We thank Referee #2 for their comments and suggestions, which are addressed point by point below.

Review of Rose et al. Sulfuric acid proxies. This paper tries out various parameterizations for sulfuric acid in the high SO_2 conditions of a volcanic plume. **A.** Motivation for this work seems thinly presented, e.g. is an aim to provide a simple equation for GCMs? **B.** 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? **C.** 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.

A. Addressing the lack of [H_2SO_4] measurements, which rely on the use of state-of-the-art instrumentation that requires specific expertise, is presented in the abstract (P1, L21-23) and in the introduction (P3, L1-4) as the primary motivation for the development of the proxies. The recent multiplication of studies dedicated to the development of such proxies listed in the introduction illustrates the interest of such tools (e.g. Lu et al., 2019; Dada et al., 2020), which are not necessarily chemically detailed, but which primary objective is, again, to compensate for the absence of measurements. As highlighted in the abstract (P1, L23) and now clearly indicated in the introduction as well, the knowledge of [H_2SO_4], and in particular the possibility of obtaining an estimate of this quantity from more commonly measured variables, is of particular interest for the study of nucleation and NPF processes:
"*However, as recently noticed by Lu et al. (2019), direct measurements of [H_2SO_4] remain challenging, because the deployment of CIMS and the analysis of the data they provide require specific expertise. Therefore, for studies in which [H_2SO_4] is an important variable (i.e. mainly for nucleation and NPF studies), is it useful to be able to predict it from more accessible observations such as SO_2 concentration and environmental parameters. This is why several proxies for [H_2SO_4] have been developed, based on the assumption that H_2SO_4 mostly results from the oxidation of the sulphur dioxide (SO_2) by the hydroxyl radical (OH).*"
This is particularly the case in volcanic plumes, where sulfuric acid is expected to play an important role in the process (P2, L18-19, Sahyoun et al., 2019); as indicated at the end of the introduction (P3, L25-28; rephrased in the revised version of the manuscript, see Reply 6), the absence of proxies dedicated to this specific environment was in the end the main reason for this study. Motivations related to the specificities of the volcanic plume conditions are also outlined in the abstract (P1, L24-27) and recalled at the end of Sect. 2.3 (P7, L10-12).

B. There is indeed a limit to the application of a given proxy, since it is built on the basis of data from a specific environment, and is therefore not necessarily suitable for the description of other conditions. This limitation is clearly evoked several times in the manuscript: in the abstract (P2, L7-10), in the results section (P20, L11-13) and in the conclusion (P24, L21-25). This limitation is also more widely illustrated by the recent multiplication of studies dedicated to the construction of proxies adapted to contrasting environments (Lu et al., 2019; Dada et al., 2020). Also, the development of proxies is obviously based on a compromise between simplicity and accuracy, but in view of the results obtained (both in this work and in the literature), this approach seems to lead to satisfactory results in various conditions.

C. Based on Boy et al. (2005), it seems that the use of such chemical model requires the knowledge of a certain number of variables, as indicated for instance in the abstract of the abovementioned paper: "Sulphuric acid concentrations were measured and calculated based on pseudo steady state model with corresponding measurements of CO, NOx, O_3, SO_2, methane and non-methane hydrocarbon (NMHC) concentrations as well as solar spectral irradiance and particle number concentrations with size distributions". Some of the listed species (in particular NMHC) are not routinely measured continuously, and therefore we would prefer to avoid considering such compounds in our study. Our willingness to use a limited number of commonly measured variables is indicated several times in the paper (P10, L7-9; P11, L5-9). More broadly, detailed

chemical investigation and/or description of the formation pathways of H_2SO_4 and its precursors in a volcanic plume is behind the scope of the present work, as now further specified in Sect. 3: "*However, since we did not observe a very specific behaviour of these species in the plume compared to regular conditions which could have motivated their inclusion, we rather chose to limit the number of variables to get as simple as possible expressions for the proxies. Similarly, the dependence of H_2SO_4 production term over absolute water concentration was left behind from the present work in order to avoid over-constraints which could prevent the use of the newly developed proxies in datasets collected in different volcanic plumes. More broadly, while Dada et al. (2020) explicitly aimed at understanding the different mechanisms of sulfuric acid formation and losses in different environments, detailed chemical investigation and/or description of the formation pathways of H_2SO_4 and its precursors in a volcanic plume was behind the scope of the present work, which objective was, again, to obtain the simplest possible description of $[H_2SO_4]$ from a limited set of commonly measured variables.*"

