

Anonymous Referee #1

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We thank the reviewer for the helpful comments. Our answers to the concerns addressed are below.

The manuscript "A statistical proxy for sulphuric acid concentrations" tries to find good parameterizations for the estimation of H₂SO₄ concentrations when no direct measurements are carried out. In general, the study is based on good quality observations and the methods are sound. It does not provide significantly new insights, but combines earlier observations in easily usable form definitely helping in other future studies (see also the general comment below). It can be published after the questions & suggestions given below are properly addressed.

A general comment on the results:

One of the findings is that a simple proxy, based on radiation and [SO₂] only, is able to predict [H₂SO₄] almost as accurately as the more complicated parametrizations. As such simple proxy is more widely applicable, it should be used instead of more complicated ones (requiring measurements of particle size distributions). So please focus on proxy L3 in the discussions.

Wider usability of proxy L3 is highlighted more in the discussion, although the proxy in Eq. 8 (Eq. 9 in the revised manuscript) will be still presented as the most accurate one. See also the uncertainty analysis done due to the request from the Referee 2. Sentence "it was also shown that it is possible to gain an approximation almost equal to the result of the best proxy with only radiation and SO₂ concentrations" can be found from the last paragraph of the Conclusion.

Abstract:

- please add in the abstract the actual parameterization equation together with the validity range of the parameterization. This validity range should preferably include: - Temperature range, RH range, altitude range, SO₂- and H₂SO₄-range, solar radiation range - types of the environment where the proxy is applicable (continental urban, rural, climate conditions, industrial etc, but not e.g. marine, arctic, night etc). This is to prevent the use of the proxy in areas/conditions where it is not tested. Please include this also to "conclusions". Best would actually be to include a small table giving the validity range & conditions of the parameterization. - Please indicate that in the final proxy, Hohenpeissenberg is used for proxy validation only, not for actual proxy creation. - Focus on the simpler proxy, as the more complicated provides little extra value compared to the increased number of measurements (& troubles) needed.

Inserting equations in the abstract would decrease its readability. Atmospheric conditions for the parameterization are introduced in Table 2 and they are pointed out in the conclusion with sentences: "Tests suggest that this universal proxy is suitable for the prediction of sulphuric acid concentration under a wide range of continental atmospheric conditions. Note, however, that the proxy has not been tested for e.g. marine, Arctic or desert conditions. Details of the conditions can be seen in Table 2."

The role of the Hohenpeissenberg data will be written more clearly. Sentence "Note that the data from Hohenpeissenberg were not used in the proxy construction." was inserted into page 16, lines 4-5 of the revised manuscript.

Intro: Page 20146, row 7: "also" - "in contrast" would suit better

Sentence changed to form: "In contrast, their findings showed that in a clean environment, Hyytiälä, Finland, SO₂ concentrations were lower on days when NPF occurred."

Data:

- Add in Table 2 the 5% and 95% percentiles for all variables to provide information on the validity range of the parameterizations - Add the temperatures for the periods.

Requested percentiles and temperature are added to Table 2.

Page 20148, row 1: The use of the median radiation is somewhat confusing - I would prefer the use of daily maximum radiation as it more clearly describes the situation, location and season - especially as the authors only utilize the data from the light periods. Or is it the mean radiation for light period only?

As stated in the caption of Table 2, only times when it was not completely dark are counted in the comparison of the Radiation entries. In order to avoid confusion, we also inserted clarification for this into page 6, lines 23-24 of the revised manuscript.

The median for daylight hours was originally reported in the table instead of daily maximum because we wanted to show the overall conditions of the period, not just the peaks. We inserted the 5% and 95% percentiles to the table to describe the magnitude of the variation. I also want to stress that in the analysis we used 10 minute averages of the hours when Radiation was $>10 \text{ W m}^{-2}$, not the daily averages.

Experimental:

Page 20148, row 22: please remove word "innovative" Page 20148, row 23: "allows"

Removed and corrected

Page 20151, row 7: reference to a wrong figure

Correct figure inserted into the revised manuscript.

Page 20151, row 25-27: Correlating H₂SO₄ with CS x RH is problematic, as H₂SO₄ variation follows solar radiation and RH daily temperatures (which are also correlated with solar radiation). Please check e.g. correlation of [H₂SO₄] and T x (CS)⁻¹ - what is the correlation coefficient now? Thus, this argumentation is on somewhat loose bases.

This is also proven by the quality predictions of the proxy L3 not including RH, and Fig 5.

Spearman correlation R between [H₂SO₄] and T x (CS)⁻¹ is less than 0.1 in all places except nwr, where R=0.55. This means that the stronger strengthening of the correlation when CS is multiplied by RH is not caused by temperature change.

Please, add a table describing the validity range of the parameterization. Show the two parameterizations given in conclusions at the top of the table.

The validity range can be seen from the updated Table 2. This is now stated in the Conclusions section, page 16, lines 7-8 of the revised manuscript. The parameterizations are already shown in the Conclusion section.