cloud-drop effective radius (r_{eff}) while other cloud properties, like the cloud optical depth and the cloud liquid water path, remained unaffected (Malavelle et al., 2017; Yuan et al., 2011). Moreover, a connection between tropical volcanic explosive eruptions and El Niño-like events has been described recently (Khodri et al., 2017). Besides explosive-effusive eruptions, small eruptions and passive degassing account for relatively stable SO_2 fluxes (23.0 ± 2.3 Tg yr^{-1}, 2005–2015), and on average emit approximately one order of magnitude more SO_2 compared to explosive eruptions (Carn et al., 2017). To what extent volcanic passive sulfur emissions can affect cloud properties is still debated (Ebmeier et al., 2014; Malavelle et al., 2017).

There are several active volcanoes in Africa. The Nyamuragira-Nyiragongo neighboring volcanoes in the Democratic Republic of the Congo (DRC) were among the most persistent passively degassing volcanoes worldwide between 2004 and 2014, with around 150 days with satellite-detected degassing in

2014 and the highest average SO_2 index observed in the period 2004 – 2014 (Carn et al., 2016). The strong passive degassing activity of Nyamuragira starting in 2011 and culminating in the formation of a lava lake in late 2014 (Campion, 2014) lead to two orders of magnitude higher SO_2 mass emissions from the Nyiragongo-Nyamuragira complex (Barrière et al., 2017; Bobrowski et al., 2017). The emissions from Nyamuragira in 2014 have been estimated at ca. 2.3 Tg SO_2 (Fioletov et al., 2016).

In the Amazon rain forest, biogenic sulfate aerosol is sustained by oceanic DMS emission, and to a lesser degree by hydrogen sulfide (H_2S), methanethiol (MeSH), and DMS emissions from soils and vegetation (Andreae and Andreae, 1988; Jardine et al., 2015; Martin et al., 2010). Occasional anthropogenic sulfur injections have been attributed to open biomass burning and fossil fuel combustion emissions, either from Brazil or, via LRT, from Africa (Andreae et al., 1990; Talbot et al., 1988). Fossil fuel sources (e.g., ship traffic, power plants) do not often influence aerosol particles measured in the remote Amazon forest (Saturno et al., 2017) but can be important downwind of populated areas like Manaus, Brazil (Kuhn et al., 2010; Martin et al., 2016). Recent measurements during the South AMerican Biomass Burning Analysis (SAMBBA) campaign, which focused on biomass burning (BB) emissions, found no correlation between sulfate aerosol and various kinds of BB aerosol particles (Brito et al., 2014). On the other hand, aircraft observations of haze layers at 2 – 4 km altitude over the Amazon rain forest have shown high sulfate enrichment in comparison to the boundary layer and the free troposphere concentrations and indicated these haze layers to be linked to LRT of aerosol particles from Africa (Andreae et al., 1988; Holanda et al., 2018). A modeled global sulfate source attribution study showed that southern Africa peak sulfate concentrations occur between June and August. For this three-month period, estimated emissions were 0.81 Tg S and decreased in the following three months to 0.66 Tg S (Yang et al., 2017). 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).

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In the Amazon rain forest, biogenic sulfate aerosol is sustained by oceanic DMS emission, and to a lesser degree by hydrogen sulfide (H_2S), methanethiol (MeSH), and DMS emissions from soils and

~~vegetation (Andreae and Andreae, 1988; Jardine et al., 2015; Martin et al., 2010). Occasional anthropogenic sulfur injections have been attributed to open biomass burning and fossil fuel combustion emissions, either from Brazil or, via LRT, from Africa (Andreae et al., 1990; Talbot et al., 1988). A recent study suggests that fossil fuel sources (e.g., ship traffic, power plants) do not significantly influence aerosol particles measured in the remote Amazon forest (Saturno et al., 2017) but can be important downwind of populated areas like Manaus, Brazil (Kuhn et al., 2010; Martin et al., 2016). Recent measurements during the South AMerican Biomass Burning Analysis (SAMBBA) campaign, which focused on biomass burning (BB) emissions, found no correlation between sulfate aerosol and various kinds of BB aerosol particles (Brito et al., 2014). On the other hand, aircraft observations of haze layers at 2 – 4 km altitude over the Amazon rain forest have shown high sulfate enrichment in comparison to the boundary layer and the free troposphere concentrations and indicated these haze layers to be linked to LRT of aerosol particles from Africa (Andreae et al., 1988; Holanda et al., 2017). A modeled global sulfate source attribution study showed that southern Africa peak sulfate concentrations occur between June and August. For this three-month period, estimated emissions were 0.81 Tg S and decreased in the following three months to 0.66 Tg S (Yang et al., 2017).~~