However, if the Referee is interested in conducting a more detailed analysis of H_2SO_4 chemistry in a volcanic plume using a model such as the one used in Boy et al. (2005), we would be pleased to contribute with the measurements we have.

Some detailed comments

Comment 1: P1, L20: 'all variables equal weight'? Do F1, F2, and F3 represent just cases of different fitted forms? 'adjusted powers' is this varying weights in the fits or also the fitted forms? Succint description of the main analysis in the abstract should not mislead a potential reader...

Reply 1: A sentence was added to help clarifying this point: "*A specific combination of some or all of these variables was tested in each of the seven proxies. In three of them (F1-F3), all considered variables were given equal weight in the prediction of $[H_2SO_4]$, while adjusted powers were allowed (and determined during the fitting procedure) for the different variables in the other four proxies (A1-A4)*".

Comment 2: P1, L25: But it seems S1 did not do well for the second case?

Reply 2: It seems that the reviewer is referring to L35 instead. However, we are not sure we understand the comment, since L35 concerns the analysis of the performance of the proxies in the first case study only, while the second case is discussed right after (from P1, L35 to P2, L5). The purpose here is to summarize the results obtained for each dataset from the elements that seem most relevant, which we believe does not include the performance of proxy S1 in the case of the STRAP data.

Comment 3: P2, L21-24: 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.

Reply 3: The sentence has been rephrased, and new reference was introduced:
"*Information about the species contributing to cluster formation with sulphuric acid and preferential formation pathways was gained from laboratory studies (Hanson et al., 2002, 2006). Laboratory experiments have also made it possible to evaluate instrumental setups and related protocols for accurate detection and quantification of the clusters and their precursors (Jen et al., 2016; Riva et al., 2019).*"

On the other hand we have kept only one paragraph, since all the sentences that it contains concern the detection of the clusters and their precursors.

Reference: Riva, M., Rantala, P., Krechmer, J. E., Peräkylä, O., Zhang, Y., Heikkinen, L., Garmash, O., Yan, C., Kulmala, M., Worsnop, D., and Ehn, M.: Evaluating the performance of five different chemical ionization techniques for detecting gaseous oxygenated organic species, Atmos. Meas. Tech., 12, 2403–2421, https://doi.org/10.5194/amt-12-2403-2019, 2019.

Comment 4: P3, L16: So a main motivation was to duplicate the Lu et al. study? Needs rewording.

Reply 4: In case there would be an ambiguity related to the use of the expression "*this work*", this expression has been replaced by "*the study of Lu and co-workers*".

Comment 5: P3, L22 : Points out a flaw in the proxy idea in general: missing information is important such as NO, NO$_2$, CH$_2$O, O$_3$, radiative environment, aerosol hygroscopicity?

Reply 5: As indicated in the paper and recalled in the reply to part A. of the main comment, the objective of the proxies is to provide an estimate of [H$_2$SO$_4$] in absence of direct measurements. While some proxies do include some more specific variables (e.g. alkenes concentration in Dada et al., 2020, [HONO] in Lu et al., 2019), most of them (including those developed in the present work) are based on a limited number of commonly measured variables to maximize their usefulness (see reply to part C. of the main comment).

Comment 6: P3, L25: 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?

Reply 6: As indicated on P3, L28, the present work reports "*the first direct measurements of [H$_2$SO$_4$] conducted in plume conditions*", implying that, in fact, measurements were not available in Rose et al. (2019) nor in Boulon et al. (2011). This is now clearly stated:
"In absence of direct measurements, and also of a specific proxy, Boulon et al. (2011) and Rose et al. (2019) […] and the Piton de la Fournaise, respectively. However, the lack of measured [H$_2$SO$_4$] obviously did not make it possible in these studies to assess the performance of the abovementioned proxies in such unusual conditions, which has motivated the present work".

