Response to the comments of referee #1:

We thank the reviewer for their helpful comments. The referee's comments are first given in black type, followed by our response to each in turn in blue type. Any changes to the manuscript in response to the comments are then given in quotation marks in red type and the line number isin the Microsoft-word version of revised MS without revision. The line number may be different in the PDF version, so please see the section number mentioned in the Response. Figure Response 1 and Figure Response 2 only appear in the Responses and not the revised MS, in order to illustrate the responses to the referee comments.

We have now added D. Moon, M. Baeza-Romero and D. Heard as co-authors to this paper since their unpublished experimental data have been included in this paper and they have contributed to enhance the manuscript.

Comment:

1. Please clarify how the ionic strength in Equation 4 is calculated. Is this calculated in the MARK model and what would a typical value be?

Response:

The ionic strength (I, mol L^{-1}), is calculated in the model via Equation 8 in section 2.2.2 on line 124. Typical values for I are in the range of 2.16 – 17.75 mol L^{-1} based on the ion concentrations in the aerosol bulk and the RH ranging from 40% - 90%.

Comment:

2. For all tables please add units where these are missing.

Response:

We added the units in Table 2, 3, 4 and 5 which is now Table S. 1, S. 2, S. 3, and S. 4 in the Supplementary Information of the revised MS.

Comment:

3. In Table 4 what are the values of k_{mt} or how are these calculated?

Response:

We added more information about the definition and importance of k_{mt} in part 2.1 on line 78 around Equation (3):

   "An approach to combine both gas phase molecular diffusion and liquid phase interface mass transport processes is through one variable called k_{mt} (Schwartz, 1984;Schwartz, 1986), which is used in the calculation for gas-liquid multiphase reactions in many modelling studies (Lelieveld and Crutzen, 1991;Chameides and Stelson, 1992;Sander, 1999;Hanson et al., 1994). The definition of k_{mt} is:

$$k_{mt} = (\frac{R_d{}^2}{3D_g} + \frac{4R_d}{3v_{HO_2}\alpha})^{-1} \qquad (3)$$

   k_{mt} is used to connect the gas phase reactions and the aerosol condensed phase reactions. The rate of gas phase reactants (X) diffusing and dissolving to the condensed phase can be calculated in the framework of aqueous phase reactions as $k_{mt\_X} \times ALWC$ (where X is the reactant molecule). Moreover, the conversion rate of aqueous phase reactants to gas phase can be calculated as $\frac{k_{mt\_X}}{H^{cc} \times RT}$ where H^{cc} is the effective Henry's law constant [M atm^{-1}]. The unit of k_{mt} is s^{-1}, as k_{mt}

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contains the conversion from m_{air}^{-3} of the gas phase molecule concentrations to m_{aq}^{-3} of the aqueous phase molecule concentrations and backward. For larger particles (radius >1μm), k_{mt} is mainly determined by gas phase diffusion of HO_2. For smaller particles (radius <1μm) k_{mt} is mainly determined by the accommodation coefficient (α). The MARK model can simultaneously simulate gas and liquid two-phase reaction systems in the same framework."

The typical value of k_{mt} of HO_2 for small particles with the radius of 50 nm is 3.85×10^5 s^{-1}, and for larger particles with the radius of 1 μm is 1.93×10^4 s^{-1}.

Comment:

4. Are all reactions in the tables included in the model? If so how does this relate to keff in Equation 11?

Response: All reactions in the tables are included in the model. k_{eff} is the comprehensive reaction rate coefficient encompasses both HO_2 dissolution equilibrium reactions and liquid phase chemical-physical reactions during HO_2 uptake process. k_{eff} is mostly affected by the concentration of copper, the HO_2/O_2^- cycle in the liquid phase and HO_2 dissolution equilibrium. HO_2/O_2^- reacting with Cu ions (R 1, R 2, R 8 and R 9) may directly affect k_{eff} thus change the model result of γ_{HO_2}. $OH_{(aq)}$, $O_{2(aq)}$, $O_{3(aq)}$, $H_2O_{2(aq)}$ will also influence the reaction system because they have direct relationship with the dissolution equilibrium and reactions of HO_2, O_2, H_2O_2 OH and O_3. H_2O_2 is also a reaction product of HO_2/O_2^- reacting with Cu ions, its concentration will also affect the reaction system. Sulfate ion, ammonium ion and nitrate ion may not directly affect the system, while they will change the concentration of aqueous HO_2, O_2, H_2O_2, OH, O_3 and their solubility. What ismore, HO_2 uptake process may influence the oxidation rate of SO_2 and other reagents, to which more research is needed.