Northeasterly and southeasterly trade winds [~~(~~north and south of the inter-tropical convergence zone (ITCZ)] ~~, respectively)~~ are able to transport aerosols over large distances, given the typically weak wet deposition in this latitude band (Wang et al., 2016). During the Amazonian dry season (August – November), the transport of African smoke from southern Africa savanna and shrubland fires is an important source of aerosol in addition to the regional emissions ~~by regional fires in South America~~ (Andreae et al., 1994). ~~When the ITCZ shifts north in the dry season, south-east trade winds originating from southern Africa are more likely to reach the central Amazon rain forest.~~ Even though the potential impact of transatlantic transport of volcanic sulfur emission has been suggested before (Yang et al., 2017), no ground-based evidence has been reported previously in the literature concerning the impact of African volcanic sources.

To analyze a major degassing period of the Nyamuragira in September 2014 and the subsequent transatlantic transport of the volcanic plume, this paper combines satellite and backward trajectory data with measurements at the Amazon Tall Tower Observatory as well as during the ACRIDICON-CHUVA

aircraft campaign in the Amazon Basin. This comprehensive data set indicates that the African volcanic SO_2 emissions were oxidized to particulate sulfate, which reached the central Amazon rain forest and influenced aerosol physical and chemical properties significantly.

~~In this paper, we present satellite observations that show volcanic SO_2 emissions in central Africa that have been transported over the South Atlantic Ocean and reach the Amazon rain forest after being oxidized to particulate sulfate. Satellite, airborne, and ground-based observations are used to show that volcanogenic sulfate can significantly affect the aerosol physical and optical properties over the Amazon Basin during the dry season.~~

2 Data and methods

2.1 Ground-based instrumentation

The ground-based aerosol data presented here have been collected at the Amazon Tall Tower Observatory (ATTO) site [called *T0a* in the GoAmazon2014/5 experiment (Martin et al., 2016)], located in the Uatumã Sustainable Development Reserve, Amazonas, Brazil. Details about the ATTO site infrastructure, instrumentation and an overview of running measurements can be found in Andreae et al. (2015). Figure 1 shows the ATTO site location and the location of the Nyamuragira volcano in the DRC, located at 1.41° S, 29.2° E, 3058 m a.s.l. The long-term measurements at ATTO started in 2012. A systematic backward trajectory (BT) analysis of air masses arriving at ATTO can be found in (C. Pöhlker et al., 2018). The ATTO aerosol measurements were taken on a triangular mast (02° 08.602' S, 59° 00.033' W, 130 m above sea level, a.s.l.) using a 25 mm diameter, 60 m high stainless steel tube with a laminar sampling flow rate of 30 L min^{-1}. The instruments were installed inside an air-conditioned container where the temperature was kept between 29 and 31 °C. All aerosol data were aggregated to hourly mean values. More details on the aerosol sampling setup can be found elsewhere (Moran-Zuloaga et al., 2017; M. L. Pöhlker et al., 2016; Saturno et al., 2017).

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Equivalent black carbon (BC_e) mass concentrations, M_{BCe}, were calculated from absorption measurements by a multi-angle absorption photometer, MAAP (Model 5012, Thermo Electron Group, Waltham, USA). The details of the instrument are described elsewhere (Petzold and Schönlinner, 2004). The BC mass absorption cross-section (MAC) was retrieved from fitting MAAP absorption coefficients at 637 nm wavelength and refractory black carbon (rBC) mass concentrations measured by using a single-particle soot photometer (SP2) revision D (Droplet Measurement Technologies, Longmont~~Boulder~~, USA). Details of the technique can be found in Stephens et al. (2003). The MAC calculations are described in Saturno et al. (2017). Light scattering coefficients were measured using a three-wavelength integrating nephelometer (Aurora 3000, Ecotech Pty Ltd., Knoxfield, Australia). For details of the instrument, see Müller et al. (2011). Absorption and interpolated scattering measurements at 637 nm wavelength were used to calculate the single scattering albedo of dry aerosol particles, ω_0, at this wavelength, which is defined as the ratio of scattering to extinction coefficients (extinction = scattering + absorption). Strong BB and FF influenced observations were defined as periods when M_{BCe} exceeded the 75th percentile (0.27 µg m^{-3}) and the absorption wavelength dependence was above or within the BC-only regime, respectively. A detailed discussion on the absorption wavelength dependence~~study of aerosol optical properties measured at the ATTO site~~ can be found in Saturno et al. (2017).

An aerosol chemical speciation monitor (ACSM) (Aerodyne Research Inc., Billerica, USA) was used to measure online non-refractory aerosol chemical composition (Carbone et al., 2018)~~7~~). These measurements started in February 2014 at the ATTO site. The technique resolves the sub-micron aerosol

chemical species in the following categories: Organics, sulfate, nitrate, ammonium and chloride (Ng et al., 2011). In this paper, we only use organics and sulfate mass concentration data, M_{org} and $M_{sulfate}$, respectively.

