We thank Referee #3 for their comments and suggestions, which are addressed point by point below.

Comment 1: One aspect I found to be missing was a discussion of the uncertainties in both the proxies and the measurements. H_2SO_4 measurements in general have associated uncertainties on the order of +/- 35-45%, however in this work none were reported. The parameters used in the proxy calculations also have associated uncertainties resulting in an overall uncertainty in the estimated H_2SO_4 concentration. These uncertainties (both the measurements and calculations) need to be discussed in terms of the comparisons.

Reply 1: The uncertainty associated to [H_2SO_4] measurements was briefly evoked in Sect. 2.3, in the comparison between OCTAVE and STRAP datasets, and is now further discussed, in the case of STRAP, at the end of Sect. 2.2:
"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 [H_2SO_4] (see Fig. S3 in the Supplement of Sayhoun et al. 2019). However, it cannot be excluded that [H_2SO_4] 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 (< ~5×10^7 cm^{-3} in the CLOUD chamber vs ~1.6×10^8 cm^{-3} on average during the flight segment of interest, see Table 1 and Fig. 2)."
Correspondingly, additional information regarding the calibration conditions during OCTAVE is now provided at the end of Sect. 2.1:
"*Note that the mass spectrometer was calibrated onsite, 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*."

It is true that the effects of measurement uncertainties of both [H_2SO_4] and predictor variables on the fitting procedure were not assessed in the original version of the manuscript. In order to address this aspect, a bootstrap procedure was applied to the data, following Dada et al. (2020), and systematic errors on the different variables were simulated in the bootstrap resamples (10 000 in total). Similar to Dada et al. (2020), we only accounted for the error related to measurement accuracy (as opposed to measurement precision) and we assumed:
- a factor of 2 uncertainty for [H_2SO_4], based on the work of Kürten et al. (2012);
- 5% uncertainty in the measurement of RH and global radiation, based on manufacturer's specifications;
-15% uncertainty in the measurement of SO_2 mixing ratio, based on calibration data;
- 20% uncertainty in the CS evaluation, similar to Dada et al. (2020).

For each proxy, the fitting parameters previously obtained from the original dataset were kept as a reference for deriving [H_2SO_4], and the results obtained from the bootstrapped data were used to evaluate the variability (25th – 75th percentile) of the fitting parameters and performance indicators associated to each proxy. This additional procedure is described in Sect. 3 of the revised manuscript:
"The data were in addition submitted to bootstrap resampling to evaluate the effect of a possible systematic error related to the measurement accuracy of [H_2SO_4] 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 [H_2SO_4] (i.e. multiplying factors for [H_2SO_4] 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 SO_2 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 [H_2SO_4]. 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 [H_2SO_4] 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.*"

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 2: Also at such high H_2SO_4 concentrations (approaching 10e^9 molecule cm-3), the reagent ion will be depleted in the NO_3 CIMS measurements. The stated concentration calculation assumes pseudo first order kinetics which most likely are no longer applicable under these conditions. Were calibrations performed using these large H_2SO_4 concentrations to prove the validity of the calculation?

Reply 2: There is no inherent need of reagent NO_3^- to be depleted with such a sulfuric acid concentration, and such a depletion was also not observed during the plume measurements (see Fig. R1 below). The sufficiency of reagent ion production ultimately depends on (i) the availability of reagent ions (= primary ion yield in the ionization source), (ii) the concentration to be measured, and (iii) the sensitivity to the target compound. If the 10^9 cm^{-3} target concentration would be a limit for smooth NO_3^- ionization operation, then NO_3^- technique could not be used to measure HOMs and other oxidized organic compounds often present in higher concentrations, and for which it is routinely applied for in field campaigns. Furthermore, as now explicitly stated at the end of Sect. 2.1, the mass spectrometer was calibrated on-site in the exact position it was also sampling the ambient air during the campaign, up to the high sulfuric acid concentrations observed during the plume measurements. The H_2SO_4 produced by the calibrator was simulated as usually by the method described in Kürten et al., (2012) which considers the present ambient conditions influencing, for example, the collision frequency.