Comment:

5. It's stated that in the model it is assumed that the surface concentration and the bulk concentration equal each other. It is also stated that this is only valid for particles with a radius less than 200 nm. However, the model and resulting parameterization are then applied to particles which are larger than this and many particles in the atmosphere are larger than this. I wonder why the authors don't seem to have used the correction in equations 10 and 11 and what impact this will have on their final results and the applicability of their parameterization to future studies?

Response:

The size of the particles is an important factor within the MARK model when considering gas phase diffusion to the particle surface and HO_2 desorption. While the HO_2 radical concentration is not a factor that influence γ_{HO_2} in the MARK model.

In ambient urban situations, the Count Median Diameter (R_d) of aerosol particles is smaller than 1μm in most instances. The ratio of $\overline{[HO_2]}$ to $[HO_{2(r)}]$ is 0.89 calculated by the MARK model simulation of k_{eff} with a RH 40%-90%, the copper ion concentration varies from 10^{-5} to 1M at 1μm diameter particles. The ratio will be even higher and close to 1 with smaller particles (>0.95 at 400 nm diameter). The MARK model is valid for particles with R_d smaller than 400 nm and may cause small deviations for particles with R_d smaller than 1μm. Thus, in this paper, we assume the surface concentration of HO_2 equals to the condensed phase average HO_2 concentration.

A model considering the influences of aerosol particle size distribution and HO_2 concentration gradients on γ_{HO_2} is currently under development.

We changed the statement in the origin MS in part 2.2.3 on line 148 and modified as: "In the copper-doped aerosol particle, because of the high value of k_{eff} and small Count Median Diameter (R_d) (usually smaller than 1μm), the ratio is close to 1. At a diameter of 1μm, and a relative humidity between 40% and 90%, the condensed phase copper ion concentration varies from 10^{-5} to 1M, the average ratio of the surface HO_2 concentration and the condensed phase HO_2 concentration is 0.89. At 400nm diameter particles for RH = 40% to 90%, the ratio is larger than 0.95. The ratios are calculated by simulation of k_{eff} and the accordingly calculations by Equation (12) and (13)."

Comment:

6. In Figure 2 what is the main cause for the decrease in the uptake coefficients between the original parameterization and the new model results. Is the difference mainly due to the different rate coefficient being used, the use of activity coefficients or something else?

Response:

The parameterization proposed by the IUPAC uses only one rate constant as the second-order reaction rate k^{II} of Cu^{2+} and HO_2. We use 1.5×10^7 L mol^{-1} s^{-1} as the secondary reaction rate k^{II} rather than the more commonly used value of 1×10^9 L $mol^{-1}s^{-1}$ in the calculation of the original parameterization.

We added the reason of using 1.5×10^7 L mol^{-1} s^{-1} in the calculation in the revised MS in Section 3.2 on line 241:

"The prior value (1.5×10^7 M^{-1} s^{-1}) reflects the rate of reaction between HO_2 and Cu^{2+}, more prevalent in acidic aerosol such as ammonium sulphate, and the latter between O_2^- and Cu^{2+} ions, which is more prevalent in aerosols with a pH greater than the pK_a of HO_2, such as NaCl (Bielski et al., 1985). This treatment within the calculation can bring predictions more in line with experimental results (Figure 2 grey dotted line) compared to the high value of 1×10^9 L $mol^{-1}s^{-1}$ used in the existing parameterized equation."