Comment 7: P4, L15:. 'compared to' -> 'than'. Nonetheless, why care about the previous one?

Reply 7: The reader is referred to earlier studies for a more detailed description of the instrumental setup, it seems therefore interesting to mention the changes made to this setup.

Comment 8: P4, L16: One has O$_3$ and NOx too... seems that a Boy 2005 model simulation could be done.

Reply 8: The Reviewer is referred to part C. of the reply to the main comment.

Comment 9: P4, L24: 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?

Reply 9: We first would like to mention that we noticed a mistake in the numerator of Eq. 1 which has been corrected, so that Eq. 1 now reads:

$$[H_2SO_4] = \frac{HSO_4^- + H_2SO_4 \cdot HSO_4^- + (H_2SO_4)_2 \cdot HSO_4^- + H_2SO_4 \cdot NO_3^-}{NO_3^- + HNO_3 \cdot NO_3^- + (HNO_3)_2 \cdot NO_3^-} \times C$$

Also the reference associated to this equation was not correct, it should have been Kürten et al. (2012) instead of 2014.

As now explicitly stated at the end of Sect. 2.1, the mass spectrometer in OCTAVE campaign was calibrated on-site, in the exact position it was sampling the ambient air during the measurement campaign, and up to the high sulfuric acid concentrations observed under the plume conditions. The H$_2$SO$_4$ produced by the calibrator was simulated as usually according to Kürten et al. (2012), which considers the present ambient conditions influencing, for example, the collision frequency. Thus, in this setup the instrumental influence was minimized, as it is indeed not straightforward to answer which of

the clusters is formed first. Yet, as shown in Fig. R1, the H$_2$SO$_4$·HSO$_4^-$ signal was always minor in comparison to HSO$_4^-$ and H$_2$SO$_4$·NO$_3^-$ signals, and the (H$_2$SO$_4$)$_2$·HSO$_4^-$ far smaller than those.

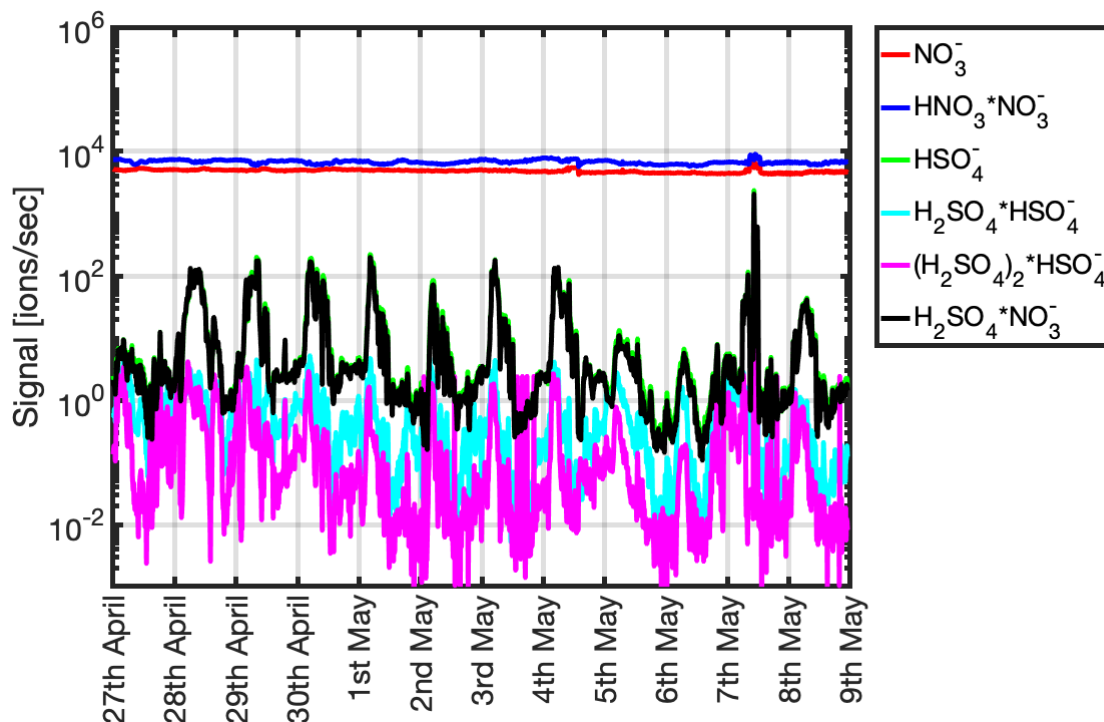Fig. R1: Measured raw ion signals at 10 minute averaging time for the reagent ions (NO$_3^-$ and HNO$_3$·NO$_3^-$) and product ions (HSO$_4^-$, H2SO4·NO3$^-$, H$_2$SO$_4$·HSO$_4^-$ and (H$_2$SO$_4$)$_2$·HSO$_4^-$) during the volcanic plume.