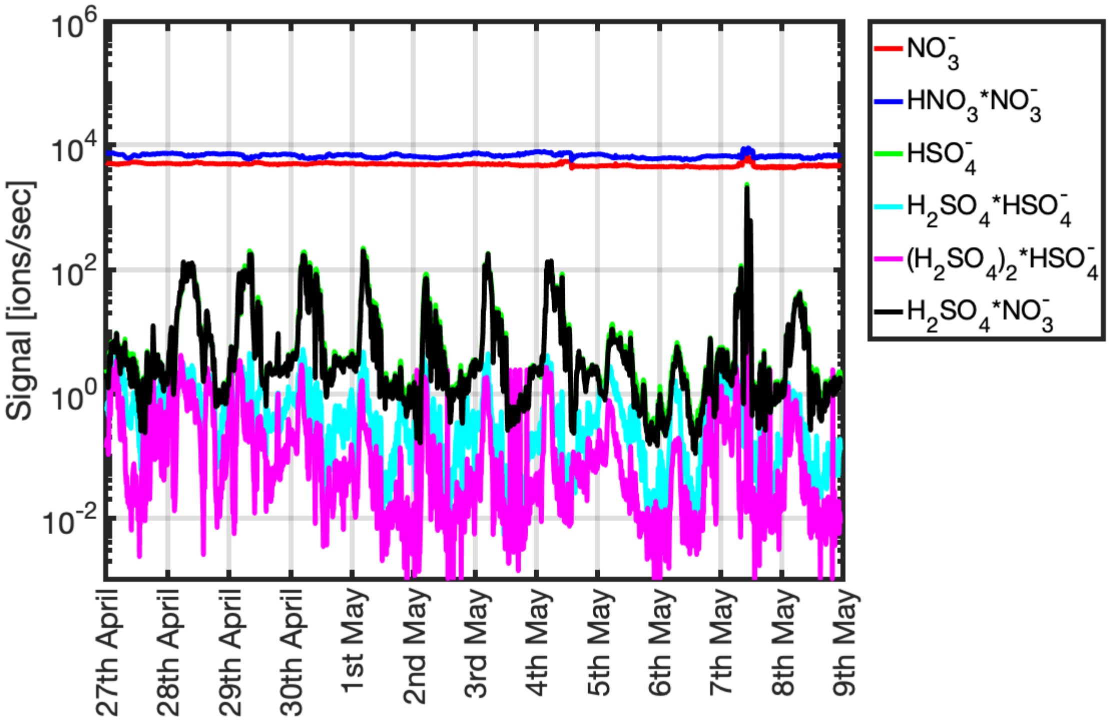

Fig. R1: Measured raw ion signals at 10 minute averaging time for the reagent ions (NO_3^- and $HNO_3 \cdot NO_3^-$) and product ions (HSO_4^-, H2SO$_4$·NO3$^-$, $H_2SO_4 \cdot HSO_4^-$ and $(H_2SO_4)_2 \cdot HSO_4^-$) during the volcanic plume.

Comment 3: Another aspect was the discussion of the airborne measurements. The best agreement between proxy and measurements was observed when no CS term was included (not very realistic). It was also stated that there were gaps in radiation data caused by improper measurements during turns, when the aircraft itself affected the amount of radiation reaching the sensor. Was upwelling radiation considered in these calculations? Assuming these measurements were made inside the plume with particles present, the nadir or reflected radiation could approach that of the zenith or incoming. An increase in the H_2SO_4 production term could balance the inclusion of the CS term.

Reply 3: Upwelling radiation was not included in the calculations. However, as shown in Fig. R2 below, upwelling radiation was relatively constant over the investigated period, and represented on average less than 10% of the downward radiation used in the calculations. Therefore, we do not believe that ignoring this term can explain our observations, for which other hypotheses are proposed, including different size ranges for the calculation of CS (P19, L5-7) and possibly contrasting origin of the particles contributing to the CS (P19, L8-11) in the two datasets.

