

African volcanic emissions influencing atmospheric aerosols 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 observational evidence for the influence of emissions from the Nyamuragira-Nyiragongo volcanoes in Africa on the Amazonian aerosol properties and atmospheric composition during September 2014. Comprehensive ground-based

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and airborne aerosol measurements together with satellite observations are used to investigate the volcanic event. Under the volcanic influence, hourly mean sulfate mass concentrations in the sub-micron size range reached up to $3.6 \mu\text{g m}^{-3}$ at the Amazon Tall Tower Observatory, the highest value ever reported in the Amazon region. The substantial sulfate injection increased the aerosol
40 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
45 forest.

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
50 condensation nuclei (CCN) (Stevens and Feingold, 2009). Anthropogenic SO_2 emissions have increased over the 20th century to a maximum ($120 \text{ Tg SO}_2 \text{ year}^{-1}$) 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
55 annual SO_2 emissions (Textor et al., 2004; Yang et al., 2017). The emitted SO_2 is oxidized in the atmosphere to gaseous sulfuric acid, which is 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. Observational evidence of drastic changes
60 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). Some examples are the

Pinatubo eruption in 1991 that reached the stratosphere (> 10 km height) (Kirchner et al., 1999), 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), and the eruptive SO_2 emissions of the Calbuco volcano in Chile, which reached southern Africa after being transported over the Atlantic Ocean (Sangeetha et al., 2018). 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 strongest passively degassing volcanoes worldwide between 2004 and 2014, with around 150 days of satellite-detected degassing in 2014 (Carn et al., 2016). The strong passive degassing activity of Nyamuragira, which started in 2011 and culminated in the formation of a lava lake in late 2014 (Bobrowski et al., 2017; Campion, 2014) led to two orders of magnitude higher SO_2 mass emissions from the Nyiragongo-Nyamuragira complex compared to the 2014-2017 average (Barrière et al., 2017). The emissions from Nyamuragira in 2014 have been estimated at ca. 2.3 Tg SO_2 year $^{-1}$ (Fioletov et al., 2016).

In the Amazon rain forest, biogenic sulfate aerosol is sustained by oceanic DMS emissions 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

90 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 (Andreae et al., 1988). These haze layers have been linked to LRT of
95 aerosol particles from Africa (Holanda et al., 2018). A modeled global sulfate source attribution study showed that peak sulfate concentrations in southern Africa occur between June and August. For this three-month period, estimated emissions from combustion sources were 0.81 Tg S, decreasing 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
100 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).

Northeasterly and southeasterly trade winds [north and south of the inter-tropical convergence zone (ITCZ)] 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
105 transport of African smoke from southern Africa savanna and shrubland fires is an important source of aerosol in addition to the regional emissions (Andreae et al., 1994). Even though the potential impact of transatlantic transport of African sulfur emissions has been suggested before based on transport models (Yang et al., 2017), no ground-based evidence has been reported previously in the literature concerning the impact of African volcanic sources.

110 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 SO₂ emissions were oxidized to particulate sulfate, which reached the central Amazon rain
115 forest and influenced aerosol physical and chemical properties significantly.

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)],
120 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.
125 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
130 (Moran-Zuloaga et al., 2017; M. L. Pöhlker et al., 2016; Saturno et al., 2017).

Equivalent black carbon (BC_e) mass concentrations, M_{BC_e} , 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
135 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, 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
140 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 ($0.27 \mu\text{g m}^{-3}$) and the absorption wavelength dependence was above or within the BC-
145 only regime, respectively. A detailed discussion on the absorption wavelength dependence 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). These measurements started in February 2014 at the ATTO site. The technique resolves the sub-micron
150 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, USA), which was deployed
155 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 ($> 5 \text{ nm}$), N_{CN} , were measured with a condensation particle counter (CPC, model 5412, Grimm Aerosol Technik, Germany).

160 **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: 13 September 2017), a modified business jet G550 (Gulfstream, Savannah, USA). The C-ToF-MS details are presented in Drewnick et al. (2005)
165 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
170 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 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
175 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 $1^\circ \times 1^\circ$ resolution were
used as meteorological input for HYSPLIT.

2.4 Satellite SO₂ vertical column density data

180 As one of the most abundant gases in a volcanic plume, SO₂ 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 SO₂ 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
185 1998, Khokhar et al. 2005 and Krueger, 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 NO₂, SO₂, and
190 HCHO with a spatial resolution of about $13 \times 24 \text{ km}^2$ 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.