Cloud condensation nuclei (CCN) number concentrations, N_{CCN}, were measured with a CCN counter (CCNC, model CCN-100, Droplet Measurement Technologies, Longmont~~Boulder~~, USA), which was deployed at the ATTO site starting in March 2014. The instrument scanned over a range of different supersaturations and particle diameters; more details can be found elsewhere (M. L. Pöhlker et al., 2016). The hygroscopicity parameter, κ, retrieved for a CCN activation ratio of 50 % is used in this study. Condensation nuclei number concentrations (> 5~~10~~ nm), N_{CN}, were measured with a condensation particle counter (CPC, model 5412, Grimm Aerosol Technik, Germany~~3022A, TSI, USA~~).

2.2 Airborne *in-situ* measurements

Chemical species of sub-micron aerosol particles were measured using a compact time-of-flight aerosol mass spectrometer (C-ToF-AMS) installed on board of the German High-Altitude and Long Range Research Aircraft (HALO, http://www.halo.dlr.de, last access:~~visited~~ 13 September 2017), a modified business jet G550 (Gulfstream, Savannah, USA). The C-ToF-MS details are presented in ~~elsewhere (~~Drewnick et al. (~~,~~2005) and Schulz et al. (2018). A single-particle soot photometer (SP2, Droplet Measurement Technologies, Longmont, USA) was used to measure rBC mass concentration. The measurements took place between 6 September and 1 October 2014, during the "Aerosol, Cloud, Precipitation, and Radiation Interactions and Dynamics of Convective Cloud Systems" (ACRIDICON) - "Cloud Processes of the Main Precipitation Systems in Brazil: A Contribution to Cloud Resolving Modeling and to the GPM (Global Precipitation Measurement)" (CHUVA) campaign over the Amazon rain forest (Machado et al., 2014). More details on the flight trajectories and instrumentation can be found in ~~elsewhere (~~Wendisch et al. (~~,~~2016). In this study, only data up to 7 km altitude have been used.

2.3 Air mass trajectories

To investigate the probability of the volcanic sulfate plume reaching the ATTO site, trajectories were calculated using the National Oceanic and Atmospheric Administration (NOAA) hybrid single-particle

Lagrangian integrated trajectory HYSPLIT model (Draxler and Hess, 1997, 1998; Stein et al., 2015). NOAA Global Data Assimilation System (GDAS) (Kleist et al., 2009) data at 1° × 1° resolution were used as meteorological input for HYSPLIT.

2.4 Satellite SO_2 vertical column density~~VCD~~ data

As one of the most abundant gases in a volcanic plume, SO_2 is often used as a tracer for volcanic emissions by a variety of spectroscopic remote sensing techniques. The strong characteristic absorption features in the UV spectral range allow the quantification of SO_2 using differential optical absorption spectroscopy (DOAS, see e.g., Platt and Stutz, 2008 and ~~;~~ Richter and Wagner, 2011), both from the ground (e.g., Bobrowski and Platt, 2007; Galle et al., 2003) and from space ((~~(~~e.g., Eisinger and Burrows~~,~~ 1998,~~;~~ Khokhar et al.~~,~~ 2005 and ~~;~~ Krueger ~~and J.~~, 1985)~~)~~).

The ozone monitoring instrument (OMI) on board of the National Aeronautics and Space Administration (NASA) Aura satellite, launched in 2004, detects backscattered solar radiation in the UV-visible range (Levelt et al., 2006). The polar-orbiting instrument crosses the equator at 13:30 local time. DOAS analysis of OMI spectra yields column densities of trace gases such as NO_2, SO_2, and HCHO with a spatial resolution of about 13 × 24 km^2 away from the swath edges. OMI's wide swath of 2600 km allowed daily global coverage until the first occurrence of the so-called row anomaly in June 2007, an instrumental problem that causes grievous radiance errors in up to half of OMI's ground pixels (Van Hoek and Claas, 2010). The row anomaly strongly affects the reliability of observations; therefore all affected pixels were removed from the data set prior to analysis.

The OMI SO_2 vertical column density (VCD) data presented in this paper were retrieved using ~~the~~ NASA's principal-component based algorithm with an a-priori vertical profile representative of a volcanic plume in the middle troposphere (TRM, Li et al., 2013, 2017). The assumption that the volcanic plume is located in the mid–troposphere is justified by the elevation of the volcano (3058 ~~k~~m), the strength of the eruption, and, particularly, the HYSPLIT trajectory analysis presented later in this paper. It is, however, important to note that the sensitivity of the satellite measurements depends systematically on plume altitude. Thus, the absolute values of the SO_2 VCD derived from the satellite

observations over- or underestimate the true values if the plume is located at a higher or lower altitude, respectively. Fortunately, this does not influence our study, as the focus of this paper is on the spatial pattern of the SO_2 plumes, and not on the SO_2 amount. The level-2 data were downloaded from: https://mirador.gsfc.nasa.gov/ (last access: 27 October 2017) and gridded to a regular, 0.1° × 0.1° grid for easily handling.