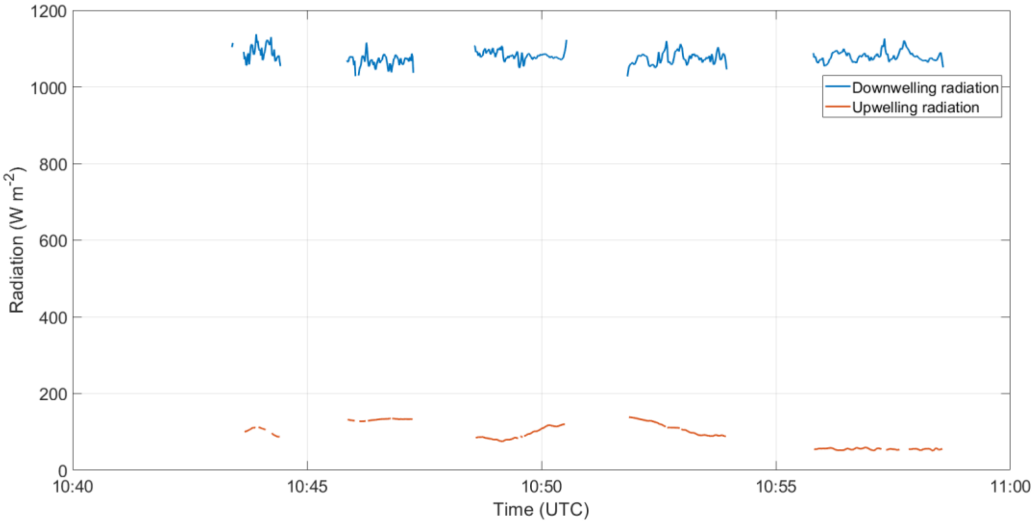

Fig. R2: Downwelling and upwelling radiation measured onboard the French ATR-42 research aircraft during the first part of flight ETNA 13 (STRAP). Gaps in the time series were caused by improper measurements during turns.

Comment 4: I would find a statement about the estimated lifetime of gas phase H_2SO_4 under these high H_2SO_4 conditions useful too.

Reply 4: An accurate evaluation of the lifetime of gas phase H_2SO_4 would require a detailed knowledge of the individual sources and sinks of this particular species under the conditions of the volcanic plume. Such an investigation is beyond the scope of the present work and is not in any case needed for the proxy derivation. Also, it would add unnecessary speculation to the current narrative, which is based solely on the measured "simple" quantities. However, the first-order CS values derived from the DMPS data serve as a measure of the lifetime for aloft species which irreversibly condense by interaction with a surface (i.e., it is a measure of how fast vapours are lost on pre-existing particles), and its inverse value can be used as a first estimate of H_2SO_4 lifetime under these conditions.

Specific comments and suggested grammatical changes:

Comment 1: One large one to mention here is the omission of the proxy equation (Eq. 11) from Mikkonen 2011. It is useful and relevant to this work and not proper to ask the reader to look up.

Reply 1: The proxy developed by Mikkonen et al. (2011) is now recalled at the end of Sect. 4.1.

Comment 2: All Fig. X should be Figure X in text

Reply 2: We are not sure about the Reviewer's expectations here, but it seems that the use of the abbreviation "Fig. X" is part of ACP usage, with the exception of the beginning of a sentence, where the use of the full expression "Figure X" is requested.

Comment 3: Consider replacing "regular conditions" with "low SO2 conditions".

Reply 3: We believe that the term "low" in the proposed expression can be considered subjective, and would therefore prefer to keep the original expression, which meaning is clarified at first use (P6, L16-17: "*i.e. outside of the volcanic plume*").

Comment 4: Page 6, line 19: with had a -> which had a

Reply 4: typo corrected, thank you for noticing!

Comment 5: Page 6, line 27: in and off-plume -> in and out of plume

Reply 5: changed

Comment 6: Lower median radiation during STRAP than OCTAVE due to diurnal observations during OCTAVE. Why not compare same time of day?

Reply 6: We do not believe that the addition of such "detailed" comparison is necessary since the purpose of Sect. 2.3 is to provide an overview of the conditions encountered during the two campaigns and to highlight the specificities of each of the datasets that possibly impact the use of these data for the development of the proxies (e.g. P12, L7-8: "*the variability of the key variables driving H_2SO_4 production was too limited in the STRAP dataset to retrieve a realistic picture of the role of these variables in predicting [H_2SO_4]*") and are also to be taken into account in the interpretation of the results.

Comment 7: Page 7, line 1: in and off-plume -> in and out of plume (you use outside plume in line 3)

Reply 7: changed

Comment 8: Figure 1 - Why not use black for radiation similar to other plots. Yellow for radiation is hard to see. Consider changing the colour.

Reply 8: Changing the colour of radiation data was indeed a good suggestion to improve the readability of Figs. 1 and 2!

Comment 9: Page 10, lines 1-3: Equation is busy and unnecessary

Reply 9: Consistent with Comments 1 and 25, we think it is indeed relevant to remind the reader of the equations useful for this study. In this approach, we believe that, like Mikkonen's proxy, the expression of the temperature-dependant reaction rate between SO_2 and OH is of interest here since it is used in the expression of each of the proxies; it was nonetheless moved to the Supplement.

Comment 10: Page 10, line 10: define k'

Reply 10: We had indeed omitted to mention the meaning of k', which is now clearly indicated after Eq. 6 (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]*".

Comment 11: Page 10, Line 11: we defined in parallel proxy A1 -> we defined a parallel proxy A1

Reply 11: commas were added instead: "*we defined, in parallel, proxy A1*"

Comment 12: Page 10, line 18: CS was first removed in proxies F2 and A2, and it was reintroduced. . . -> the CS was first removed in proxies F2 and A2, and re-introduced. . .

