Heterogeneous uptake of gaseous hydrogen peroxide on mineral dust

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ABSTRACT

The heterogeneous uptake processes of hydrogen peroxide on Arizona test dust and two types of authentic Chinese mineral dusts, i.e., Inner Mongolia desert dust and Xinjiang calciferous dust, were investigated using a Knudsen cell reactor coupled with a quadrupole mass spectrometer. The uptake coefficients were measured as a function of the initial concentration of H2O2 from 2.6 × 10¹¹ to 1.2 × 10¹² molecules/cm³, and the temperature dependence of the uptake coefficients was investigated over a range from 253 to 313 K. The concentration of H2O2 showed little effect on the uptake coefficients of these heterogeneous processes. As a function of temperature, the initial uptake coefficients decrease with increasing temperature, whereas the steady state uptake coefficients of Arizona test dust and Inner Mongolia desert dust increase with increasing temperature. Implications for the understanding of the uptake processes onto mineral dust samples were also discussed.

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Introduction

Mineral dust plays an important role as particulate matter in the troposphere. The amount of mineral dust injected into the atmosphere is about 1000–3000 Tg/year (Dentener et al., 1996; Usher et al., 2003). As one of the largest arid regions in the world, the Taklimakan Desert, Gobi Desert and loess areas are the main source of mineral dust in Asia. Mineral aerosols (at ~35%) in most circumstances are major components of the total aerosols in China (Zhang et al., 2012). Because wind-blown dust can transport long distances and has a long atmospheric lifetime, anthropogenic gas pollutants such as SO₂, NOₓ and O₃ can react over East Asia by interacting with mineral dust and producing secondary pollutants in this region.

In the atmosphere, as the precursor of odd-oxygen and reservoir of odd-hydrogen radicals (Lee et al., 2000), hydrogen peroxide is an important secondary photochemical product, which is related to the bimolecular recombination of hydroperoxyl (HO₂) radicals (Hua et al., 2008; Jackson and Hewitt, 1999; Reeves and Penkett, 2003). The self-reaction of two hydroperoxyl radicals to form hydrogen peroxide has a particularly high reaction rate in photochemical smog. In the cloud aqueous phase, hydrogen peroxide is an important oxidant, which can oxidize sulfur dioxide to sulfuric acid (Husain et al., 2000). Hydrogen peroxide has also been used in the manufacturing industry field, for example as an antiseptic, disinfectant, or detergent (Vione et al., 2003). Normally, photolysis, reaction with OH, wet or dry deposition can remove the hydroperoxide radicals and lower the oxidizing capacity of the atmosphere (Rubio et al., 2006). However, in field measurement studies, gaseous H₂O₂ can be observed, which means that hydrogen peroxide is more stable in the gas phase (Bales et al., 1995; Jacobi et al., 2002). Field studies have
also shown that gaseous H$_2$O$_2$ can react with ambient aerosols through heterogeneous reactions, which may be an important sink for H$_2$O$_2$ (de Reus et al., 2005; He et al., 2010).

Therefore, recently several laboratory studies have investigated the kinetics and mechanisms of H$_2$O$_2$ interactions with mineral aerosol surfaces, making it possible to reduce the discrepancies between field-measured and modeled H$_2$O$_2$ concentrations (El Zein et al., 2013, 2014; Pradhan et al., 2010a, 2010b; Romanias et al., 2012, 2013; Wang et al., 2011; Zhao et al., 2011, 2013; Zhou et al., 2012). However, most of the available data concern the reactions of H$_2$O$_2$ with model mineral oxides. Study on the reactivity of realistic mineral dust toward gaseous H$_2$O$_2$ is rather scarce and laboratory investigation on the combined action of hydrogen peroxide and other contaminants in the gas phase on aerosol surfaces is also very limited (Chu et al., 2000; Clegg and Abbatt, 2001a, 2001b).