~~The level-2 data were downloaded from: https://mirador.gsfc.nasa.gov/ (last visited 27 October 2017) and gridded to a regular, 0.1° × 0.1° grid for easily handling.~~

~~Multi-year OMI SO_2 VCD daily averages from 11° S to 17° N are summarized in Fig. S1 as a function of time and longitude. The figure shows observations during the ATTO measurement period (March 2012 to July 2017) and a snapshot of September 2014. Given that the Nyamuragira and Nyiragongo volcanoes are so close to each other (within ~15 km), their emissions detected by remote sensors are often treated as a paired source (Carn et al., 2017). Hereafter, the term "Nyamuragira" refers to the couple Nyamuragira-Nyiragongo in this text. Time series of area-averaged OMI SO_2 observations are shown in Fig. 2a. Nyamuragira produced frequent intensive SO_2 emission events especially from 2012 to the end of 2015. The area where the average was calculated (Fig. 2b) corresponds to approximately 200,000 km^2 around Nyamuragira. Emissions from Nyamuragira were often transported westward, as can be observed in HYSPLIT forward trajectories calculated for 2014 (Fig. S2).~~

3 Results and discussion

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 M. L. 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. All these values and also measurements over the South Atlantic Ocean (Huang et al., 2018) were well below the $M_{sulfate}$ median values measured at ATTO

between 21 and 30 September 2014 (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.

3.1 Satellite measurements and trajectory analysis of the volcanic plume

Given that the Nyamuragira and Nyiragongo volcanoes are so close to each other (within ~15 km), their emissions detected by remote sensors are often treated as a paired source (Carn et al., 2017). Hereafter, the term "Nyamuragira" refers to the couple Nyamuragira-Nyiragongo in this text. Time series of area-averaged OMI SO_2 observations are shown in Fig. 3a. The area where the average was calculated (Fig. 3b) corresponds to approximately 200,000 km^2 around the volcanoes. 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). Multi-year OMI SO_2 VCD daily averages from 11° S to 17° N are summarized in Fig. S1 as a function of time and longitude. The figure shows observations during the ATTO measurement period (March 2012 to July 2017) and a snapshot of September 2014. According to Barrière et al. (2017), the emissions from June to October 2014 where mostly due to lava fountaining activity in Nyamuragira, characterized by strong tremors. The high emission activity of Nyamuragira at the end of 2014 was also observed by in-situ measurements from November 2014 to October 2015, when SO_2 emission fluxes reached up to 12.7×10^6 kg day^{-1} from Nyamuragira (Bobrowski et al., 2017). These emissions were often transported westward, as can be observed in HYSPLIT forward trajectories calculated for 2014 (Fig. S2).

Fortunately, during the high emission period mentioned above, specifically in September – October 2014, ground and airborne measurement campaigns took place in the Amazon Basin (Andreae et al., 2015; Wendisch et al., 2016; Martin et al., 2017). Satellite SO_2 VCD observations over central Africa and the Atlantic Ocean were examined during this period in order to precisely identify the plume trajectory. A map of gridded OMI SO_2 TRM VCD observations from 7 to 17 September 2014, is available as supplementary material (Fig. S3). Two important emission events were observed at the Nyamuragira location, one on 7 September and another on 12 September. The latter exhibits a clear westward transport of the SO_2 plume starting from 13 to 17 September. Figure 4 shows SO_2 VCD observations during this period within the region between 20 W – 30 E, and 15 S – 5 N with SO_2 VCD larger than 2.5×10^{16} molecules cm^{-2}. Several sets of trajectory calculations were performed. First, three to seven starting locations were selected within the SO_2 plumes detected by OMI on 12–17 September 2014. At each location, 15-day (360 hours) forward trajectories were started at the time of the satellite overpass (11 – 14 UTC) at seven altitudes spaced equally between 1 and 7 km. The resulting trajectories initialized at 4 km altitude on 13 September are in best agreement with the satellite data and are shown in Fig. 4. All starting parameters were systematically varied and very consistent patterns were found (see Fig. S4). The trajectories are superimposed on the map presenting all SO_2 plumes detected by OMI between 12 and 17 September. Trajectories started within the leading edge of the plume are in good agreement with the OMI data, as after 24 hours (second marker) both trajectories are located within the plume detected on 14 September, and after 48 hours (third marker) both trajectories are located within the plume detected on 15 September.

