

Dear Dr. Mao,

We gratefully thank you for the review of our manuscript and enclose responses to the various comments and suggestions by the review.

The comments and suggestions are received and manuscript is revised. We would like to answer suggestions by reviews

### 1. Temperature dependence of HO<sub>2</sub> uptake coefficient.

We agree with the reviewer on the importance of temperature dependence. However, making additional experiments at lower temperatures is beyond the scope of this study. Thus we added the following sentences in Summary:

A few studies suggested that uptake coefficients of HO<sub>2</sub> radicals depend on temperature for aerosol particles with certain compositions (Thornton and Abbatt 2005, and Mao et al 2010). Further experiments on the temperature dependence would be needed for better representation of this process in chemical transport models.

### 2. Aerosol sampling and preparation.

### 3. Calculation of HO<sub>2</sub> loss in aqueous phase.

The actual free form Cu and Fe ions concentrations in the wet particles would depend on the following three factors, as mentioned by the reviewer. First, it is suggested that the solubility for Cu and Fe decrease with pH (Deguillaume et al., 2005). In this study, the pH of extracts for Mts. Tai and Mang were measured in ranges of 6.4–7.1 and 5.6–6.7, respectively.. From the viewpoint of chemical composition, ammonium is abundantly present for the studied particles, achieving almost full neutralization. Based on these facts, here we study uptake of HO<sub>2</sub> onto neutral particles, for which the pH could be reproduced at least roughly in the laboratory, although uptake onto acidic particles (pH < 3) might be another interesting issue, with higher solubility for Cu as the reviewer suggested.

Second, we have to consider the activity coefficients of Cu and Fe in the aqueous phase. These values are roughly estimated to be less than 0.1, based on (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> particles at 68% RH (Ross and Noone, 1991), suggesting the free form metal ions will be limited in the particles.

Third, the sampled particle size range might have affected the determined coefficient.

As suggested by the reviewer, our regenerated particles have trace metal concentrations averaged over the sampled size range and they are not weighted by the aerosol surface area size distribution. Thus it is important to examine if Cu concentration is dominant in submicron size range (Lannefors et al., 1983) or not. At Mt. Tai, on the contrary, Xu et al. (ACPD, 2009) suggested a relatively flat size distribution of Cu. For Mt. Mang, we made no size-segregated observations of trace metals. Hence this effect could be variable and we will mention this as possibility in the revised text.

Based on the considerations of these factors, we will mainly include discussion on the second effect (i.e., reduced activity) in the revised text, while the first and third points will also be mentioned as factors giving uncertainties.

We modified the results and discussion in Pages 13797 and 13798

#### 4. Catalytic cycle of Cu(I)/Cu(II) and Fe(II)/Fe(III).

We added more reaction processes for Fe(III). And we modified estimated accommodation coefficient and Fe level to achieve the  $\gamma$  values we measured in page 13798.

#### 5. Gas-phase diffusion.

We already considered the effect of gas-phase diffusion in the model calculations, by using the Fuchs-Sutugin equation. To explain this, we added the following sentences and equations in page 13800

We assumed that the reaction of HO<sub>2</sub> with aerosol does not have any gaseous products.



The first order decay rate  $k_r$  of equation (13) is determined by the Fuchs-Sutugin equation (Fuchs and Sutugin, 1970),

$$k_r = \int \frac{4\pi D r}{1 + K(r) \left( \frac{1.333 + 0.71 K(r)^{-1}}{1 + K(r)^{-1}} + \frac{4(1-\gamma)}{3\gamma} \right)} N(r) dr, \quad (14)$$

where  $r$  is the radius of the aerosol particle,  $K(r)$  is the Knudsen number for the aerosol particle radius,  $D$  is the gas-phase diffusion coefficient ( $D$ ) of HO<sub>2</sub> as  $\sim 0.25 \text{ cm}^2 \text{ s}^{-1}$  and  $N(r)$  is the number density distribution function of the aerosol.