Due to the scarcity of experimental measurements of kinetic data for important atmospheric processes and how these data vary with temperature, much uncertainty still remains in atmosphere science. This work presents a detailed investigation of the uptake coefficients of hydrogen peroxide on three types of mineral dust particles (Arizona Test Dust, Inner Mongolia desert dust and Xinjiang sierozem), which was motivated by a desire to mimic the behavior of mineral aerosol particles that are generally present in the atmosphere, over the temperature region from 253 to 313 K. The temperature dependence of these uptake processes and atmospheric implications of these reactions are discussed in detail in the following section. Through these series of experiments, we have been able to probe the effects of H$_2$O$_2$ as an important oxidant on mineral dust particles. The present studies provide useful information to understand the mechanisms of heterogeneous processes of hydrogen peroxide on these mineral particles. The experimentally determined data will also contribute to model studies of the atmosphere.

1. Experimental section

1.1. Materials

Arizona test dust with a nominal 0–5 μm size used in this study was purchased from Powder Technology Inc. (Powder Technology Inc., USA). Inner Mongolia desert dust and Xinjiang sierozem were purchased from the Chinese standard material center (the Chinese standard material center, China), consisting of <75 μm diameter fractions representative of mineral dust from the Inner Mongolia desert and Xinjiang arid region in the northwest of China. A Quantachrome Autosorb-iQ, Quantachrome Instruments, USA) using multipoint Brunauer–Emmett–Teller (BET) analysis was applied to measure the surface areas of these powders. The BET areas were determined to be 5.30 m$^2$/g for Arizona test dust, 5.06 m$^2$/g for Inner Mongolia desert dust, and 20.98 m$^2$/g for Xinjiang sierozem. The main fractions of the mineral dust samples are quartz and feldspar. The certified chemical compositions of the mineral dust reference materials are listed in Table 1.

Aqueous solutions of H$_2$O$_2$ (35 wt.%, Alfa Aesar, China) were prepared as described in our previous work (Zhou et al., 2012). Before using, solutions were concentrated to greater than 95% by weight percentage by drying.

### Table 1 – Certified reference materials for the chemical composition of mineral dusts.

<table>
<thead>
<tr>
<th>Main composition</th>
<th>Arizona test dust (%)</th>
<th>Inner Mongolia desert dust (%)</th>
<th>Xinjiang calciferous dust (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO$_2$</td>
<td>68–76</td>
<td>78.30 ± 0.33</td>
<td>60.3 ± 0.41</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>10–15</td>
<td>9.65 ± 0.09</td>
<td>11.96 ± 0.09</td>
</tr>
<tr>
<td>Fe$_2$O$_3$</td>
<td>2.0–5.0</td>
<td>2.07 ± 0.03</td>
<td>4.07 ± 0.06</td>
</tr>
<tr>
<td>Na$_2$O</td>
<td>2.0–4.0</td>
<td>2.31 ± 0.04</td>
<td>2.02 ± 0.04</td>
</tr>
<tr>
<td>CaO</td>
<td>2.0–5.0</td>
<td>1.83 ± 0.05</td>
<td>7.40 ± 0.09</td>
</tr>
<tr>
<td>MgO</td>
<td>2.0–5.0</td>
<td>0.78 ± 0.08</td>
<td>2.04 ± 0.04</td>
</tr>
<tr>
<td>K$_2$O</td>
<td>2.0–5.0</td>
<td>2.56 ± 0.03</td>
<td>2.43 ± 0.04</td>
</tr>
<tr>
<td>TiO$_2$</td>
<td>0.5–1.0</td>
<td>&lt;0.2</td>
<td>&lt;0.2</td>
</tr>
</tbody>
</table>

1.2. Knudsen cell reactor

The measurements of the uptake coefficients of H$_2$O$_2$ on the mineral dust particles were conducted in a Knudsen cell reactor, and the signals of reactants were monitored by a quadrupole mass spectrometer (HAL 3F 501, Hiden, UK). The details of our experimental apparatus have been described in our previous publication (Wang et al., 2011). The characteristics of the reactor used in this work are summarized in Table 2. The uptake of hydrogen peroxide on mineral dust was monitored by mass charge ratio $m/z = 34$ (H$_2$O$_2$) channel as in our previous work (Zhou et al., 2012), and the temperature of the reaction cell was heated or cooled by a refrigerated circulator.