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 dispersive 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 SO_2 plume for the initial days, and then calculate forward trajectories from the observed location of the plume.

The two southernmost trajectories in Fig. 4 make a sharp turn after 15 September, which is in agreement with the observed pattern, although there is no longer an exact match with the respective OMI observations (in red and maroon). This discrepancy may be due to inaccuracy of the individual trajectories, or the SO_2 plumes might have been below OMI's detection limit. 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.

~~The most important activity at the Nyamuragira volcano since 2011 occurred in September 2014 (Global Volcanism Program, 2017), coinciding with ground and airborne measurement campaigns in the Amazon Basin (Andreae et al., 2015; Wendisch et al., 2016; Martin et al., 2017). Satellite SO_2 VCD observations over central Africa and the Atlantic Ocean were examined during this period in order to precisely identify the volcanic eruptive period and the plume trajectory. A map of gridded OMI SO_2 TRM VCD observations from 7 to 17 September 2014, is available as supplementary material (Fig. S3). Two important emission events were observed at the Nyamuragira location, one on 7 September and another on 12 September. The latter one exhibits a clear westward transport of the SO_2 plume starting from 13 to 17 September. Figure 3 shows SO_2 VCD observations during this period within the region between 20 W – 30 E, and 15 S – 5 N with SO_2 VCD larger than 2.5×10^{16} molecules cm^{-2}. Several sets of trajectory calculations were performed. First, three to seven starting locations were selected within the SO_2 plumes detected by OMI on 12–17 September 2014. At each location, 15-day (360 hours)~~

~~forward trajectories were started at the time of the satellite overpass (11 – 14 UTC) at seven altitudes spaced equally between 1 and 7 km. The resulting trajectories initialized at 4 km altitude on 13 September are in best agreement with the satellite data and are shown in Fig. 3. All starting parameters were systematically varied and very consistent patterns were found. The trajectories are superimposed on the map presenting all SO_2 plumes detected by OMI between 12 and 17 September. Trajectories started within the leading edge of the plume are in good agreement with the OMI data, as after 24 hours (second marker) both trajectories are located within the plume detected on 14 September, and after 48 hours (third marker) both trajectories are located within the plume detected on 15 September. The two southernmost trajectories make a sharp turn after 15 September, which is in agreement with the observed pattern, although there is no longer an exact match with the respective OMI observations (in red and maroon). This discrepancy may be due to inaccuracy of the individual trajectories, or the SO_2 plumes might have been below OMI's detection limit. 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.~~

In addition to the plume forward trajectory analysis, backward trajectories initiated at the ATTO site at an altitude of 300 m (approximately 170 m above ground) were calculated for 360 hours. These trajectories were initiated every hour from 20 September 0:00 UTC up to 30 September 23:00 UTC. The results, presented as a trajectory density plot in Fig. 5~~4~~, indicate that although during this time period essentially all air masses come from southern Africa, they usually come from further south. Nevertheless, a significant number of trajectories originates close to the volcano and its plume.

3.2 Airborne aerosol observation~~s of the volcanogenic aerosol particles~~

Enhanced sulfate aerosol mass concentrations were observed above 3 km height over the Amazon Basin during the ACRIDICON-CHUVA campaign, which were substantially greater than the concentrations measured at lower altitudes for some flights. A map including all airborne observations on the different flights can be found in the supplementary material (Fig. S5). However, given the multiple sulfate aerosol sources, sulfate itself cannot be used as a tracer of volcanic emissions. In order to distinguish the

volcanogenic sulfate from other aerosol sources like BB, which is important during this time of the year, we examined the $M_{sulfate}$ vertical profiles together with their sulfate-to-OA mass ratio ($M_{sulfate}$ / M_{org}). A list of the ACRIDICON-CHUVA flights and $M_{sulfate}$ vertical profiles are presented as supplementary information in Table S1 and Fig. S6, respectively.

From the different airborne observations, the elevated plumes observed on flights AC14 and AC17 exhibited median sulfate-to-OA mass ratios larger than 1.0, indicating strong volcanogenic influence. The $M_{sulfate}$ vertical profile measured on 21 September 2014 (AC14) is presented in Fig. 6. The observations show a sulfate-enhanced layer between 4 and 5 km height, which was also observed during flight AC17 on 27 September 2014. The average $M_{sulfate}$ observed during flight AC14 was 1.1 ± 0.5 µg m^{-3} between 3 and 6 km height. This sulfate-enhanced layer exhibits a sulfate-to-OA ratio generally larger than 1. It can be distinguished from lower layers, below 3 km height, which are characteristically rich in OA due to BB and biogenic emissions. Usually, BB aerosol particles have been shown to have higher OA mass concentrations than other aerosol particles (McNaughton et al., 2011; Saturno et al., 2017). 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.

