Review of Dada et al "Sources and sinks driving sulphuric acid concentrations in contrasting environments: implications on proxy calculations" by Anonymous Referee

The manuscript, "Sources and sinks driving sulphuric acid concentrations in contrasting environments: implications on proxy calculations," by Dada et al. describes a new method for estimating gas phase H2SO4 concentrations using relatively common measurements. The development of these so-called "proxies" for H2SO4 is important as this species is often used in global models for simulating the timing and intensity of new particle formation events. Additional proxies are especially needed for representing regions that were not include in previous attempts (e.g., China) or during time periods that we not considered previously (e.g., nighttime). Thus, this manuscript is potentially valuable and is, in principle, worthy of publication in ACP. I do however, wish to point out a one main item and a few minor issues that I would like the authors to respond to prior to recommending publication.

We thank the reviewer for their valuable comments and suggestions, we think that these help improve the presentation of the proxy and the overall quality of the study. We provided point-by-point answers in purple. Insertions to the text are in *Italics*. Line numbers refer to the old version of the ACPD version of the text.

As a major concern: In the abstract of this manuscript and throughout the text the authors claim that the new proxy is "a more flexible and an important improvement of previous proxies." While that may be true, we only are provided a comparison to the previous proxy developed in a pristine boreal forest atmosphere (the Petaja proxy). Nowhere do the authors compare their new proxy to that developed by Mikkonen et al. First of all, this makes little sense as the Mikkonen model was developed for a broader range of conditions than the Petaja model. If there is a valid reason to disregard the Mikkonen model then the authors should state that, or else they should show model predictions from that on all relevant figures as they did with the Petaja model. Otherwise they should remove the statement that the model is an improvement over other proxies, as they are only comparing to one.

We agree with the reviewer that it is rather crucial to compare to Mikkonen et al. as it has been developed for several locations including a broad range of conditions. However, since our proxy includes periods that we have not considered previously (e.g., nighttime), we still think that it is an improvement over previous proxies.

We compared our proxies with Mikkonen et al. 2011 in all 4 locations, and added the diurnal Mikkonen plot to the main text (Figures 2,4, 6 and 8) while the scatter plots between measured sulphuric acid concentrations and both of Petäjä and Mikkonen proxies during daytime (GlobRad >= 50 W/m^2) in Figures S13 and S14, respectively.