195 The OMI SO₂ vertical column density (VCD) data presented in this paper were retrieved using 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 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 SO₂ 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 SO₂ plumes, and not on the SO₂ amount. The level-2 data were downloaded from:
205 <https://mirador.gsfc.nasa.gov/> (last access: 27 October 2017) and gridded to a regular, 0.1° × 0.1° grid for easily handling.

3 Results and discussion

The Amazonian dry season (August – November) M_{sulfate} hourly-means median over 3 years of measurements at the ATTO site was 0.60 $\mu\text{g m}^{-3}$ (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 $\mu\text{g m}^{-3}$) and did not change significantly during FF combustion influence (median: 0.56 $\mu\text{g m}^{-3}$). 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 $\sim 0.44 \mu\text{g m}^{-3}$ using energy-dispersive X-ray analysis and M. L. Pöhlker et al. (2017) reported a M_{sulfate} mean of 0.25 $\pm 0.19 \mu\text{g m}^{-3}$ 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 $\mu\text{g m}^{-3}$, see Fig. 2a). Note that the M_{sulfate} median corresponding to 21-30 September 2014 is higher than 99.3 % of the data measured during the dry seasons and the FF combustion influenced periods, and it is higher than at least 75 % of the data measured in the BB-influenced periods. Regarding the single scattering albedo, $\omega_{0,637}$, shown in
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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 and an air mass trajectory analysis are 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 SO₂ observations are shown in Fig. 3a. The area where the average was calculated (Fig. 3b) corresponds to approximately 200,000 km² around the volcanoes. Nyamuragira produced frequent intensive SO₂ emission events in 2014 with a mean emission of 14.4×10^6 kg SO₂ day⁻¹ (Barrière et al., 2017). Multi-year OMI SO₂ 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 were 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 SO₂ 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 SO₂ 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 SO₂ TRM VCD observations from 7 to 17 September 2014, is available as supplementary material (Fig. S3). Two 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 SO₂ plume starting from 13 to 17 September. Figure 4 shows SO₂ VCD observations during this period within the region between 20 W – 30 E, and 15 S – 5 N with SO₂ 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 SO₂ 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 SO₂ 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

275 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.

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 SO₂ 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 trajectory reached the direct vicinity of ATTO on 25 September at 1.8 km altitude, whereas another trajectory passed an area south of ATTO at an altitude of 1.5 km 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 (Andreae et al., 1988).

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, indicate that during this time period essentially all air masses come from southern Africa and a significant number of trajectories passes the area west of the volcano (e.g., 0-10° E, 0-10° S), where the volcanic plume was detected.

3.2 Airborne aerosol observations

Enhanced sulfate aerosol mass concentrations were observed above 3 km altitude 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 ($M_{\text{sulfate}} / M_{\text{org}}$). 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 (21 September 2014) and AC17 (27 September 2014) 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 $1.1 \pm 0.5 \mu\text{g m}^{-3}$ between 3 and 6 km height. This sulfate-enhanced layer exhibits the highest sulfate-to-OA ratio and 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}) (Holanda et al., 2018), 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 mass concentration) the most plausible explanation for the source of this sulfate plume is 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 (21 September 2014) 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

330 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).

Figures 4, 5, and 7 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
335 trajectories. 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. Combined with the westward transport pattern derived from SO₂ satellite data and the lack of an alternative strong sulfate source makes this a quite convincing case.

340 3.3 Ground-based aerosol observations

The arrival of the African volcanic emissions over the Amazon rain forest is reflected in the aerosol physical and chemical properties measured at the ATTO site. The most evident effect was the significant increase in M_{sulfate} . The 90th percentile of M_{sulfate} 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
345 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 shows different aerosol parameters measured before, during and after the Nya2014 event. The N_{CN} , shown in Fig. 8a, did not vary greatly from the values typical of the season (M. L. Pöhlker et al., 2016). The average N_{CN} during the dry season in 2014 was $(1.3 \pm 0.6) \times 10^3$ particles cm⁻³, whereas during the Nya2014 event, there were three peaks lasting
350 for few hours with particle number concentrations higher than 3000 particles cm⁻³ on 27, 29, and 30 September 2014, likely related to BB plumes (details in M. L. Pöhlker et al., 2017).