With regard to the setup used during STRAP, although it was previously described in detail in Sahyoun et al. (2019), certain indications should indeed have been recalled in this work, both concerning the instrument itself and the calibration procedure. Those have been added to Sect. 2.2, with, as suggested, a short discussion on the possible effects related to calibration conditions different from those encountered during the campaign on the estimation of [H$_2$SO$_4$]:

"*As previously explained in details by Sahyoun et al. (2019), sulfuric acid concentrations were measured with an APi-TOF equipped with an ambient ionization (AI) inlet adapted to airborne measurements and used for the first time during STRAP. In contrast with the CI inlet, the AI inlet does not require the use of any chemicals, and only includes a soft X-ray source (Hamamatsu L9490) to ionize the sample flow. This direct ionization process was sufficient to get a high enough signal and allow a time resolution as high as 1s for the corresponding measurements. Also, in order to avoid possible effects related to pressure changes on the detection of the AI-Api-TOF, a pressure stabilizing unit was installed in front of the instrument. As detailed in Sahyoun et al. (2019), calibration of this new setup was performed (with respect to [H$_2$SO$_4$] measurement) during fall 2016 at the CLOUD CERN facility (Kirkby et al., 2011; Duplissy et al., 2016 and references therein) by comparison with the measurements performed with a nitrate based CI-APi-TOF in various conditions representative of the atmosphere. During these experiments, O$_2^-$ was assumed to be the main ionizing agent of H$_2$SO$_4$, as on board the aircraft during the measurement campaign, but contribution of NO$_3^-$ could not be excluded, in particular in presence of higher NOx levels (up to 33 ppb) in the CLOUD chamber. Therefore, estimates of [H$_2$SO$_4$] were finally obtained by the mean of Eq. (2) using the signals measured at m/z = 97 Th (HSO$_4^-$) and m/z = 160 Th (NO$_3^-$ · H$_2$SO$_4$) by the AI-APi-TOF and a calibration coefficient C = 4.5 ×10^9 molecule cm^{-3}:*

$$[H_2SO_4] = \frac{HSO_4^- + NO_3^- \cdot H_2SO_4}{Total\ ion\ count} \times C \qquad (2)$$

The good correlation obtained between the signals of the well characterized CI-APi-TOF and the AI-APi-TOF during the calibration experiments can undoubtedly be seen as an indicator of the satisfactory performance of the newly developed inlet, and further on the derivation of [H₂SO₄] (see Fig. S3 in the Supplement of Sayhoun et al. 2019). However, it cannot be excluded that [H₂SO₄] inferred from the measurements carried out during the STRAP campaign were subject to greater uncertainty due to the specific conditions of the volcanic plume, in particular with respect to H_2SO_4 concentrations, which were on average slightly higher in the plume than in the simulation chamber ($< \sim 5 \times 10^7 \ cm^{-3}$ in the CLOUD chamber vs $\sim 1.6 \times 10^8 \ cm^{-3}$ on average during the flight segment of interest, see Table 1 and Fig. 2)".