Backward trajectories were calculated from several points along the flight paths. Figure 7 shows backward trajectories started at nine points along the AC14 flight track, where sulfate-to-OA ratios larger (colored lines) or smaller than 1 (gray lines) were detected; the flight and aerosol data measured at each point are presented in Table 1. Figure 7 clearly shows that the colored trajectories, with one exception, initialized within the sulfate plume originate from central Africa, whereas the gray trajectories, started outside of the sulfate plume, appear to originate from South America or from more southern regions over or across the Atlantic Ocean. The air mass trajectory analysis supports our hypothesis that the AC14 observations were likely the result of probing the volcanic plume emitted on 7 September, the first one detected by OMI (see Fig. S3). For flight AC17 a similar pattern is observed, with three out of four (colored) trajectories started within the sulfate plume originating from central

Africa and half of the other trajectories clearly coming from regions more to the South (see supplement, Fig. S7 and Table S2).

~~Enhanced sulfate aerosol mass concentrations were observed above 3 km height over the Amazon Basin during the ACRIDICON-CHUVA campaign compared to lower altitudes. A map including all airborne observations on the different flights can be found in the supplementary material (Fig. S4). However, given the multiple sulfate aerosol sources, sulfate itself can not be used as a tracer of volcanic emissions. In order to distinguish the volcanogenic sulfate from additional aerosol sources like BB, which is important during this time of the year, we examined the $M_{sulfate}$ vertical profiles together with their sulfate-to-OA mass ratio ($M_{sulfate}$ / M_{org}). A list of the ACRIDICON-CHUVA flights and $M_{sulfate}$ vertical profiles are presented as supplementary information in Table S1 and Fig. S5, respectively. From the different airborne observations, flight AC14 showed the highest sulfate-to-OA mass ratio, indicating the strongest volcanogenic influence. The $M_{sulfate}$ vertical profile measured on 21 September 2014 (AC14) is presented in Fig. 5. The observations show a sulfate-enhanced layer between 4 and 5 km height. The average $M_{sulfate}$ observed during flight AC14 was 1.1 ± 0.5 µg m^{-3} between 3 and 6 km height. This sulfate-enhanced layer exhibits a sulfate-to-OA ratio generally larger than 1. It can be distinguished from lower layers, below 3 km height, which are characteristically rich in OA due to BB and biogenic emissions. Usually, BB aerosol particles have been shown to have higher OA mass concentrations than other aerosol particles (McNaughton et al., 2011; Saturno et al., 2017). Therefore, the high sulfate-to-OA ratio is an indication of the volcanic origin of the probed aerosol. Backward trajectories were calculated from several points along the flight paths. Figure 6 shows backward trajectories started at nine points along the AC14 flight track where sulfate-to-OA ratios larger (colored lines) or smaller than 1 (gray lines) were detected; the flight and aerosol data measured at each point are presented in Table 1. Figure 6 clearly shows that the (colored) trajectories, with one exception, initialized within the sulfate plume originate from central Africa, whereas the gray trajectories, started outside of the sulfate plume, appear to originate from South America or from more southern regions over or across the Atlantic Ocean. The air mass trajectory analysis indicates that the AC14 observations were likely the result of probing the volcanic plume emitted on 7 September, the first one detected by OMI (see Fig. S3). For flight AC17 a similar pattern is observed, with three out of four (colored)~~

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Figures 4, ~~5~~3, ~~4~~, and ~~7~~6 show that the trajectories agree well, but not perfectly with the ground-based, airborne, and satellite measurements, which is mainly caused by the uncertainty of such long trajectories. Nevertheless, the fact that forward and backward trajectories calculated from various starting points and times agree on the sulfate source is a further ~~a strong~~ indication that the sulfate plumes observed at and near ATTO originate from the Nyamuragira volcano. Combined with the westward transport pattern derived from SO_2 satellite data and the lack of an alternative strong sulfate source makes this a quite convincing case.

3.3 Ground-based aerosol observations

~~3.3 Volcanic emission effects on the aerosol particle properties~~

The arrival of the African volcanic emissions over the Amazon rain forest is reflected in~~affected~~ the aerosol physical and chemical properties measured at the ATTO site. The most evident effect was the significant increase in Msulfate. The 90th percentile of Msulfate measured at the ATTO site during the dry season 2014 was used as a threshold to define the volcanic influence event (Nya2014) as the time when this threshold was exceeded. By this criterion, the Nya2014 event spanned from 21 September 2014 at 02:00 UTC to 1 October 2014 at 01:00 UTC. Figure 8~~7~~ shows different aerosol parameters measured before, during and after the Nya2014 event. The NCN, shown in Fig. 8~~7~~a, did not vary greatly from the values typical of the season (M. L. Pöhlker et al., 2016). The average NCN during the dry season in 2014 was (1.3 ± 0.6) × 103 particles cm-3, whereas during the Nya2014 event, there were three peaks lasting for few hours with particle number concentrations higher than 3.0 × 103 particles cm-3 on 27, 29, and 30 September 2014, likely related to BB plumes (M. L. Pöhlker et al., 2017).

