

Interactive comment on “Unified observation co-existing volcanic sulphur dioxide and sulphate aerosols using ground-based Fourier transform infrared spectroscopy” by Pasquale Sellitto et al.

Anonymous Referee #2

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The manuscript (MS) introduces a new method to simultaneously retrieve the co-existing volcanogenic SO₂ and sulfate aerosols (SA) from ground-based FTIR observations. Data are collected from Masaya volcano during a bi-annual (1998- 1999) measurement campaign. Based on Mie calculations and using the non-linear least square fitting algorithm, the total mass concentration of the SA and SO₂ (and their ratios) are derived that are consistent with previous observations at Masaya. The results show that ignoring co-existent SA can lead to substantial errors in SO₂ estimations. This has very important implications for remote sensing of volcanic plumes.

The MS is very well structured and written. Methods and assumptions are clearly

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explained. From my perspective, it is a significant contribution to volcanic plume observation and thus, should be published at AMT after addressing the following points.

Comments:

1- The term “co-emitted” is misleading. How can you make sure that H₂SO₄ is directly emitted during degassing? There are several studies that show high-temperature oxidation of H₂S and SO₂ affect the sulphur speciation in the plume (e.g. Martin et al 2006; Hoshyaripour et al 2012). Moreover, in ash-free plumes in the troposphere OH could oxidize the SO₂ relatively quickly. Please replace this term with “co-existing”.

2- P4.L2-4: The authors state “the spectral signature of ash and condensed water, both very different and distinguishable from SA spectral signature, is not observed in our dataset, so we exclude the presence of both types of particles”. This makes me wonder:

2-1- What is the source of pre-existing particles on which H₂SO₄ condenses? If there is no water, why there is a 65% sulfate solution? This even contradicts the assumptions made later in the Mie calculations (binary solution).

2-2- What if there is a lot of ash particles and/or water droplets in the plume? Does this method work with ash- and droplet-free plumes (I seriously doubt if the second one exists in nature) only? If so, this statement should be revised: “This method is easily exportable to other volcanoes, to monitor magma extraction processes and the atmospheric sulphur cycle”

3- The authors claim that the method is “easily exportable to other volcanoes” but have analyzed only 30 minutes of data from 2-years measurements conducted 2 decades ago. What is specific about the data that makes it “high quality spectra” and how likely is it to get such data elsewhere? This will elaborate on the requirements and limitations of the method. It would be great to see a second example that shows the applicability of the method to other eruptions/volcanoes.

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4- This MS is submitted to the special issue “StratoClim stratospheric and upper tropospheric processes for better climate predictions”. Please explain how the results obtained from passively degassing volcano like Masaya (with tropospheric plumes) can be generalized and used for UTLS studies. Is it directly exportable to UTLS plumes? If not, what are the key aspects to consider?

References:

Hoshyaripour, G., M. Hort, and B. Langmann (2012), How does the hot core of a volcanic plume control the sulfur speciation in volcanic emission?, *Geochem. Geophys. Geosyst.*, 13, Q07004, doi:10.1029/2011GC004020.

Martin, R. S., T. A. Mather, and D. M. Pyle (2006), High-temperature mixtures of magmatic and atmospheric gases, *Geochem. Geophys. Geosyst.*, 7, Q04006, doi:10.1029/2005GC001186

Interactive comment on *Atmos. Meas. Tech. Discuss.*, doi:10.5194/amt-2019-186, 2019.

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