On the other hand, during the Nya2014 period, M_{sulfate} averaged $1.7 \pm 0.6 \mu\text{g m}^{-3}$, which was significantly above the dry season 2014 average of $0.7 \pm 0.3 \mu\text{g m}^{-3}$, see Fig. 8b. The highest M_{sulfate} value observed at the ATTO site was $3.6 \mu\text{g m}^{-3}$ (hourly mean) on 26 September 2014. For comparison,

355 during the SAMBBA campaign in southern Amazonia, M_{sulfate} barely exceeded $1.0 \mu\text{g m}^{-3}$, despite organics nearly reaching $100 \mu\text{g m}^{-3}$, M_{BCe} of $5 \mu\text{g m}^{-3}$ 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 $1.1 \pm 0.7 \mu\text{g m}^{-3}$ during the dry season, with a maximum of $3.3 \mu\text{g m}^{-3}$ for aerosol particles with diameters smaller than $2 \mu\text{m}$ (Artaxo et al., 2002). It is important to
360 note that the observations by Artaxo et al. (2002) were done under strong BB influence with average elemental carbon (EC) mass concentrations, M_{EC} , of $3.8 \pm 4.2 \mu\text{g m}^{-3}$, with a maximum of $25 \mu\text{g m}^{-3}$. In contrast, the BC_e measurements at the ATTO site during the Nya2014 event had an average of $0.4 \pm 0.1 \mu\text{g m}^{-3}$, with a maximum of $0.8 \mu\text{g m}^{-3}$, 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
365 Fig. 8b. At a sampling site impacted by Manaus emissions, the sub-micron M_{sulfate} was about $0.2 \mu\text{g m}^{-3}$ during the wet season, rarely exceeding $0.6 \mu\text{g m}^{-3}$ (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
370 airborne measurements are also included in Fig. 8b. The M_{sulfate} measured on flight AC14 was significantly enhanced between 3 to 6 km altitude, reaching a median of $1.0 \mu\text{g m}^{-3}$ and a 75th percentile of $1.6 \mu\text{g m}^{-3}$. Previous aircraft measurements during the SAMBBA campaign reported a M_{sulfate} flight average of $0.48 \mu\text{g m}^{-3}$ (Allan et al., 2014).

The increased M_{sulfate} period was accompanied by an enhanced sulfate-to-OA mass ratio, according to
375 the ATTO observations (Fig. 8c). The Nya2014 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
380 (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 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. 8c. 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. Similarly, the particle hygroscopicity was also increased 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).

Summary and conclusions

Satellite SO₂ observations showed two explosive events at the Nyamuragira volcano on 7 and 12 September 2014. These emissions were observed to be transported far 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.

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.

410 The ground-based M_{sulfate} measured at the ATTO site reached hourly mean levels up to $3.6 \mu\text{g m}^{-3}$ 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 415 (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).

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 420 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 425 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/>.

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

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Table 1. Measurements at points along track of the flight AC14 (21 September 2014) selected as starting points for backward trajectories presented in Fig. 7. Data points with sulfate-to-OA > 1 are emphasized by bold font.

Time (UTC)	Latitude [°N]	Longitude [°E]	Altitude [km]	M_{sulfate} [$\mu\text{g m}^{-3}$]	Sulfate-to-OA	Color in Fig. 7
15:14	-2.75	-60.34	1.61	1.2	0.5	Gray
16:19	-3.19	-60.21	4.50	1.0	1.1	Blue
16:36	-4.00	-59.50	4.50	3.0	2.9	Green
16:54	-5.20	-59.25	4.49	3.1	7.8	Red
17:11	-6.45	-58.98	6.45	0.6	0.8	Gray
17:46	-4.52	-59.40	6.43	0.5	0.8	Gray
19:40	-4.44	-59.41	7.68	1.2	1.0	Gray
21:41	-3.79	-59.55	4.79	1.8	2.2	Light blue
21:59	-3.04	-60.20	0.90	1.2	0.3	Gray