And on line 256 the main reason of the differences between original parameterization and the MARK model:

"The main reason for the differences between the original parameterization and the MARK model is the effect of including the activity coefficients of Cu ion and HO2 and the effects of reactions of different valence states of copper ions."

7. For Figure 2 what is the aerosol pH and how is it calculated?

Response:

The main components of the aerosols used in the laboratory measurements of γ_{HO_2} shown in Figure 2 are ammonium sulfate and a small amount of copper sulfate. According to the calculation based on the aqion 7.0.8 interface (for details please see https://www.aqion.de/), the pH is around 4.54 considering aerosol dehydration with 2M aerosol bulk sulfate concentration and 1M Cu ion at 25°C. In the MARK model, we set aerosol pH as 4.5 when compared to the laboratory results. We have re-calculated γ_{HO_2} at pH=4.5 and added data points from Lakey et al., JPCA (2016) based on the short comments from Pro. Heard. In the revised MS, Figure 2 is modified as follows:
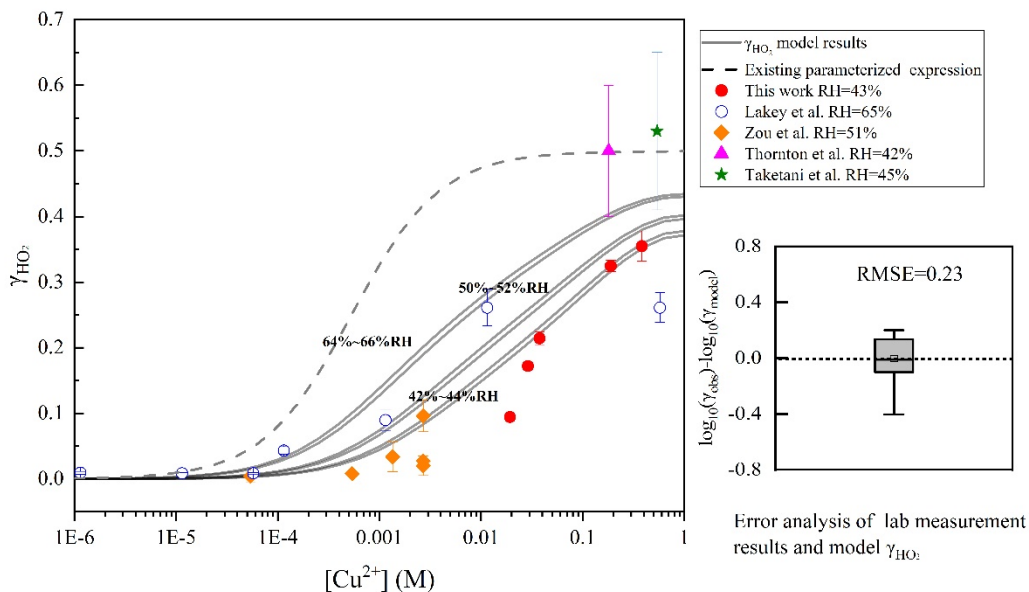
"Figure 2: Dependence of γ_{HO_2} on aerosol copper concentration. Red filled circles denote the results at 43% *RH* measured at Leeds. Blue hollow circles at 65% *RH* (Lakey et al., 2016). Yellow filled diamonds denote results at 51% *RH* (Zou et al., 2019), filled purple triangle at 42% *RH* (Thornton and Abbatt, 2005)) and filled green star at 45% *RH* (Taketani et al., 2008)). The grey dotted line denotes the current parameterized equation (Thornton et al., 2008;Hanson et al., 1992;Hanson et al., 1994;Jacob, 2000;Kolb et al., 1995) and the solid grey lines represent the model results of MARK model in this study at various *RH*. The root mean square error (RMSE) between the MARK modelled values and the full dataset (0.23). Aerosol pH is set as 4.5 based on aqion 7.0.8 interface considering the participation of Cu ion (for details please see https://www.aqion.de/)."

