

Interactive comment on “A proxy for atmospheric daytime gaseous sulfuric acid concentration in urban Beijing” by Yiqun Lu et al.

Anonymous Referee #2

Received and published: 19 December 2018

The manuscript by Lu et al. evaluates different equations for the calculation of gas-phase sulfuric acid from proxy parameters (like SO₂, UVB radiation, condensation sink, etc.). Different sets of parameters are tested and the performance of the proxy equations is evaluated against in-situ measurements of gas phase H₂SO₄ (measured with a chemical ionization mass spectrometer) and the measured proxy parameters. The measurements took place in Beijing during February/March 2018. While similar proxy expressions for sulfuric acid have been derived for other locations (see Mikkonen et al., 2011, ACP, 11(21), 11319–11334) this is the first one applying such methods for the conditions of a Chinese mega city. Unlike previous studies, the present one includes also HONO as an OH precursor and not just ozone. This leads to a slightly better correlation coefficient between the proxy-derived and measured H₂SO₄. The

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manuscript is scientifically relevant and should be published in ACP after the authors have considered the points listed below. Besides the general comments, I have also listed a number of suggestions to improve the language.

Comments:

(1) Page 1, line 25: “remains a major challenge” is a bit exaggerated, please revise this statement

(2) Page 2, line 43: How is it known that HONO is formed heterogeneously? Isn't the gas-phase reaction between OH and NO also an efficient source?

(3) Page 6, line 169: Is the upper size limit of 700 nm sufficient to include all relevant particles contributing to the condensation sink? During dust events, larger particles can probably contribute quite significantly to the CS.

(4) Page 8, line 231 to 234: It should be explained in more detail how k is calculated and how it relates to k_0 .

(5) Page 9, line 281 to 284: Could the correlation between HONO and NO₂ also be caused by the gas phase reaction between OH and NO (because NO correlates probably strongly with NO₂)?

(6) Page 10, line 317 to 319: The mean absolute error does not seem to be the best metric for evaluating the fitting of data that vary over more than one order of magnitude. This can lead to a bias where the high values are well represented but the relative deviation for the small values can be large. A better metric could be the average ratio (sum over all $\max(V_{i_measured}, V_{i_fitted}) / \min(V_{i_measured}, V_{i_fitted})$), where V_i is a data point for the considered parameter, e.g. [H₂SO₄]).

(7) Page 11, line 346/347: The improvement relates to an increase of R from 0.85 to 0.86; this does not justify the word “significantly”; “improved slightly” instead of “improved significantly” is more appropriate.

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(8) Page 13, line 396: Aren't the different values of CS taken into account in the calculation, or do the authors mean that the different exponents for CS make the difference? What is the exponent for the CS in the Petäjä et al. study?

(9) Page 14, line 439: How can the proxies be evaluated for past measurements? If measurements exist for both sulfuric acid and the proxy parameters, these should be included in the present study.

(10) Page 14, line 440: The importance of heterogeneous chemistry cannot be concluded from the presented data; this statement should be removed or supported with further data.

(11) Section 5: Discussion on the expected seasonality of the proxy-H₂SO₄ relation should be discussed. Very likely, the exponents can have different values for other meteorological condition, e.g., when desert dust contributes to the particle burden. In such a case, the exponent for the CS could become negative. This possibility should be mentioned/discussed.

(12) Table 1: The values for ozone are quite low. Are these low values typical for wintertime conditions in Beijing?

(13) Table 3: What is the definition of the scaling factor k_0 ? What are its units?

(14) Figure 3: The yellow areas are hardly visible; it would probably be better to use colored lines instead of filled areas for this figure. In addition, the shape of the curves suggests a pronounced bias (the high values are on average underestimated, while the low values are overestimated). This bias can also be seen in the SI Figures. Using a different metric for the fitting (ratios instead of absolute differences, see above) could solve this issue.

Language:

Page 2, line 46: "less than" rather than "up to"?

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Page 3, line 57: “involve” instead of “demand”

Page 3, line 73/74: delete “using a passive CIMS”

Page 3, line 76: delete “associated equilibrium or”

Page 3, line 79: replace “will be” with “are”

Page 4, line 90: replace “supposing” with “assuming”

Page 4, line 102: delete “, a proxy for condensational sink”

Page 4, line 108: delete “that characterized“

Page 4, line 114: replace “between” with “in”

Page 6, line 156: “the sample flow”

Page 6, line 177: “dependent on” instead of “that could be defined as a function of”

Page 8, line 230: What is meant by the symbol ξ ?

Page 9, line 255: “in the 5-95% percentile range” instead of “was in . . . range of”

Page 9, line 280: delete “an”

Page 10, line 290: “lamination” does not seem to be the right word here, maybe better to use “layering with”

Page 10, line 305: “has” instead of “have”

Page 11, line 352: either delete “are tended to” or use “tend to”

Page 12, line 382: Do the authors mean “deviations” instead of “derivations”?

Page 13, line 390: “when averaged to a time resolution of 5 min” instead of “with a time resolution of 5 min”

Page 13, line 411: Please provide the units for the parameters in this equation.

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Page 14, line 418/419: Please provide the units for the parameters in this equation.

Page 14, line 425/426 Please provide the units for the parameters in this equation.

Page 14, 433/434: Please provide also the equation numbers and not just the proxy numbers (N2 and N7).

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