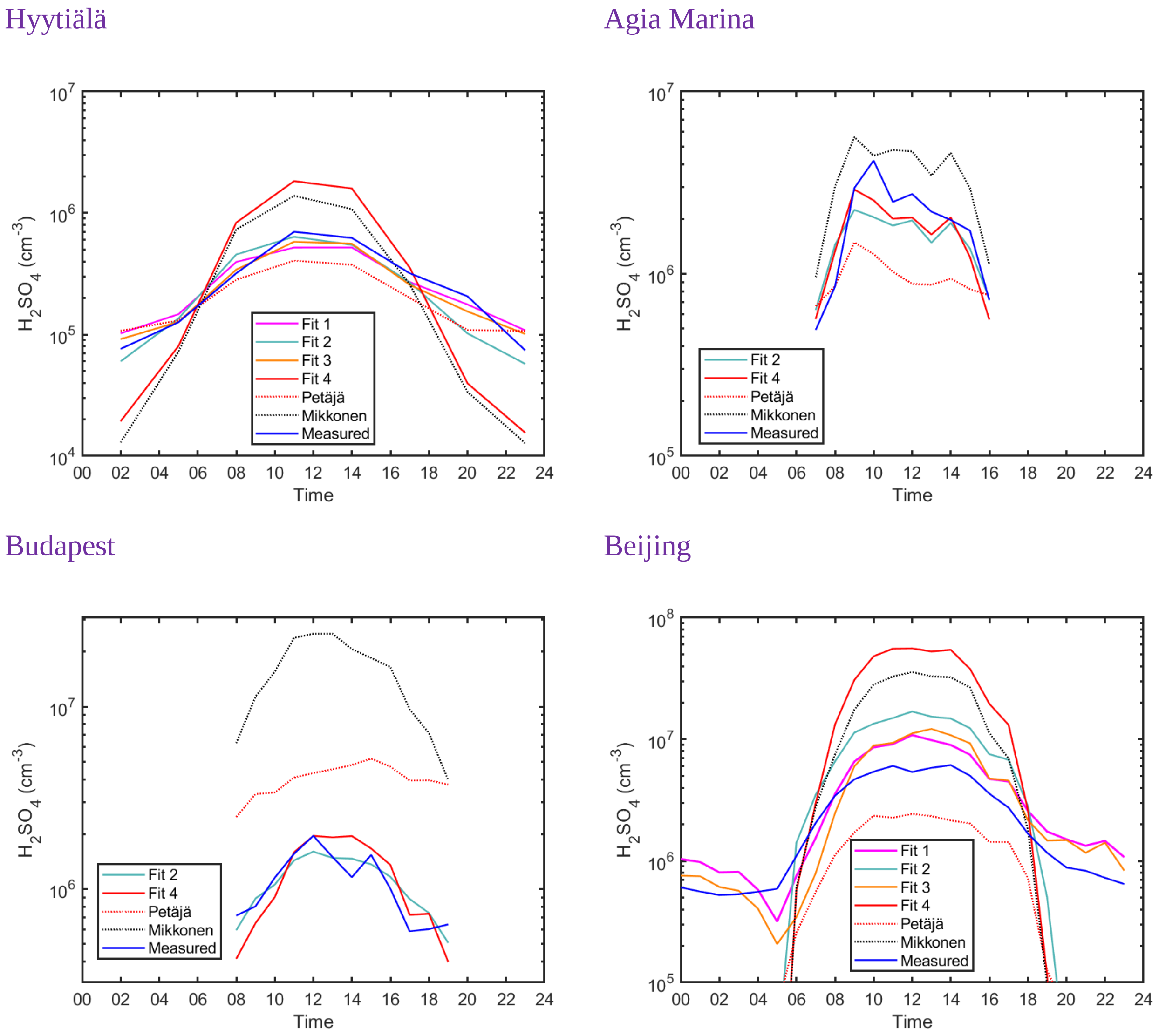
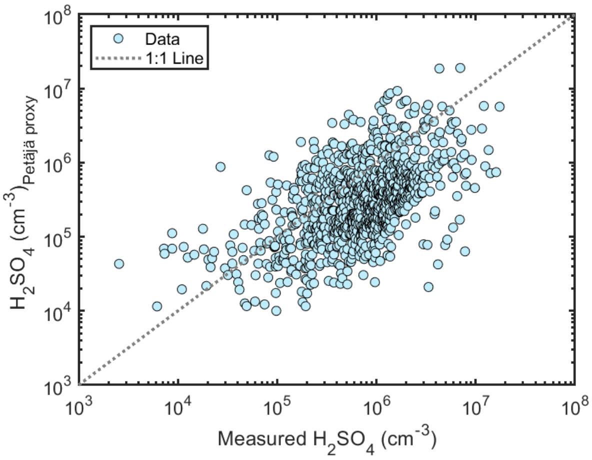
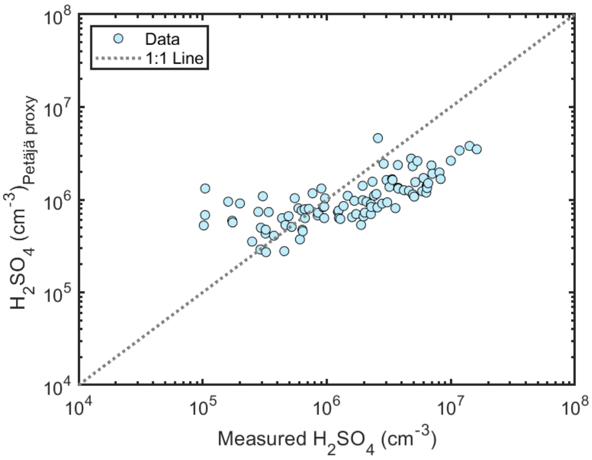