Reference: Kürten, A., Rondo, L., Ehrhart, S., and Curtius, J.: Calibration of a Chemical Ionization Mass Spectrometer for the Measurement of Gaseous Sulfuric Acid, J. Phys. Chem. A, 116, 6375–6386, https://doi.org/10.1021/jp212123n, 2012

Comment 10: P5, L30-32: 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. conditions perhaps introduce more factors (e.g. ambient pressure dependencies of the C factors)?

Reply 10: As suggested by the Reviewer, the two abovementioned sentences were removed.

The Reviewer is referred to Reply 9 with regard to the possible effects of the particular conditions of deployment of the instruments on the [H₂SO₄] measurements.

Also, a complementary investigation on the effect of a possible systematic error related to the measurement accuracy of [H₂SO₄] and predictor variables on the fitting parameters and performance indicators (i.e. R, RE and SSR) of the newly developed proxies was added at the end of Sect. 3:
"*The data were in addition submitted to bootstrap resampling to evaluate the effect of a possible systematic error related to the measurement accuracy of [H₂SO₄] and predictor variables on the fitting parameters and performance indicators (i.e. R, RE and SSR). The method is described in detail in Dada et al. (2020) and is only briefly recalled here. 10 000 bootstrap resamples were generated from the original dataset by randomly replacing an existing data point with another, and the resulting time series were further multiplied by a set of random factors to simulate the presence of independent systematic errors on the different variables. For each variable, these factors (one per bootstrap sample, i.e. 10 000 in total) were drawn from a uniform distribution (in logarithmic scale) of possible biases in their respective uncertainty range. Specifically, uncertainties in the range between -50% and 100% were considered for measured [H₂SO₄] (i.e. multiplying factors for [H₂SO₄] in the bootstrap resamples were between 0.5 and 2) following the work of Kürten et al. (2012). According to calibration data, we assumed an uncertainty of 15% in the measurement of SO₂ mixing ratio and, similar to Dada et al. (2020), we assumed 20% uncertainty in the CS evaluation. An uncertainty of 5% in the measurement of the remaining variables of interest (i.e. RH and global radiation) was finally accounted for based on manufacturer's specifications. For each function listed in Table 2, the fitting procedure was first applied to the original dataset to obtain a set of reference parameters for deriving [H₂SO₄]. The variability of the fitting parameters and performance indicators was then evaluated for each proxy by repeating the same procedure on the bootstrap resamples.*"

Corresponding results are presented in Table 3, which is introduced at the beginning of Sect. 4.1:
"The following discussion focuses on the fitting parameters and performance indicators (i.e. R, RE and SSR) obtained for the original dataset, but Table 3 presents as well an estimate of their variability (25th and 75th percentiles) inferred from the bootstrap procedure introduced in the previous section."

As specified in Sect. 4.2, STRAP data were in contrast not submitted to bootstrap resampling in the development of the proxy F2': "*Note that the evaluation of a possible systematic error related to the measurement accuracy of [H₂SO₄] and predictor variables on the fitting parameters and performance*

indicators was left behind this last test, which purpose was simply to get an estimate of the improvement in proxy performance related to the derivation of location specific coefficients."

Comment 11: P7, L3: Boy 2005 showed that NO+HO$_2$ was a major source. In fact, why not run that model in the plume for comparison?

Reply 11: We did not have continuous measurement of [OH] during OCTAVE, so we cannot compare model outputs with observations. Also, we tried to quantify [HO$_2$] with Br- mass spectrometer but were unsuccessful; we know it is not a simple quantity to estimate, and thus we do not want to attempt it either. More broadly, as already mentioned in the reply to the main comment, the aim of this work was not to investigate nor describe in detail the formation pathways of H$_2$SO$_4$ and its precursors in a volcanic plume, but to provide simple expressions to estimate [H$_2$SO$_4$] with a sufficient level of confidence.