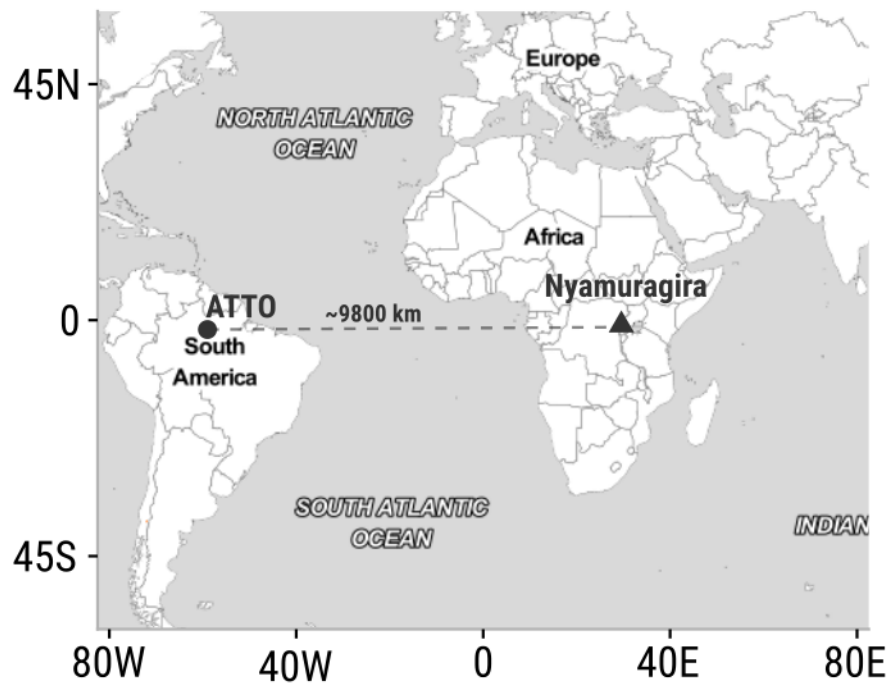


Figure 1. ATTO site and Nyamuragira volcano locations.

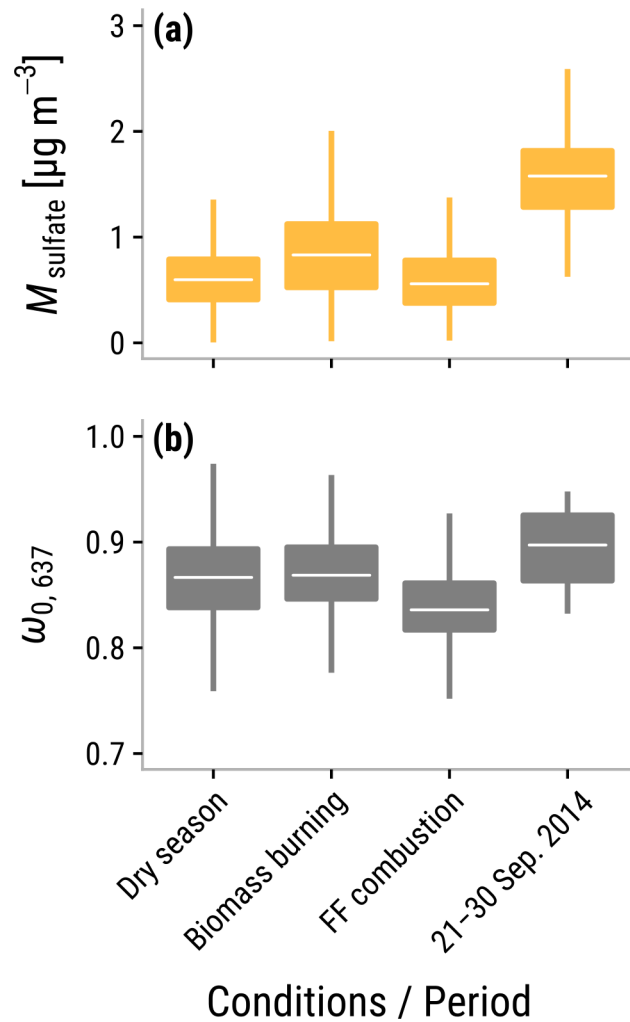


Figure 2. Box and whisker plots of hourly-mean (a) sulfate mass concentration, M_{sulfate} , and (b) single scattering albedo at 637 nm, $\omega_{0,637}$, during different periods and conditions including dry season average 2014 – 2016, biomass burning and fossil-fuel combustion influenced conditions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