Comment:

8. Figure 2 seems to be missing some previously published data point(s) from Lakey et al., JPCA (2016). It seems that the point at the highest copper concentration in that work (which is not shown in Figure 2) would not fit the modeled line. The authors should include any previously published missing points for completeness. Are they able to model or at least speculate as to why this data point does not fit their model.

Response:

We have now added these data points from Lakey et al., JPCA (2016) at the highest and the lowest copper concentration used to Figure 2 for completeness. Please also see the response to the Short Comment from Professor Heard, University of Leeds. Prof. Heard, Dr. Moon and Dr. M. Baeza-Romero from Leeds are now added as co-authors to the paper, with the correct data from Lakey et al., (2016) used for *RH*=65% and from the PhD thesis of Dr. Moon for *RH*=43% included. Moreover, in order to model the results from Lakey et al., JPCA (2016), we change the accommodation coefficient to 0.26 in the MARK model as they recommended in the paper, and get the result as follows:
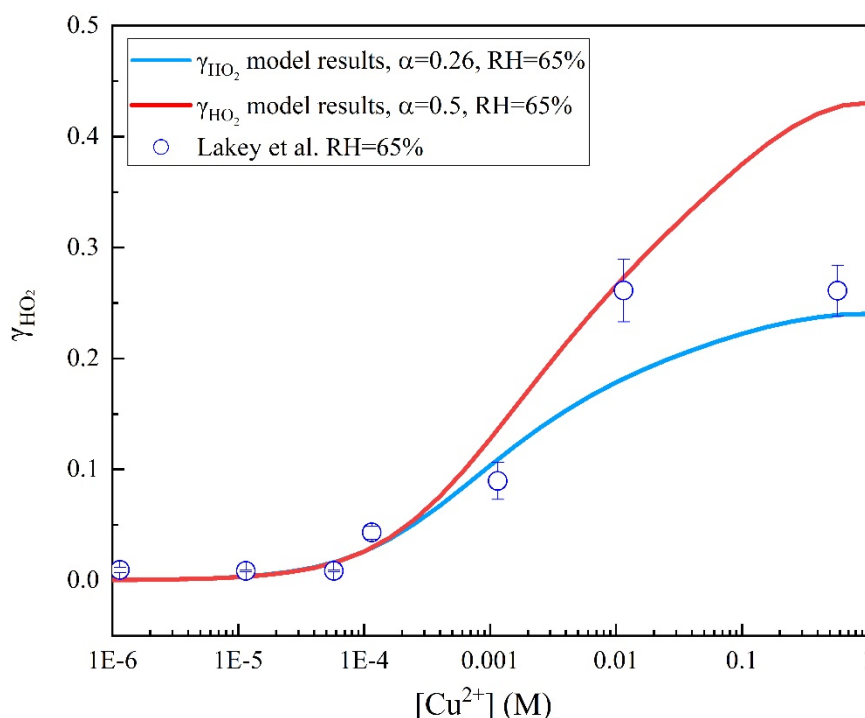
Figure Response. 1 Comparison of the dependence of γ_{HO_2} on aerosol copper ion concentration with α_{HO_2} as 0.26 and 0.5 at 65% *RH*. The solid line denotes the MARK model results, and the blue hollow circles denote the results at 65% *RH* measured by Lakey et al. (2016).

Model results using α_{HO_2} as 0.5 fits well with the results from Lakey et al. (2016) when $[Cu^{2+}]$ smaller than 0.1M while have larger deviation for higher $[Cu^{2+}]$. α_{HO_2} as 0.26 fits unsatisfactorily at $[Cu^{2+}]$ around 0.01M. Considering the ambient aerosol condensed phase $[Cu^{2+}]$ (for example, ranging from 0.003 to 0.012 M in Wangdu campaign), in this MS we still use α_{HO_2} as 0.5 to get the novel HO$_2$ uptake parametrization.