Figure R 1 The diurnal variation of sulphuric acid proxy concentrations using different fits and observed concentrations. Median values are shown. Fits 1,2, 3 and 4 corresponds to the Equations 2, 4, 5, and 6, respectively. Petäjä fit shown is applied using the coefficients reported in Petäjä et al. 2009 (Equation 7) and Mikkonen et al. 2011 (Equation 8).

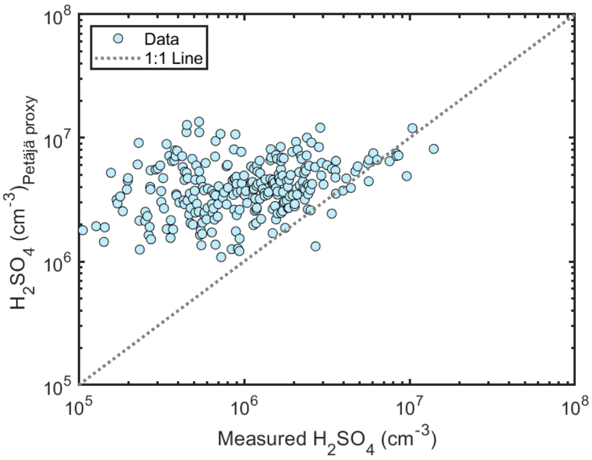
Hyytiälä



Agia Marina



Budapest



Beijing

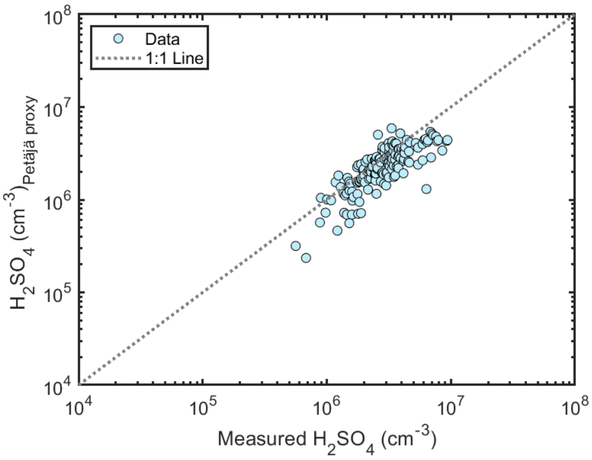


Figure R 2 Scatter plot showing the correlation between measured sulphuric acid and the sulphuric acid concentrations derived from the Petäjä et al. 2009 proxy at the 4 locations during daytime (GlobRad >= 50 W/m^2): Hyytiälä, Agia Marina, Budapest and Beijing.