On the other hand, d~~D~~uring the Nya2014 period, $M_{sulfate}$ averaged 1.7 ± 0.6 μg m^{-3}, which was significantly above the dry season 2014 average of 0.7 ± 0.3 μg m^{-3}, see Fig. 8~~7~~b. The highest $M_{sulfate}$ value observed at the ATTO site was 3.6 μg m^{-3} (hourly mean) on 26 September 2014. For comparison, during the SAMBBA campaign in southern Amazonia, $M_{sulfate}$ barely exceeded 1.0 μg m^{-3}, despite

organics nearly reaching 100 μg m^{-3}, M_{BCe} of 5 μg m^{-3} and N_{CN} above 25 × 10^3 particles cm^{-3} during the peak of biomass burning (Brito et al., 2014). A long-term measurement study, also conducted in southern Amazonia, reported $M_{sulfate}$ of 1.1 ± 0.7 μg m^{-3} during the dry season, with a maximum of 3.3 μg m^{-3} for aerosol particles with mobility diameters smaller than 2 μm (Artaxo et al., 2002). It is important to note that the observations by Artaxo et al.~~'s~~ (2002) ~~sulfate observations~~ were done under strong BB influence with average elemental carbon (EC) mass concentrations, M_{EC}, of 3.8 ± 4.2 μg m^{-3}, with a maximum of 25 μg m^{-3}. In contrast, the BC_e measurements at the ATTO site during the Nya2014 event had an average of 0.4 ± 0.1 μg m^{-3}, with a maximum of 0.8 μg m^{-3}, indicating that the BB influence was relatively weak during the period of interest, with some short (few hours) spikes due to the influence of near-by fire events, see Fig. 8~~7~~b. At a sampling site impacted by Manaus emissions, the sub-micron $M_{sulfate}$ was about 0.2 μg m^{-3} during the wet season, rarely exceeding 0.6 μg m^{-3} (de Sá et al., 2017). Therefore, even considering a range of pollution sources, our measurements at ATTO during the Nya2014 event are the highest sub-micron sulfate concentration ever reported in the Amazon Basin; see Martin et al. (2010) for a summary of wet and dry season aerosol observations. For comparison, the ACRIDICON-CHUVA airborne measurements are also included in Fig. 8~~7~~b. The $M_{sulfate}$ measured on flight AC14 was significantly enhanced between 3 to 6 km altitude, reaching a median of 1.0 μg m^{-3} and a 75th percentile of 1.6 μg m^{-3}. Previous aircraft measurements during the SAMBBA campaign reported a $M_{sulfate}$ flight average of 0.48 μg m^{-3} (Allan et al., 2014).

The increased $M_{sulfate}$ period was accompanied by an enhanced sulfate-to-OA mass ratio, according to the ATTO observations (Fig. 8~~7~~c). The Ny2014 sulfate-to-OA average over about 10 days was 0.38 ± 0.09, significantly higher than the dry season average of 0.18 ± 0.15. During some BB pulses, decreased sulfate-to-OA ratios were observed, but the whole Nya2014 period was exceptionally high compared to the typical dry season conditions. The sulfate-to-OA values measured at ground level were usually lower than the airborne values observed between 3 and 6 km height because the OA sources (BB and biogenic emissions) are located at ground level and the LRT sulfate that arrives at higher altitudes is diluted upon vertical mixing. The possibility of FF~~fossil-fuel (FF)~~ burning was ruled out as an important sulfur source during the event discussed here because of the particularly high dry-aerosol ω_0 measured during the event (0.89 ± 0.04), as can be observed in the color code data in Fig. 8~~7~~c.

Usually FF emissions, rich in BC, present characteristically low ω_0 (0.2 – 0.3) (Bond and Bergstrom, 2006) and its addition would have lowered the value of ω_0. Instead, an increase in ω_0 was observed to values higher than 0.90 during the period of maximum $M_{sulfate}$ (26 – 27 September 2014).

