

Interactive comment on “Investigation of several proxies to estimate sulfuric acid concentration in volcanic plume conditions” by Clémence Rose et al.

Anonymous Referee #2

Received and published: 29 November 2020

Review of Rose et al. Sulfuric acid proxies. This paper tries out various parameterizations for sulfuric acid in the high SO₂ conditions of a volcanic plume. Motivation for this work seems thinly presented, e.g. is an aim to provide a simple equation for GCMs? One set of field data was simulated somewhat reasonably but the other was not. Is that a sign that these proxies are limited in their application? A third finding (p.23 line 11) could be the limited scope of SA proxies and thus a chemical model (e.g. Boy 2005) will be needed in lieu of widespread measurements of SA.

Some detailed comments

p1,line20 'all variables equal weight'? Do F1, F2, and F3 represent just cases of differ-

C1

ent fitted forms? 'adjusted powers' is this varying weights in the fits or also the fitted forms? Succinct description of the main analysis in the abstract should not mislead a potential reader...

p1.line25 But it seems S1 did not do well for the second case ?

p2.lines21-24. This is a mangled sentence that needs to be parsed out or eliminated. The next sentence begins a new thought and deserves a new paragraph.

p3.line16. So a main motivation was to duplicate the Lu et al. study? Needs rewording.

p3.line22. Points out a flaw in the proxy idea in general: missing information is important such as NO, NO₂, CH₂O, O₃, radiative environment, aerosol hygroscopicity ?

p3.line25. This is an unusual statement: they used the proxies but did not test them ? Do you mean compared to measured H₂SO₄? Did not Rose 2019 have measurements?

p4.line15. 'compared to' -> 'than'. Nonetheless, why care about the previous one?

p4.line16. One has O₃ and NO_x too... seems that a Boy 2005 model simulation could be done.

p4.line24. made a non-negligible contribution to what ? Read on... Is there ion-molecule clustering going on or are these ionization of neutral clusters? Should 195u be multiplied by 2 and 293u by 3 to get the total SA being produced? How does one separate ionization of H₂SO₄ dimer and trimer from the clustering of HSO₄- with H₂SO₄? Still no reference for this equation. Or for the AI instrument, next page. Was the calibrations carried out to test the limits of these equations?

p5.line30-32. These two sentences add nothing to the paper, instead they distract. Instead here is where some discussion of the uncertainties in the H₂SO₄ measurements and whether the particulars of the deployment(s) have affected them. Did exp. con-

C2

ditions perhaps introduce more factors (e.g. ambient pressure dependencies of the C factors) ?

p7.line3 Boy 2005 showed that NO +HO2 was a major source. In fact, why not run that model in the plume for comparison?

p10.line8. This seems to be not only wrong chemically but misguided. O3 times radiation (hv) is the correct parameter and you should use it as such. Let potential future users input the ozone for their situation. Also, when water is low such as cold temperatures at altitude, quenching of O(1D) by air molecules would compete. Is SO2 in the plume so high that it is optically thick? Would the UVB be affected more than global radiation (this reveals that 'global radiation' and its measurement should be explained in depth in this context)? Again, Boy 2005 showed that NO + HO2 was the major direct source for OH. All these questions could be addressed by some sort of OH box model run for plume conditions.

p10.line10. kprime is what? The parameter K must have some physical meaning.

Also, CS concerns here. On the face of it, the CS used is not correct and one should apply correction factors as the aerosol is probably hygroscopic (loads of SO2 here.) Furthermore: below 40%? The level of drying is not even known? Uncertainties pile up. A nominally hygroscopic aerosol should be assumed and the CS should be corrected as best as possible, perhaps using half the nom.hyg.aer. correction.

p10.line25 That is huge,35%. It would be a dominant contribution to the signal at 195u.

p10.line30 Give some insight on this term. It represents a net flux to the dimer. That this is a constant for a whole measurement campaign is not realistic. Agreement with any estimated dimer abundance / loss rate should be discussed.

p11.line9. What ? Essentially saying: We don't want to know too much. Also arguing that proxies are not meant to be all that useful? Instead, put as much as you know into it. Then knowledge develops, limitations in it become known.

C3

These last few comments underscore the many levers of uncertainty here. The final section has too-much-summary of the various proxies and there is not enough presentation of a firm conclusion regarding the limits of their chosen proxy (let's not have 8 or 9 to choose from) in the light of the uncertainties.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-636>, 2020.

C4