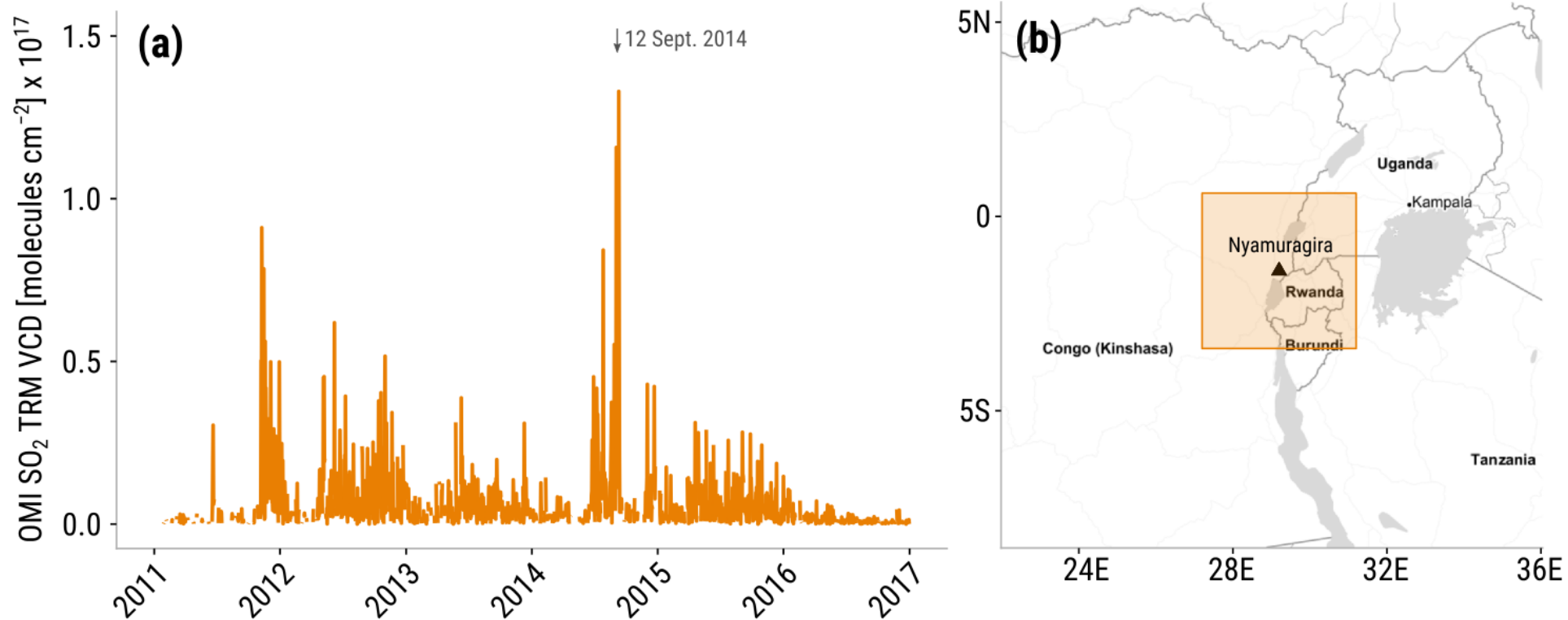


Figure 3. (a) Time series of daily-averaged OMI SO₂ TRM VCD observations corresponding to the averages over the area delimited by 27.2° E, 3.4° S, 31.2° E, and 0.6° N (b). Map of eastern Africa showing the averaging area (orange square). The location of Nyamuragira is represented by a black triangle.