The accommodation coefficient may change in the process of the experiments for some reasons. For example, the reaction time and HO$_2$ initial concentration (see next comment by the reviewer). Aerosol phase state is also an important factor influenced α_{HO_2}. Moreover, during the efflorescence of aerosol particles in drying nafion tube, different concentrations of copper ions may have a certain effect on particle phase state which will influence α_{HO_2}. This part should be further studied.

9. George et al. PCCP (2013) noticed higher uptake coefficients for lower HO$_2$ concentrations for copper doped particles. Did the authors do any sensitivity tests with different HO$_2$ concentrations and do they see any difference?
Response:
The γ_{HO_2} measured in the flow tube experiments indeed depends both on the HO$_2$ concentration and also the reaction time between HO$_2$ and the aerosols.. However, sensitivity tests in the MARK model show no γ_{HO_2} decreasing trend with increasing $[HO_2]_0$ in the absence of Cu ions, and γ_{HO_2} will slightly increase with the $[HO_2]_0$ in the presence of Cu ions in the MARK model.

A possible explanation for the results from George et al. PCCP (2013) could be the Fenton-like reactions of Cu ions and H_2O_2 that is an additional source of HO_2. More H_2O_2 will be generated with greater light intensity and may accumulate along the flow tube with the reaction of HO_2 with aerosol for H_2O_2 is one product of HO_2 uptake. However, H_2O_2 and Cu ions reactions need to be of the same order of magnitude or no more than one magnitude lower than that of HO_2 reaction with Cu ions to make obvious differences in the measurement of γ_{HO_2}. In the MARK model, H_2O_2 only reacts with Cu^+ and the reaction rate constant is 7×10^3 L mol^{-1} s^{-1} which is too small to influence γ_{HO_2} with the changes of H_2O_2 concentration, and so it may explain the lack of sensitivity of the uptake coefficient with HO_2 concentration.

10. Figure 3: Is lg on y axis log10?

11. Figure 3: Please explain this figure in a more detailed fashion. It is unclear to me what the markers are and why there is a range of values. Why does there seem to be a larger difference between the model and the parameterization at low relative humidity?

Response:

We have changed the y-axis label in accordance with the referee´s comment in Figure 3 in the revised MS. We calculated the RMSE of γ_{HO_2} predicted by MARK and the corresponding calculated values from the new parameterized equation at different *RH* and Cu ion concentrations.

We added the explanation of the larger difference between the model and the parameterization at low relative humidity in line 290 on the second last paragraph of section 3.3 in the revised MS:

"The range of values shows the difference between the modeled data and parameterized equation data at different Cu concentration. At low *RH* and consequently relatively low ALWC, γ_{HO_2} is more sensitive to $[Cu^{2+}]$ expecially at low $[Cu^{2+}]$ ($<10^{-4}$M). This sensitivity can not be fully represented in the parameterized equation. What is more, at low $[Cu^{2+}]$ and low *RH*, the value of γ_{HO_2} is smaller than in other conditions, small changes of γ_{HO_2} will cause larger RMSE values. "

Comment：

12. Figure 4: Is the data shown measurements or a simulation?

13. Figure 5: This figure is not mentioned at all in the text and as such I don't know what the difference is between Figures 4 and 5.