### Table 2 – Knudsen reactor parameters.

<table>
<thead>
<tr>
<th>Knudsen reactor parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume (V) (cm$^3$)</td>
<td>461</td>
</tr>
<tr>
<td>Temperature (T) (K)</td>
<td>253–313</td>
</tr>
<tr>
<td>Surface-to-volume ratio</td>
<td>0.57</td>
</tr>
<tr>
<td>Total pressure (P) (Pa)</td>
<td>1.5 × 10$^{-3}$</td>
</tr>
<tr>
<td>Escape orifice diameter (mm)</td>
<td>3</td>
</tr>
<tr>
<td>Escape orifice escape rate (sec$^{-1}$)</td>
<td>0.297(T/M)$^{1/2}$</td>
</tr>
<tr>
<td>Effective area for the escape aperture (cm$^2$)</td>
<td>0.161</td>
</tr>
<tr>
<td>Sample surface area (A$_s$) (cm$^2$)</td>
<td>5.3</td>
</tr>
<tr>
<td>Sample collision frequency (ω) (sec$^{-1}$)</td>
<td>124.2</td>
</tr>
</tbody>
</table>

The effective area of the escape aperture was measured in each independent experiment according to the attenuation of the N$_2$ signal from one steady state to another (Li et al., 2002). M: Relative molecular mass of the gas molecular in the knudsen cell.
mass spectrometry (QMS) signal response of each mineral dust samples exposed to gaseous hydrogen peroxide at 298 K is depicted in Fig. 1. The initial concentration of gaseous hydrogen peroxide was first established in the chamber, and then one powder sample was exposed to contact with the reactant. The signal of hydrogen peroxide \((m/z = 34)\) dropped below its original value quickly initially, and then the signal intensity recovered gradually to a new steady state. An observed uptake coefficient \(\gamma_{\text{obs}}\), can be derived from the Knudsen cell Eq. (1) (Usher et al., 2002):

\[
\gamma_{\text{obs}} = \frac{A_h}{A_s} \left( \frac{I_0 - I}{I} \right)
\]

where, \(A_h\) is the effective area of the escape hole, \(A_s\) is the geometric area of the sample holder, and \(I_0\) and \(I\) are the QMS intensities detected when sample holder is covered and exposed, respectively. The values obtained from this equation were either the initial uptake coefficients \(\gamma_{\text{obs,init}}\) or the steady state uptake coefficients \(\gamma_{\text{obs,ss}}\) representing the uptake coefficient observed at the initial stages or the steady stages of the reactions.

During the experiments, the concentration of H\(_2\)O\(_2\) is related to the pressure in the reactor, which was kept approximately at \(3.0 \times 10^{-3}\) Pa equal to \(3.7 \times 10^{11}\) molecules/cm\(^3\). There were no remarkable variations of other monitored channels such as O\(_2\) \((m/z = 32)\) and CO\(_2\) \((m/z = 44)\), which meant that no catalytic oxidation reactions took place during the uptake experiments.

Eq. (1) is derived by assuming that gas–surface collisions only take place with the top layer of the dust. In order to determine the surface area of dust samples involved in the uptake processes, the uptake coefficients \(\gamma_{\text{obs}}\) were measured using the geometric surface area of the sample as a function of sample mass. As can be seen from Fig. 2, the observed uptake values are linearly dependent on the mass of these three types of dust. We determined the range of this mass dependence, and in this range the diffusion of the reactant gas can enter into the underlying layers of the dust, which can increase the collisions with the dust surface. In our study, the experiments were conducted in the linear mass regime. The entire surface area of the dust sample was accessible to H\(_2\)O\(_2\); where the entire sample participates in the reaction, the BET surface area is applicable (Underwood et al., 2000). From Eq. (2), a mass independent uptake coefficient can be derived

\[
\gamma_{\text{BET}} = \frac{A_h}{A_{\text{BET}}} \left( \frac{I_0 - I}{I} \right) = \gamma_{\text{obs}} \frac{A_s}{A_{\text{BET}}}
\]

where, \(A_{\text{BET}}\) is the surface area of the sample, taken as the BET area, which is equal to the specific BET area of the powder times the sample mass (Underwood et al., 2001). The results of all of our experiments are given in Table 3.