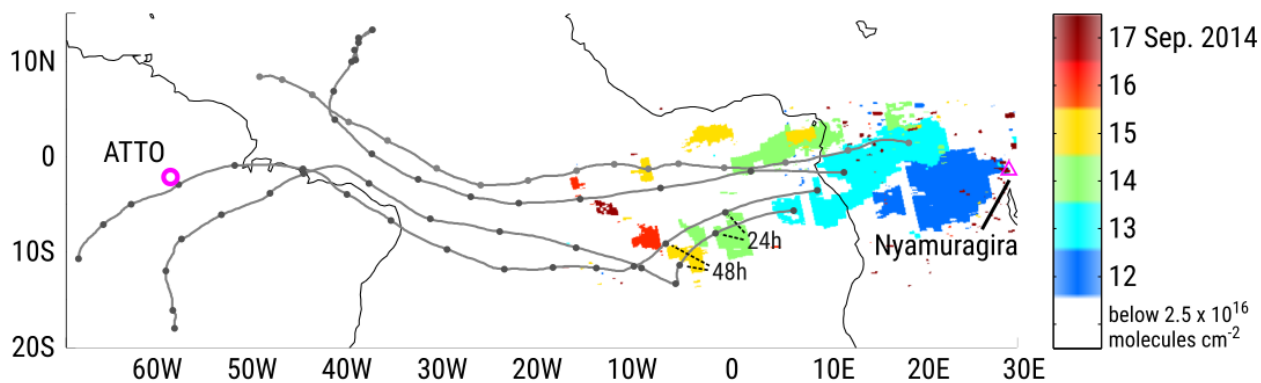


Figure 4. Map of SO₂ plumes with VCD > 2.5 × 10¹⁶ molecules cm⁻² color-coded by date of observation. 15-day forward trajectories started at 4 km (above mean sea level, a.m.s.l.) at four locations within the plume detected on 13 September 2014 (light blue) are indicated by black lines with markers at 24-hour intervals.

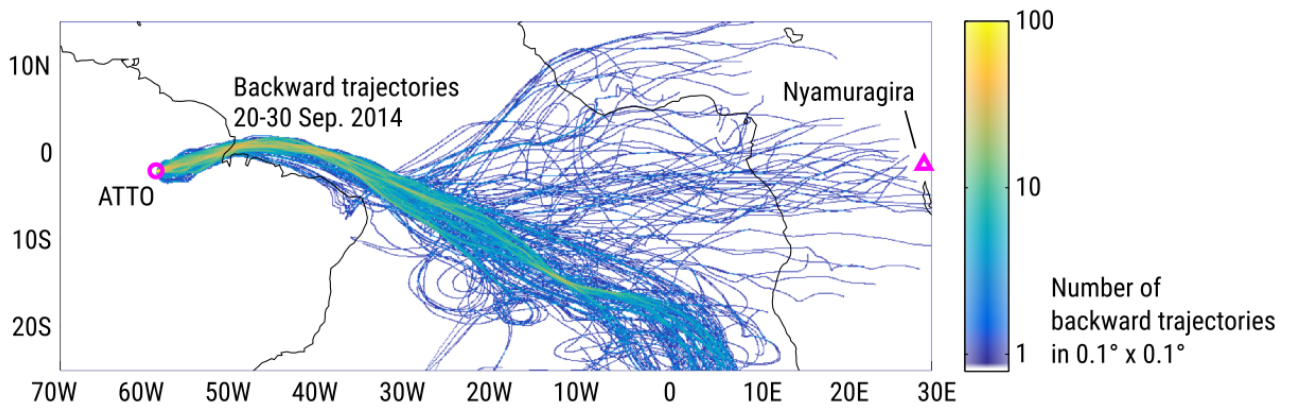


Figure 5. Density of 15-day backward trajectories started from ATTO at an altitude of 300 m a.m.s.l. on every hour starting at 0:00 UTC on 20 September 2014 up to 23:00 UTC on 30 September 2014.