14. Why not combine Figures 4 and 5 for better comparisons?

Response：

Figure S. 1 shows the averaged particle surface-area size distribution (PSASD) and the particle number size distribution (PNSD) of aerosol measured in the Wangdu field campaign. We deleted Figure 5 in the original MS because the information of aerosol size distribution is redundant. In the SI of the revised MS, we have added Figure S.1 as follows:
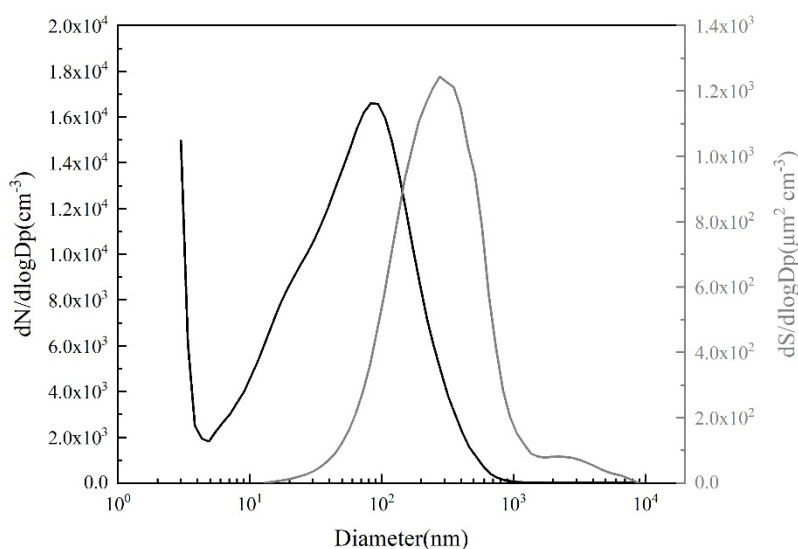
Figure S.1: The dry-state particle number size distribution (PNSD) (black line) and particle surface-area size distribution (PSASD) (grey line) of aerosol for conditions encountered during the Wangdu field campaign.

15. Can the authors speculate as to why the HO_2 uptake coefficient is higher at night (Figure 6)?

Response:

We add the table below in the SI for revised MS as Table S. 5. Table S.5 shows the median and average values of the copper concentration, PM loading and *RH* during the day and at night. During the day the copper concentration is larger, but the lower *RH* may limit γ_{HO_2}. Thus, k_{het}, the quasi-first order reaction rate constant of HO_2 heterogeneous uptake is slightly higher at night compared to the daytime, contributing to the higher γ_{HO_2} predicted at night.

Table S.5. The median and average values used in the calculation of γ_{HO_2} in Wangdu

	Value	Cu [ng/m^3]	PM$_{2.5}$ mass [μg/m^3]	*RH* [%]	γ_{HO_2}	k_{het}[s^{-1}]
Day	median	33.42	77.9	55.4	0.119	0.017
	average	44.66	85.0	57.6	0.126	0.020
Night	median	19.01	70.6	68.9	0.134	0.021
	average	34.16	67.9	67.4	0.147	0.023

16. In Figure 6 what is the main cause of the distribution in HO_2 uptake coefficients? Is it due to different copper concentrations in the particles or something else?

Response:

The distribution of γ_{HO_2} is mainly due to the different copper concentrations and ambient *RH*. Although the PM mass is also a parameter in the empirical equation proposed by this MS, it shows small partial correlation on γ_{HO_2}. Figure S 2 below shows the partial correlation coefficient between γ_{HO_2}, field measured [HO$_2$], [OH], $TR_{HO2uptake}$ and R_1 with aerosol mass loading in Wangdu

campain. This figure is now added in the SI of the revised MS.
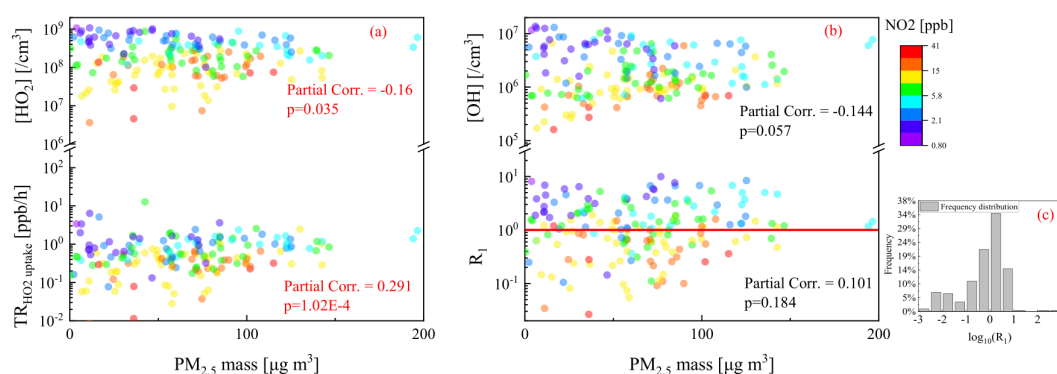
"Figure S.2. Impact of the HO_2 uptake evaluated with the novel empirical equation for conditions encountered during the Wangdu field campaign. Partial correlation of logarithmic values of $TR_{HO2uptake}$ and R_1 with respect to aerosol loading were calculated. The partial correlation coefficient in panel (a) means that $TR_{HO2uptake}$ has a small partial correlation with aerosol loading. No partial correlation of R_1, $[HO_2]$ and $[OH]$ to aerosol loading is observed. The different coloured dots show different $[NO_2]$. Panel (c) is the distribution of $log_{10}R_1$."