At 298 K, the effect of the gaseous hydrogen peroxide concentration on the uptake coefficient was also considerable in the range of \(2.6 \times 10^{11} - 1.2 \times 10^{12}\) molecules/cm\(^3\). When the initial concentration of gaseous hydrogen peroxide was changed in this range, there was no distinct dependence of initial uptake coefficients on this explored range. This concentration independence observed for gaseous H\(_2\)O\(_2\) initial uptake to mineral dust is very similar to the results reported by El Zein et al. (2014). They measured the heterogeneous interaction of H\(_2\)O\(_2\) with solid films of Arizona test dust using a low pressure flow tube reactor. After long time exposure, the uptake of hydrogen peroxide on the mineral dust surface showed relative saturation and became inactive toward further H\(_2\)O\(_2\) decomposition. There was clearly steady state uptake for all of the mineral dust samples. These phenomena suggest that the adsorption of H\(_2\)O\(_2\) on these three types of mineral dust was the

Fig. 1 – Uptake curves of hydrogen peroxide on Arizona test dust, Inner Mongolia desert dust, and Xinjiang sierozem at 298 K in Knudsen cell reactor. QMS: quadrupole mass spectrometry.
first step of the uptake process, and then there were continuing surface reactions. From the results of the experimental measurement, we inferred that the heterogeneous uptake on mineral dust surface took place through two steps, physical adsorption and heterogeneous reactions. This phenomenon also supported the results of field studies showing that heterogeneous uptake on ambient mineral dust aerosol seems to be an important sink for H$_2$O$_2$ (de Reus et al., 2005; He et al., 2010).

2.2. Effects of temperature on uptake kinetics of hydrogen peroxide on mineral dust

The temperature dependence of the uptake coefficients of heterogeneous processes on these mineral dusts were further investigated with an initial concentration of H$_2$O$_2$ of about $3.7 \times 10^{11}$ molecules/cm$^3$ over the temperature region of 253–313 K.

Table 3 summarizes all the results from the experiments. The initial uptake coefficients of Arizona test dust changed from $1.47 \times 10^{-4}$ to $2.71 \times 10^{-4}$, the initial uptake coefficients of Inner Mongolia desert dust changed from $2.19 \times 10^{-4}$ to $3.56 \times 10^{-4}$, and those of Xinjiang sierozem were in the range of $4.46 \times 10^{-5}$–$7.34 \times 10^{-5}$. From these results, it can be seen that the initial uptake coefficients of gas phase hydrogen peroxide uptake on these mineral dusts showed similar phenomena, in that they all decreased with increasing temperature. This phenomenon of the coefficient change trend with temperature conforms well to the physical adsorption process, which is the first main process in these systems. A similar temperature dependence of initial uptake coefficients was also observed in previous studies (El Zein et al., 2014; Romanias et al., 2013; Zhou et al., 2012). However, the steady state uptake coefficients of the mineral dust samples showed different phenomena. The $\gamma_{ss}$ of Arizona test dust increased slowly with increasing temperature from $5.57 \times 10^{-5}$ to $9.40 \times 10^{-5}$, the $\gamma_{ss}$ of Inner Mongolia desert dust increased markedly in the range of $2.50 \times 10^{-5}$–$1.31 \times 10^{-4}$, and the steady state uptake coefficients of Xinjiang sierozem had little correlation with temperature. El Zein et al. (2014) investigated the interaction of H$_2$O$_2$ with the Arizona test dust ($0$–$3 \, \mu$m) surface over the temperature range from 268 to 320 K at initial concentration of H$_2$O$_2$ ($[\text{H}_2\text{O}_2]_0$) $– 1.0 \times 10^{12}$ molecules/cm$^3$. They found that $\gamma_{ss}$ was independent of temperature. The observed temperature dependence indicates that the structure or the components may influence the steady state uptake processes.