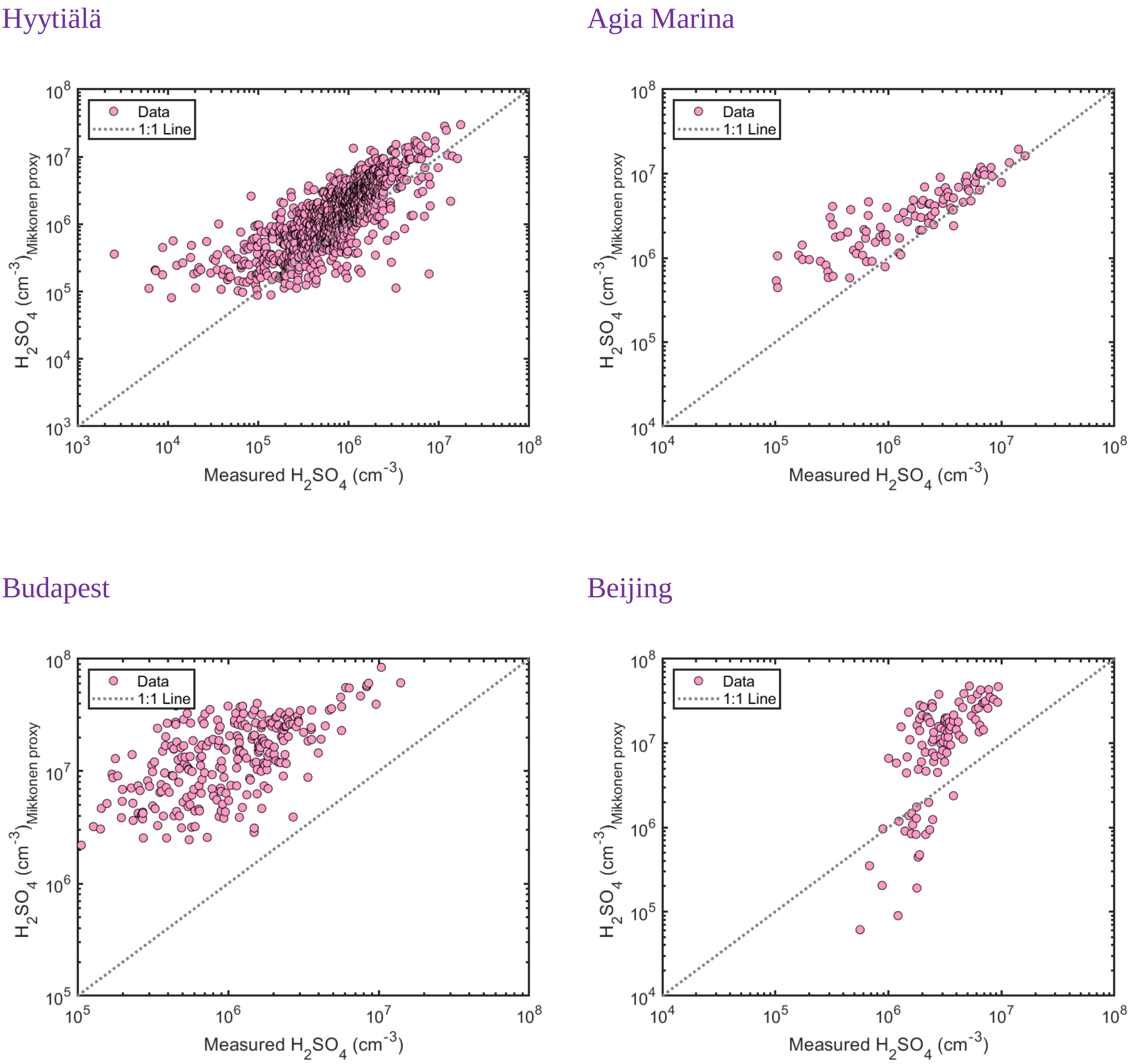


Figure R 3 Scatter plot showing the correlation between measured sulphuric acid and the sulphuric acid concentrations derived from the Mikkonen et al. 2011 proxy at the 4 locations during daytime (GlobRad >= 50 W/m^2): Hyytiälä, Agia Marina, Budapest and Beijing.

As minor issues:

1. Line 27: Just to be slightly fussy with wording, H2SO4 is important in new particle formation for actually two reasons: it has low volatility and also has strong intramolecular bonding abilities. Merely mentioning low volatility misses qualities that make this compound special.