17. Were any HO_2 measurements made during the Wangdu field campaign and if so was any box modeling of the Wangdu campaign performed to determine whether there was a discrepency between measured and modeled HO2 uptake coefficients? Were predicted HO2 uptake coefficients in the range that was expected? If no HO2 measurements were made, could the authors clarify why they chose this particular field campaign to apply their model to?

Response:

Yes, HO_2 and other radical concentration measurements were made during the comprehensive campaign in Wangdu.

We added the following statements in the revised MS in section 3.4.3 the second paragraph on line 347:

"Tan et al. (2017) had compared the measured and modelled OH, HO_2 and RO_2 radicals in the Wangdu campaign. However, in this paper, they did not discuss the influence of HO_2 uptake. A very recent publication (Tan et al., 2020) calculated γ_{HO_2} in the Wangdu campaign based on the comparison of field measurement data for HO_2 and concentrations calculated by the box model. The paper proposes that all γ_{HO_2} calculated in this way from the Wangdu campaign can be fitted to a Gaussian distribution around the value of 0.08 ± 0.13. This value is similar to our estimation in this paper considering the indirect measurement uncertainty (please see the SI)."

What is more, we recalculated the ratio ($R1$) of HO_2 uptake to ROx termination rate with measured RO_2 concentration which is now in consist to the dataset of (Tan et al., 2020). In the original MS, we used the modeled RO_2 radical concentrations which is underestimated compared to the measured results. Thus, $R1$ decreases obviously due to the competition of larger proportion of RO_2+HO_2, RO_2+NO and RO_2+RO_2 reactions in the ROx radical termination budget.

18. Line 313: The authors may want to clarify that 'aerosol properties' may include phase state and that previous measurements have shown lower uptake coefficients for semi-solid and solid particles (e.g. Lakey et al. ACP (2016)). The authors should also clarify somewhere that one of the major limitations of their model is that they assume steady-state concentrations and do not consider concentration gradients which will occur and could change over time for semi-solid particles.

Response:

We changed the word "properties" as "aerosol particle condensed phase component concentrations" in the revised MS in Part 2.2.2 on line 116 at the first paragraph and Part 3.4 on line 314 at the second paragraph to avoid the inaccurate statement of aerosol properties including phase state that is not considered in the MARK model used in this MS.

We added the following statement in the revised MS in section 4 on line 377:

"The novel empirical equation is applicable under the assumption of steady-state concentrations and with metastable or liquid aerosol particles (if the ambient *RH* over a completely liquid aerosol decreases below the deliquescence *RH*, the aerosol may not crystalize immediately but may constitute a supersaturated aqueous solution (i.e., in the metastable state) (Song et al., 2018)). The approximate calculation of HO_2 concentration gradients within the aerosol particle condensed phase also cause deviations for larger particles. The bulk diffusion coefficient of HO_2 and other reactive molecules should be lower in the situation of semi-solid particles (Berkemeier et al., 2016;Shiraiwa et al., 2010;Mikhailov et al., 2009) and would change with the water activity and the organic components (Price et al., 2015). This aspect needs further studies. For crystalline or amorphous solid aerosol particles, HO_2 will undergo surface reactions and diffuse across the surface rather than be accommodated within the aerosol bulk. The MARK model has limitations in the calculation of γ_{HO_2} with semi-solid aerosol particles. In the Wangdu campaign, κ_{sca} (optical aerosol hygroscopicity parameter) ranges from 0.05 to 0.35 with an average of 0.22. The ambient *RH* during the Wangdu campaign shows significant diurnal variations and varies greatly from 15% to 97%, with an average value of 61% (Kuang et al., 2019) indicating that the percentage of solid aerosol particles is relatively low and hence do not significantly influence γ_{HO_2}.
"

19. The authors fix the solubility of copper at 25%. In reality solubility can vary considerably. How sensitive is this parameter in their model?