Comment 12: P10, L8: A. This seems to be not only wrong chemically but misguided. O$_3$ 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. B. 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)? C. 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.

Reply 12:

A. Replacing [OH] with global radiation was approved in previous studies dedicated to the development of proxies for [H$_2$SO$_4$] in various environments (Petäjä et al., 2009; Mikkonen et al., 2011; Dada et al., 2020). Following the Reviewer's suggestion, we have nonetheless tried to replace global radiation by the product $Rad \times [O_3]$ in all the proxies. As now shown in the Supplement (Table S1 and Figs. S3-S4), this did not lead to any significant improvement of the predictive ability of the proxies which, with the exception of A2, all saw a deterioration of their performance indicators. This sensitivity test and the corresponding results are briefly discussed in Sect. 4.1:

   "*As a sensitivity test, an attempt to replace global radiation by the product $Rad \times [O_3]$ was made in all the proxies, to investigate if the explicit consideration of O$_3$, which photolysis is the main pathway for OH formation during daytime, would allow to further optimize the prediction of [H$_2$SO$_4$]. Corresponding results are reported in the Supplement (Table S1 and Figs. S3-S4), but do not highlight any improvement in the performance of the proxies, which, with the exception of A2, all display worse performance indicators than when considering global radiation alone. Limited improvement in the predictive ability of the proxies was also noticed by Lu et al. (2019) when considering [O$_3$] with UVB in the urban atmosphere of Beijing, where the concurrent inclusion of [HONO] seemed in contrast to be more critical.*"

   Regarding the conditions, they were not particularly dry at Maïdo during the OCTAVE campaign (see Table 1 and Fig. 1, median RH ~ 68% in plume conditions), and are not in general, as reflected by the frequent presence of clouds in the vicinity of the station. Moreover, despite its altitude, this station does not experience very cold temperatures due to its geographical location (see Fig. 8 in Foucart et al., 2018, monthly averages > ~9°C in 2015). More broadly, even if high altitude sites (in particular those located at higher latitudes) do in general experience colder temperatures compared to lowland sites, they are also often associated with high frequency of cloud occurrence (e.g. 60% on average during winter at puy de Dôme, France, 1465 m a.s.l.; Baray et al., 2019), which indicates that they are not characterized by particularly dry conditions. So we do not think there is a need to consider any term in the proxies related to quenching of O(1D) by air molecules that would be justified by specific conditions at high altitude.

Reference: Baray, J.-L., Bah, A., Cacault, P., Sellegri, K., Pichon, J.-M., Deguillaume, L., Montoux, N., Noel, V., Seze, G., Gabarrot, F., Payen, G. and Duflot, V.: Cloud Occurrence Frequency at Puy de Dôme (France) Deduced from an Automatic Camera Image Analysis: Method, Validation, and Comparisons with Larger Scale Parameters, Atmosphere, 10(12), 808, doi:10.3390/atmos10120808, 2019.

B. No, the plume was not optically thick, because SO$_2$ concentration was very high only for brief moments; the highest concentrations measured during OCTAVE (> 200 ppb) have furthermore been excluded from the analysis (P12, L16-18). It cannot be excluded, however, that the response of UVB radiation was slightly different from that of global radiation to plume conditions. However, this analysis is outside the objectives of this work, where, consistent with the abovementioned studies, the choice was made to consider global radiation, once again to favour the reuse of the proposed proxies (P10, L7-9), since UVB is generally not systematically measured.

C. The Reviewer is referred to reply 11 as well as reply to part C. of the main comment.

Comment 13: P10, L10: 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 SO$_2$ 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 corrected as best as possible, perhaps using half the nom.hyg.aer. correction.