We agree with the reviewer that H_2SO_4 is distinct for its strong hydrogen bonding ability which makes it possible to interact with other species and is found to be important for the first step of cluster formation. We have modified the relevant sentence on Line 58 to the following:

Sulphuric acid (H_2SO_4), which has a very low saturation vapor pressure and strong hydrogen bonding capability (Zhang et al., 2011), has been found to be the major precursor of atmospheric NPF (Weber et al., 1996; Kulmala et al., 2004; Sihto et al., 2006; Sipilä et al., 2010; Erupe et al., 2011; Lehtipalo et al., 2018; Ma et al., 2019) and is often used in global models for simulating the occurrence and intensity of new particle formation events.

2. Line 64: I suggest that the authors put a sentence or two here to state why it is important to develop a proxy for H2SO4. Many readers may be aware of the reason but it's a small thing to do and will be a great benefit to those who would otherwise be left wondering why so much effort is being placed in this.

We added the following sentences as per recommendation from the reviewer:

Line 60: *Sulphuric acid (H_2SO_4), which has a very low saturation vapor pressure, has been found to be the major precursor of atmospheric NPF (Weber et al., 1996; Kulmala et al., 2004; Sihto et al., 2006; Sipilä et al., 2010; Erupe et al., 2011; Lehtipalo et al., 2018; Ma et al., 2019) and is often used in global models for simulating the occurrence and intensity of new particle formation events (Dunne et al., 2016).*

and to Line 80:

Besides the abovementioned-previously-developed proxies, an additional proxy is still needed for representing nighttime periods which were not considered previously.

3. Line 75: I notice that Dr. Mikkonen is a reviewer of this article, so perhaps he will make this point (and I hope he also raises the concern that I express above). While the statement that his parameterization does not include condensation sink it technically correct, I believe that he considered this in his statistical analysis and found that condensation sink, or rather higher aerosol loading, is associated both with the source and sink of H2SO4, and that is the reason why on average it does not appear in the parameterization. If true then perhaps more accurate to state it this way rather than to leave the reader to conclude that this model overlooked the potential role of condensation sink.

We did not intend to say that Mikkonen et al. (2011) have overlooked the potential role of condensation sink, we have however referred to their sentence in the abstract copied below.

Sentence from Dada et al. 2020: "Proxies developed by Mikkonen et al. (2011) suggested that the sulphuric acid concentration depends mostly on the available radiation and SO_2 concentration, with little influence of CS."

Sentence from Mikkonen et al. 2011: "Interestingly, the role of the condensation sink in the proxy was only minor, since similarly accurate proxies could be constructed with global solar radiation and SO2 concentration alone."

4. Line 86: I suggest you choose a better word than "goodness"

We modified the sentence to the following:

In order to evaluate the accuracy of the our hypothesized sources and sinks and derive the proxy equations goodness of our new proxy, we utilize measurements from four different locations: (1) Hyytiälä, Finland, (2) Agia Marina, Cyprus, (3) Budapest, Hungary and (4) Beijing, China, representing a semi-pristine boreal forest environment, rural environment in the Mediterranean area, urban environment and heavily polluted megacity, respectively. To evaluate the predictive power of the derived proxies, the equations are further tested on independent data sets.

5. Line 249: this reference to Petaja paper seems strange. Why wasn't standard referencing used is referring to Equation 7 in the text (e.g., on line 245)?

We thank the reviewer for noticing; we modified the related text to the following:

We also refitted the data using the simple proxy proposed by Petäjä et al. (2009) by excluding the formation of sulphuric acid via stabilized Criegee intermediates source pathway and loss of sulphuric acid via the cluster formation pathway using Equation 6 and evaluated it by comparing to the original Petäjä et al. (2009) proxy using Equation 7.

$$\frac{d[H_2SO_4]}{dt} = k_1 GlobRad[SO_2] - CS[H_2SO_4] \qquad (1)$$

$$\frac{d[H_2SO_4]}{dt} = 1.4x\ 10^7 x\ GlobRad^{-0.7}[SO_2]\ GlobRad - CS[H_2SO_4] \qquad (2)$$

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