The effects of the volcanic sulfur plume on the aerosol hygroscopicity was explored by analyzing the κ values measured at different supersaturations. Higher κ values were measured for the accumulation mode aerosol (particles with diameter greater than 100 nm) (Fig. 8d; note the color-coded particle activation diameter, D_a). During the Nya2014 event, the κ values increased significantly, especially when the maximum $M_{sulfate}$ was observed. For example, the average κ for a supersaturation of 0.10 % (D_a = 167 – 179 nm, 25th and 75th percentile, respectively) was 0.26 ± 0.04 during the Nya2014 event, with a maximum of 0.36. A similar increase was observed at Jungfraujoch (Switzerland) under the influence of the Eyjafjallajökull volcanic emissions in 2010 (Bukowiecki et al., 2011). In that case the Aitken mode particles (10 to 50 nm diameter) were more strongly affected than the accumulation mode particles because of the shorter ageing period of the sulfate particles. The Nya2014 κ average was slightly higher than the 2014 dry season average of 0.21 ± 0.03 for 0.10 % supersaturation (excluding the volcanic event), and significantly higher than a strong BB event average of 0.18 ± 0.01 for 0.10 % supersaturation, whose high OA content (sulfate-to-OA < 0.04) caused a significant drop in κ (M. L. Pöhlker et al., 2017).

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Summary and conclusions

Satellite SO_2 observations showed two explosive events at the Nyamuragira volcano on 7 and 12 September 2014. These emissions were observed to be transported over the South Atlantic Ocean. Modelled air mass trajectories, starting at the satellite-observed plume locations, showed that the plume was transported towards South America, eventually reaching the region of the Amazon Tall Tower Observatory in central Amazonia. Airborne observations during the ACRIDICON-CHUVA campaign probed a sulfate-enhanced layer between 4 and 5 km height on 21 September 2014 (flight AC14). Additionally, this layer exhibited very low rBC concentrations and an increased sulfate-to-OA mass ratio with medians higher than 1 for measurements between 3 and 6 km height.

The ground-based $M_{sulfate}$ measured at the ATTO site reached hourly mean levels up to 3.6 µg m^{-3} on 26 September 2014, the highest values ever reported in the Amazon Basin. The sulfate-to-OA mass ratio increased from a dry-season average of 0.18 ± 0.15 to an average of 0.38 ± 0.09 upon volcanic influence, which spanned over about 10 days. Increased sulfate-to-OA and single scattering albedo (ω_0) suggest a rather low influence of BB and FF sources. In terms of aerosol hygroscopicity, the values of κ (for 0.10 % supersaturation) measured during the volcanic event reached an average of 0.26 ± 0.04 (and a maximum of 0.36), the highest levels observed under dry season conditions (average of 0.21 ± 0.03).

~~Satellite SO_2 observations showed two explosive events at the Nyamuragira volcano on 7 and 12 September 2014. These emissions were observed to be transported over the South Atlantic Ocean. Air mass trajectory modeling from the plume location showed that the plume was transported towards South America, likely over the ATTO site and its surroundings in central Amazonia. Airborne observations during the ACRIDICON-CHUVA campaign showed a sulfate-enhanced layer between 4 and 5 km height on 21 September 2014 (flight AC14). Additionally, this layer exhibited an increased sulfate-to-OA mass ratio with medians higher than 1 for measurements between 3 and 6 km height.~~

~~The ground-based $M_{sulfate}$ measured at the ATTO site reached an hourly mean of 3.6 µg m^{-3} on 26 September 2014, the highest values ever reported in the Amazon Basin. The sulfate-to-OA mass ratio increased from a dry-season average of 0.18 ± 0.15 to 0.38 ± 0.09 during the volcanic influence event, which spanned for a period of about 10 days. Increased sulfate-to-OA and single scattering albedo (ω_0)~~

~~were assumed as an indication of the low influence of BB and FF sources, respectively. In terms of aerosol hygroscopicity, the values of κ (for 0.10 % supersaturation) measured during the volcanic event reached an average of 0.26 ± 0.04 (and a maximum of 0.36), slightly higher than the dry season average of 0.21 ± 0.03.~~

The evidence presented here shows one particular event of volcanic SO_2 emissions in Africa influencing the aerosol particles' chemical composition, hygroscopicity, and optical properties in the Amazon Basin. Therefore, our study indicates that these emissions and their transatlantic transport could potentially affect the Amazonian cloud microphysical properties. However, the extent and relevance of the episodic volcanic influence on the Amazonian atmosphere would require more extensive studies. Beyond the effects and implications of this particular event, the results of our study represent a reference case of the dynamics and conditions of transatlantic aerosol transport from southern Africa to South America. This could help to understand the inter-continental advection of other aerosol species, such as combustion aerosol particles that are more difficult to trace.

Data availability. The data presented in this paper can be accessed via e-mail request to Jorge Saturno (j.saturno@mpic.de) or Christopher Pöhlker (c.pohlker@mpic.de). OMI data are available online at https://disc.gsfc.nasa.gov/.

Competing interests. The authors declare that they have no conflict of interest.

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