

Interactive comment on “African volcanic emissions influencing atmospheric aerosol particles over the Amazon rain forest” by Jorge Saturno et al.

Anonymous Referee #2

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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

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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 prove 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 ?

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?

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

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SO₂ into the atmosphere?

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 ?

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.

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

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?

2- Surprisingly, another peak in sulfate-to-OA ($\gg 1$) is also observed on 27/09/2014

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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 ?

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 Nyamuragira-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.

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 explicitly distinguish the impacts of tropospheric vs. stratospheric eruptions, which are different and presently mixed in the current draft.

- As it is the topic of their paper, the authors should precisely focus on the atmospheric

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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)

- 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.

- 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.

- 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.

- 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 ?

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

- 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 ?

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

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