Response:

The MARK model is sensitive to Cu ion concentrations that exceed 1×10^{-4} M. We tested the sensitivity of soluble copper ion concentration in the Wangdu campaign between the value of 10% to 70% (Fang et al., 2017;Hsu et al., 2004;Hsu et al., 2010). γ_{HO_2} will increase 1.57 times from 0.075±0.031 at 10% solubility to 0.193±0.079 at 70% solubility based on the GaussAmp fitting of data from Wangdu campaign. Even at 70% solubility (which is unlikely true for most situations), the central value of γ_{HO_2} is lower than 0.2. The figure below shows the variation of the uptake coefficient with Cu ion solubility.
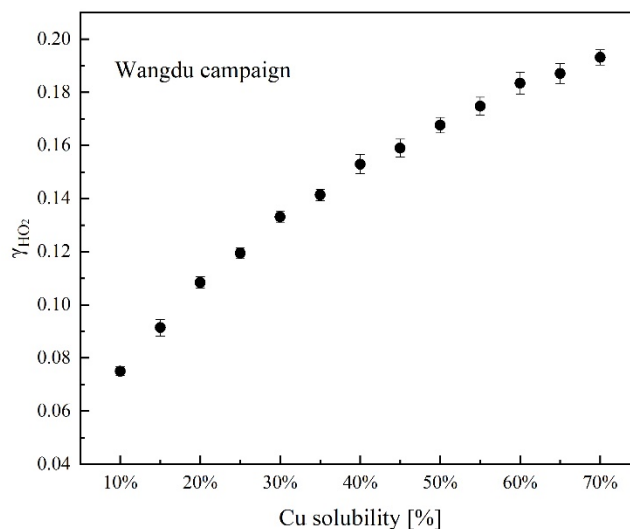
Figure Response.2 Sensitivity analysis of Cu solubility in the calculation of γ_{HO_2} for conditions encountered during the Wangdu campaign.

In the revised MS part 3.4.2 we discussed the influence factors of Cu solubility in the aerosol.

20. Another limitation of the model is that they don't consider reactions between different metal ions (such as Reaction 4 in Mao et al. ACP (2013)) which they have stated. However, could they also speculate how this could impact the estimated uptake coefficients for atmospheric aerosols (e.g. is it expected that this would increase the uptake coefficient)?
Response:
According to our understanding, there is no direct published laboratory measurement evidence of Cu-Fe redox coupling mechanism in HO$_2$ uptake. We speculate that the upper limitation of γ_{HO_2} may not change because of the low solubility of Fe and the influence of organic matters. However, whether the product of HO$_2$ uptake is H$_2$O$_2$ or H$_2$O will affect atmospheric oxidation capacity, as outlined by Mao et all (Mao et al., 2013). This should be studied in the future. In the revised MS Section 4, we discussed the influence of other TMI on γ_{HO_2}.

21. Please check the references carefully as many seem to wrong (e.g. Schwartz and Meyer 1986 line 108 and references in Figure 2).
Response:
We have checked the references in the updated manuscript, and the references in Figure 2 are also checked based on the Short Comment from Professor Heard (please see the first short comment). On line 43 in Section 1, line 79 in Section 2.1, line 142 in Section 2.2.3 and line 263 in Section 3.3, we changed the reference as Schwartz (1984) and Schwartz (1986). Moreover, we removed inaccurate references on line 33, 206 in the original MS and in Table S.3 and part S.1 in the revised MS. We also checked the references for the misuse of capitalization and subscripting.

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