Reply 12: changed

Comment 13: Page 10, line 22: respectively for "F" and "A" proxies. -> respectively for the "F" and "A" proxies.

Reply 13: changed

Comment 14: Page 10, line 24: contribute up to ∼35% to -> contribute up to ∼35% of

Reply 14: changed

Comment 15: Page 10, lines 29-30: definitions of alpha and beta need to come earlier.

Reply 15: We are not sure we understand the expectations of the Reviewer, since alpha and beta are defined right after the equation of proxy S1, in which they are used.

Comment 16: Page 11, line 13: which is defined as follow for -> which is defined as follows for

Reply 16: changed

Comment 17: Page 11 Table 2: parameters need to be defined, especially k. Is K x k = k' in equation 6? As general rule the reader should not have to read the text to understand a figure or table.

Reply 17: As we have now indicated (see Reply 10), k' reflects the presence, in addition to k, of a factor that partially takes into account the use of global radiation instead of [OH] in the determination of [H_2SO_4]. As indicated in Sect. 3 (P10, L21-22), this "scaling factor" is indeed included in the pre-factors K (for proxies F1-F3) and a (for proxies A1-A4), and in parameter α for S1. This information has been added in the legend of Table 2, together with the definition of k:

"*Table 2 Proxy functions. F1-F3 are the proxies with powers fixed to -1 or 1 for all variables, as predicted by the theory, while A1-A4 have individual adjusted powers for each variable. S1 includes the additional H_2SO_4 sink related to cluster formation. In each of the proxies, k corresponds to the temperature dependant reaction rate between SO_2 and OH. Fitting parameters K in F1-F3, a – f in A1-A4 and α - β in S1 were determined iteratively to minimise the sum of squared residuals associated to each proxy. 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.*"

Comment 18: Page 12, line 12: in a different volcanic plume, in which the conditions however overall resembled the average conditions encountered during OCTAVE. -> in a different volcanic plume with conditions similar to those encountered during OCTAVE.

Reply 18: changed

Comment 19: Page 12, line 13: and we believe that their behaviour was not well caught due to their limited number. -> and due to the limited number of measurements probably do not represent H2SO4 concentrations under such large concentrations of SO2 as a whole.

Reply 19 : changed

Comment 20: Page 12, line 30: measurement -> measurements

Reply 20: changed

Comment 21: Page 13, line 1: make them better -> improve results

Reply 21: changed

Comment 22: Page 13, line 6: in regular conditions, who noticed a better performance of the proxies when taking RH into account. -> who noticed a better performance of the proxies when taking RH into account under regular conditions.

Reply 22: changed

Comment 23: Page 13, line 22: point up that overall -> point out that the overall

Reply 23: changed

Comment 24: Page 13, line 23: shown on -> shown in

Reply 24: changed

Comment 25: Page 14, line 23: Eq. (11). At first look I thought this referred to Eq. (11) in the present work, which doesn't exist. I'm assuming this refers to Eq. (11) in Mikkonen et al. (2011). The equation needs to be added here so the reader doesn't have to go look it up!

Reply 25: As already mentioned in Reply 1 above, the proxy developed by Mikkonen et al. (2011) is now explicitly recalled at the end of Sect. 4.1.

Comment 26: Page 19, line 5: CS -> CSs

Reply 26: changed

Comment 27: Page 23, line 15: The condensation sink (CS) was in a first approach considered as the only sink contributing to the balance of [H_2SO_4] to derive seven proxies based on the knowledge of SO_2 mixing ratios, global radiation, CS and RH, which inclusion in the sink term was tested in several formulations. -> The condensation sink (CS) was in a first approach considered as the only sink contributing to the balance of [H2SO4] to derive seven proxies based on the knowledge of SO2 mixing ratios, global radiation, and CS. RH, included in the sink term was tested in several formulations.

Reply 27: changed

Comment 28: Page 23, line 28: In contrast, the inclusion of RH,. . . This sentence is long and runs on. Needs to be rewritten and split up to convey conclusions.

Reply 28: As suggested, the sentence was split into three shorter sentences: *"In contrast, the inclusion of RH, either with the CS or with a separate power, did not improve the performance of proxies A3 and A4, respectively, compared to A1. This observation suggested a limited effect of RH on the sink regulating* H_2SO_4 *production, in spite of the relatively high negative correlation observed between these two variables. More importantly, power -1 attributed to RH in proxy F3 lead to a systematic underestimation of [*H_2SO_4*]."*

Comment 29: Page 24, line 1: and on average higher at – but was on average higher at

Reply 29: changed

Comment 30: Page 24, line 4: the literature -> that work

Reply 30: changed