Taketani et al. presents the measurement of HO_2 uptake coefficient for ambient aerosols at 298K at Mts. Tai and Mang in China. They show that the measured HO_2 uptake coefficient is significantly higher than the measured values in previous laboratory studies. To my knowledge this is so far the first attempt to measure HO_2 uptake coefficient from ambient aerosols, and therefore it adds valuable information to current knowledge of HO_2 uptake. However, this paper could be greatly improved by addressing the following issues.

1. Temperature dependence of HO_2 uptake coefficient. There are extensive discussions on the strong temperature dependence of HO_2 uptake coefficients (Thornton and Abbatt, 2005; Thornton et al., 2008; Mao et al., 2010), which suggest large increase of HO_2 uptake coefficient with decreasing temperature. And this is driven by the Henry's law constant for HO_2 and applicable to all aqueous aerosols. However, the current measurements in this paper were only conducted at 298 K. I think some measurements at lower temperature (such as 283K and 273K) would greatly strengthen this paper. Or at least the authors should make it clear that current measurements are likely the lower limit for ambient HO_2 uptake coefficient.

2. Aerosol sampling and preparation. This experiment first extracts the ambient aerosols (TSP for Mt. Tai and PM2.5 for Mt. Mang) with water. Then they generate aerosols using this extract from the filter with an atomizer. There are a few problems with this procedure. First, extracting the ambient aerosols with water (presumably pH = 7), will remove most dissolved Cu and Fe in ambient aerosols (pH is likely below 3) given that their pH-solubility relationship(Deguillaume et al., 2005). In other words, most dissolved Cu and Fe in ambient aerosols will not be atomized for subsequent HO_2 uptake experiments, and this will cause severe underestimate of HO_2 loss in the flow tube experiment. Second, extracting and regenerating aerosols will largely change the size distribution of aerosol trace metals. For example, if Cu is mainly in submicron aerosols (Lannefors et al., 1983) and total aerosol mass is largely in supermicron aerosols, this extracting will further dilute Cu concentration and therefore underestimate HO_2 loss rate for ambient aerosols (ambient aerosol surface area is mainly provided by submicron aerosols). Some discussions are needed on these problems.

3. Calculation of HO_2 loss in aqueous phase. In order to calculate HO_2 loss in such concentrated solution, each transitional metal ion has to be corrected by its activity coefficient for the non-ideal behavior (Ross and Noone, 1991). The resulting activity coefficient can be as small as 0.01 for Fe^{3+} and 0.05 for Cu^{2+}. Therefore the discussion of Cu and Fe forms in Page 13797 and 13798 must consider the correction by activity coefficients of Cu and Fe ions.

4. Catalytic cycle of Cu(I)/Cu(II) and Fe(II)/Fe(III). Cu and Fe both act as catalyst for HO_2 loss in aqueous phase. Therefore reaction (9) and reaction (10) are incomplete for

the catalytic cycle of Fe(II)/Fe(III), which is initiated by Fe(III) + HO_2/O_2^-. Cu(I)/Cu(II) is also initiated by Cu(II) + HO_2/O_2^-. Cu^+ and Fe^{2+} are only intermediate products of these catalytic cycles (actually the majority of dissolved Cu is Cu(II)). So Line 19 -21 in Page 13796 should be rephrased.

5. Gas-phase diffusion. I did a rough calculation for the following equation for standard aerosol uptake parameterization:

$$k = -\left(\frac{r_e}{D_g} + \frac{4}{v\gamma}\right)^{-1} A$$

where v is the mean molecular speed of the gas, D_g is the gas-phase molecular diffusion coefficient, γ is the reactive uptake coefficient for the gas, and A is the aerosol surface area per unit volume of air calculated from the mass concentration and r_e is the effective radius of that aerosol component.

It turns out when $\gamma(HO_2)$ =0.8 and r_e = 0.4 μm (this is likely a good estimate for the effective radius for sulfate when corrected by hygroscopic growth factor), the gas-diffusion term is equivalent to the interfacial mass transport term. In fact, the gas-phase diffusion can often be the limiting step when $\gamma(HO_2)$ approaches unity. Therefore the gas-phase diffusion term cannot be ignored for the calculation of ambient aerosols, but it could be negligible for rather small aerosols under laboratory conditions (mean surface-area-weighted radius 50-75 nm in this study). I think this should be addressed when applying the measured $\gamma(HO_2)$ to the calculations for ambient aerosols.

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