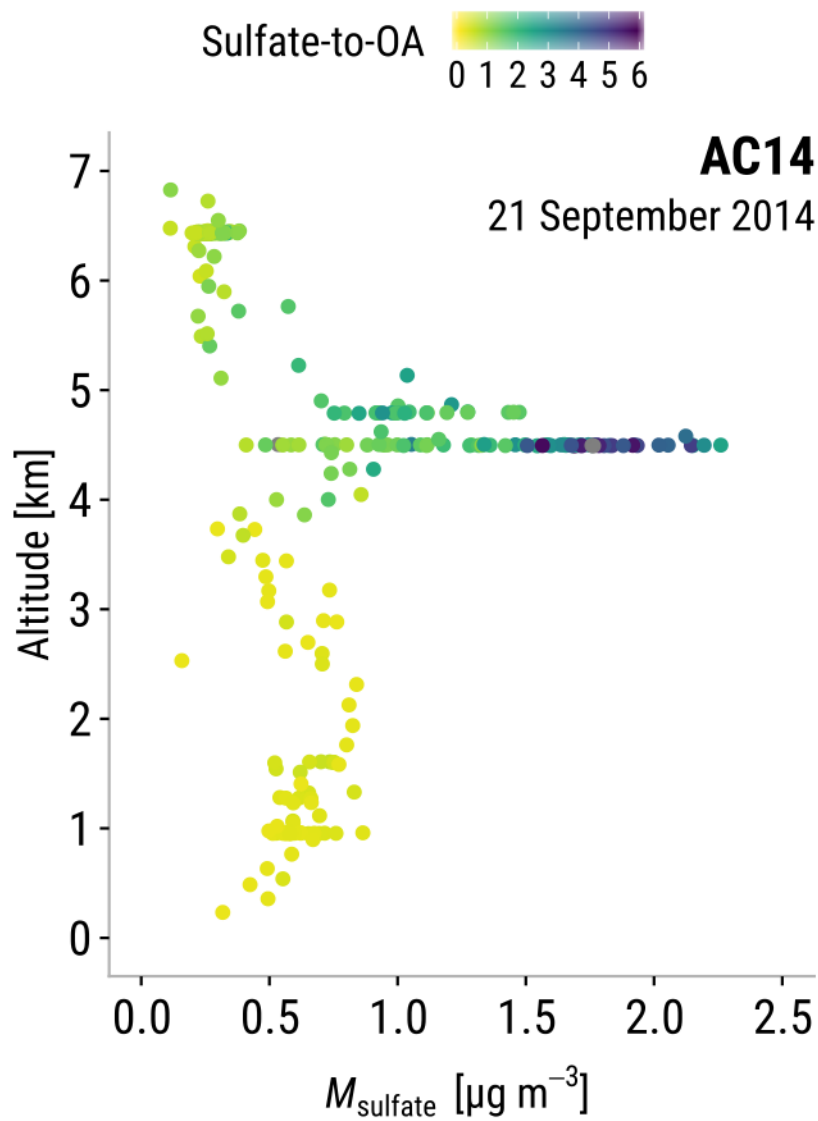


Figure 6. Sulfate layer observations over the Amazon rain forest. The figure shows the M_{sulfate} vertical profile observed during flight AC14 (21 September 2014). Color-coded sulfate-to-OA values are truncated at a maximum of 6.

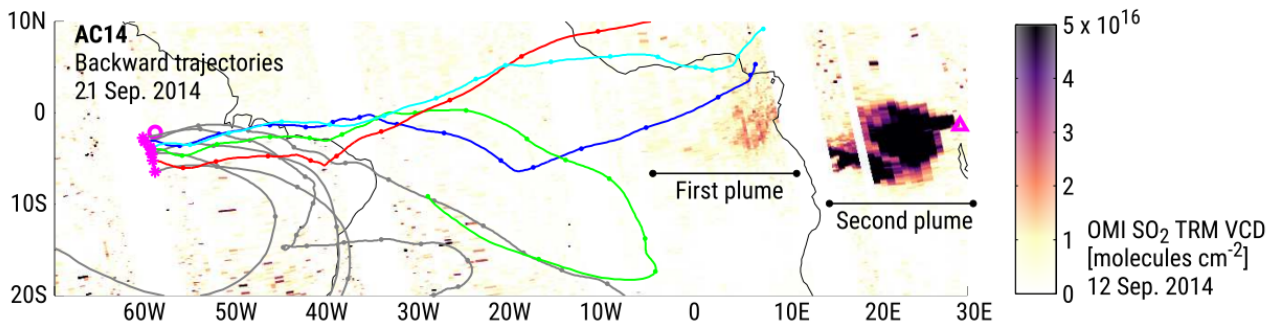


Figure 7. Map of gridded OMI SO₂ VCD, observed on 12 September 2014. Two plumes are shown on the map; the first one emitted on 7 September 2014, and the second one emitted on 12 September 2014. 15-day backward trajectories initialized at several points along track of the flight AC14 (21 September 2014) at flight altitude are shown as continuous lines with dots. Trajectories starting at points where sulfate-to-OA > 1 are shown in color (see Table 1 for details), all other trajectories are shown in gray; dots are placed at 24-hour intervals. The path of flight AC14 is marked in pink, with stars denoting the starting points of the backward trajectories. The locations of the ATTO site and Nyamuragira are marked with a pink circle and a triangle, respectively.