Since the initial uptake coefficients showed negative temperature dependence, the changes of observed enthalpy ($\Delta H_{obs}$) and entropy ($\Delta S_{obs}$) for H$_2$O$_2$ adsorption on mineral dust and the reactive energy ($E_a$) for Inner Mongolia desert dust could be calculated from the following equations (Hudson et al., 2002; Jayne et al., 1991).

$$\ln \left( \frac{\gamma_{\text{BET}, \text{int}}}{1 - \gamma_{\text{BET}, \text{int}}} \right) = -\frac{\Delta H_{obs}}{RT} + \frac{\Delta S_{obs}}{R} \quad (3)$$

$$\ln \left( \frac{\gamma_{\text{BET}, \text{ss}}}{1 - \gamma_{\text{BET}, \text{ss}}} \right) = \frac{E_a}{RT} + A \quad (4)$$

Consequently, as shown in Fig. 3, the enthalpy ($\Delta H_{obs}$) and entropy ($\Delta S_{obs}$) were determined to be $-(7.7 \pm 1.5)$ kJ/mol and $-(98.6 \pm 19.7)$ J K/mol for Arizona test dust, $-(5.4 \pm 1.1)$ kJ/mol and $-(87.2 \pm 17.4)$ J K/mol for Inner Mongolia desert dust, and $-(7.8 \pm 1.6)$ kJ/mol and $-(108.3 \pm 21.7)$ J K/mol for Xinjiang sierozem. The activation energy ($E_a$) for Arizona test dust and Inner Mongolia desert dust is 8.64 and 18.4 kJ/mol respectively. In the atmosphere, when the activation energy of a given reaction is greater than $20$ kJ/mol, the role of the reaction in atmospheric chemistry is regarded as unimportant.
This suggests that the heterogeneous reaction of H$_2$O$_2$ on Inner Mongolia desert dust should not be neglected in the troposphere.

At low relative humidity conditions, the generally accepted mechanism for these uptake processes on oxide surfaces is as shown below (Lin and Gurol, 1998; Pradhan et al., 2010b; Zhao et al., 2011).

\[
\text{H}_2\text{O}_2(g) + S \rightarrow \text{S(OH)}(\text{S}^+ \text{+ OH}^-) + \text{OH}^-; \\
\text{OH}^- + \text{H}_2\text{O}_2 \rightarrow \text{H}_2\text{O} + \text{HO}_2; \\
\text{HO}_2^- + \text{HO}_2 \rightarrow \text{H}_2\text{O}_2 + \text{O}_2.
\]

The $\Delta H_{\text{obs}}$ value can demonstrate that H$_2$O$_2$ adsorbs on mineral dust samples in a similar capacity. The empirical formula between $\gamma_{\text{BET,\text{int}}}$ of H$_2$O$_2$ on mineral dusts with temperature can be calculated as:

\[
\gamma_{\text{BET,\text{int}}}(\text{Arizona test dust}) = \frac{\exp(931.0/T - 11.9)}{1 + \exp(931.0/T - 11.9)} \tag{5}
\]

\[
\gamma_{\text{BET,\text{int}}}(\text{Inner Mongolia desert dust}) = \frac{\exp(643.7/T - 10.5)}{1 + \exp(643.7/T - 10.5)} \tag{6}
\]

\[
\gamma_{\text{BET,\text{int}}}(\text{Xinjiang sierozem}) = \frac{\exp(941.3/T - 13.0)}{1 + \exp(941.3/T - 13.0)} \tag{7}
\]

Thus using this equation, the $\gamma_{\text{BET}}$ at other temperatures can be obtained. The initial uptake coefficients of hydrogen peroxide on these mineral dusts resembled those of other mineral aerosols.