Reply 13: The meaning of k'is now indicated after Eq. 6 (which is now Eq. 5): "*where k′ corresponds to the multiplication of k by a factor (to be determined in the fitting procedure) which partly takes into account the use of global radiation instead of [OH]*". Some information about the meaning of K was provided on P10, L20-22, and is now recalled also in the legend of Table 2: "*The pre-factors a and K as well as parameter α are assumed to take into account the use of global radiation instead of [OH] in the different proxies.*"

Regarding the treatment of CS, we used the same approach as Mikkonen et al. (2011), who also calculated the CS in dry conditions, and did not apply a specific correction for the hygroscopic particle growth (this is also the case for the data from Budapest in Dada et al., 2020). There is no detailed characterisation of the particle hygroscopic growth factor at Maïdo, so, in line with the following lines from Mikkonen et al. (2011), we rather used the simple approach of including RH directly in some of the proxies: "Also, hygroscopic particle growth is expected to differ between different measurement sites and is, moreover, a function of season and air mass. A careful consideration of the hygroscopic growth effect on CS would thus require considerable additional effort." Still following the work of Mikkonen and co-workers, detailed consideration of the particle hygroscopicity might in addition have limited value only: "Sensitivity tests for Hyytiala data indicate that the hygroscopicity correction is not of significant magnitude to remarkably improve the calculated sulphuric acid proxies".

These aspects are now explicitly mentioned in the revised version of the paper (Sect. 3):
"*Applying a specific correction for the hygroscopic particle growth would have required a detailed characterisation of this process (e.g. as a function of air mass type, season) which has not been performed at Maïdo, and is likely not either available at a number of sites where the newly developed proxies could be used. Also, according to Mikkonen at al. (2011), such hygroscopicity correction might, at least in some environments, have only limited effect on the prediction of [H$_2$SO$_4$]. Therefore, similar to Mikkonen et al. (2011), inclusion of RH in the sink term was tested instead*".

Comment 14: P10, L25: That is huge,35%. It would be a dominant contribution to the signal at 195u.

Reply 14: There seems to be a misunderstanding here, because the term that is included in proxy S1 $(-\beta \times [H_2SO_4]^2)$ does not directly correspond to the signal measured at 195 amu, but to the square of the acid concentration to which all the ions present in the numerator of Eq. 1 are expected to contribute.

Comment 15: P10, L30 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.

Reply 15: As indicated in the paper (P10, L23-26), the term $-\beta \times [H_2SO_4]^2$ is introduced in S1 as a proxy for the sink of H_2SO_4 related to molecular cluster formation. As recalled in Reply 14, it is expressed as a second order function of the sulfuric acid concentration, which is calculated from the signals of all the ions present in Eq 1., among which the so-called dimer.

The systematic consideration of this term in the proxy may indeed be questionable, since the occurrence of molecular cluster formation/nucleation is not continuous. However, the high H₂SO₄ levels encountered in volcanic plumes seem to favour the occurrence of the process, as evidenced by the high NPF frequencies reported by Rose et al. (2019), thus indicating that the inclusion of this term is certainly often relevant/justified under plume conditions. Finally, the fairly good performance observed for S1 (among the best for OCTAVE with A1) confirms that considering this additional term contributes to satisfactory predictions for [H₂SO₄], or at least does not strongly alter them.

Comment 16: P11, L9: 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.

Reply 16: The Reviewer is referred to the replies to the main comment.

Comment 17: 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.

Reply 17: We hope that with the answers previously proposed and the clarifications made to the objectives (compensating for the absence of measurements, not aiming at a detailed understanding of H₂SO₄ formation pathways) and to the approach chosen for the development of proxies (based on a limited number of commonly measured variables), the uncertainty levers identified by the Reviewer will no longer be considered as such.

We are not sure what the Reviewer means by «The final section has too-much-summary of the various proxies". Concerning the fact that no firm indication is given regarding the proxy to select, it is true. This is explained by the fact that, overall, all suggested proxies display performances which, although slightly contrasted over the different [H₂SO₄] ranges and possibly variable depending on the specificities of the datasets and/or associated environments, can be globally considered acceptable. It is hardly possible, on the basis of our observations/results, to provide more directive indications. The only criterion that is not a priori subjective and that can possibly guide the choice is based on the availability, for a given site, of the data necessary for the application of the various proxies.