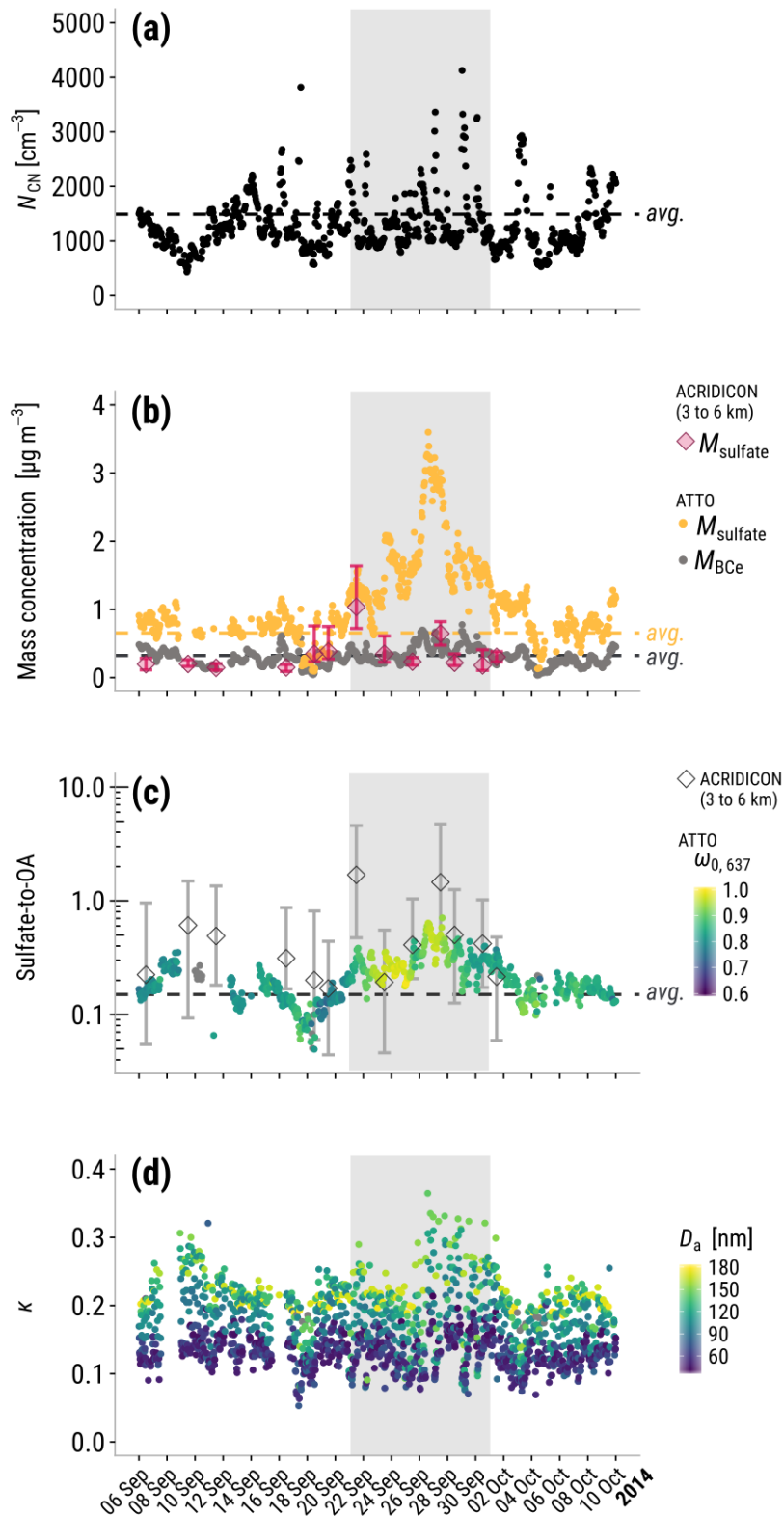


Figure 8. Different aerosol properties measured during the Nya2014 event (gray shaded area), These time series include ATTO hourly means of (a) aerosol particle number concentration, N_{CN} , (b) sulfate and BC_e mass concentration, (c) sulfate-to-OA mass ratio with color-coded aerosol particle single scattering albedo at 637 nm wavelength, and (d) hygroscopicity parameter, κ , original time resolution, with color-coded particle activation diameter. Dry season averages are shown as dashed lines. ACRIDICON-CHUVA flight medians and inter-quartile ranges from 3 to 6 km altitude are shown in (b) and (c).