### 3. Conclusions and atmospheric implications

To sum up, mineral dust plays an important role as particulate matter in the troposphere, and can act as a sink or reactive surface for a variety of atmospheric chemical species. In this study, we investigated heterogeneous uptake of H$_2$O$_2$ on three different mineral dust samples as a function of concentration of H$_2$O$_2$ over a temperature range from 253 to 313 K in a Knudsen cell reactor. Reactant concentrations were kept closer to ambient atmospheric levels compared to all previous studies of this reaction. The uptake coefficients showed little observable dependence on the concentration of hydrogen peroxide; while as a function of temperature, the initial uptake coefficients decreased with increasing temperature for these mineral dust samples. The temperature dependences of these processes can be expressed by Eqs. (5)–(7) for each mineral dust. For these three different kinds of mineral dust, with increasing temperature, the gap between initial uptake coefficient and steady state uptake coefficient became smaller and smaller. This meant that increasing temperature can promote the uptake processes from adsorption to reaction. According to the temperature dependence of uptake coefficients, the enthalpy ($\Delta H_{\text{obs}}$), entropy ($\Delta S_{\text{obs}}$) and activation energy ($E_A$) for each mineral dust were also determined.

The lifetime of H$_2$O$_2$ due to uptake onto Arizona test dust, Inner Mongolia desert dust and Xinjiang sierozem can be estimated by Eq. (8).

\[
\tau = \frac{4}{\gamma A} \tag{8}
\]

where, $A$ (cm$^2$/cm$^3$) is the dust surface area density, $\tau$ is the mean molecular speed, and $\gamma$ is the uptake coefficient. Because BET areas are applicable for calculating the uptake coefficients, here we use them as dust surface areas. If we assume a conservatively lowest dust loading of 5 to a highest dust loading of 600 $\mu$g/m$^3$ (Aymoz et al., 2004; Guo et al., 2013; Li et al., 2012), our measured uptake coefficients at room temperature are about (1.51 ± 0.30) × 10$^{-5}$, (2.41 ± 0.48) × 10$^{-4}$ and (5.30 ± 1.06) × 10$^{-5}$ for Arizona test dust, Inner Mongolia desert dust and Xinjiang sierozem, which leads to the related atmospheric lifetimes of gaseous H$_2$O$_2$ consumed by heterogeneous on Arizona test dust, Inner Mongolia desert dust and Xinjiang sierozem of 2.3 hr to 11.9 day, 1.5 hr to 7.5 day and 6.5 hr to 43.2 day respectively. The lifetimes of H$_2$O$_2$ via uptake onto mineral dust samples at different temperatures are listed in Table 4. Because the uptake coefficients were calculated based on the BET surface areas of the dust samples, they represent the lower limit. Since for Arizona test dust the BET surface area is about 4 times larger than the geometric area, this may cause $\gamma$ to be 4 times smaller than that calculated basing on the geometric surface area (Wagner et al., 2009). This gap should also be taken into account in box model studies, where it would cause an underestimation of H$_2$O$_2$ transformation. When sand storms occur, the surface mass aerosol concentrations may reach a peak at 3000 $\mu$g/m$^3$. Our results suggested that the heterogeneous uptake of H$_2$O$_2$ on mineral aerosol can significantly influence the concentration of H$_2$O$_2$ in the atmosphere.
Fig. 3 – Plot to determine the temperature dependence of hydrogen peroxide uptake on Arizona test dust, Inner Mongolia desert dust, and Xinjiang sierozem. $\gamma_{\text{BET,init}}$: initial uptake coefficient; $\gamma_{\text{BET,ss}}$: steady state uptake coefficient; $T$: temperature.

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**Table 4 – Atmospheric lifetime of hydrogen peroxide by uptake onto mineral dust samples.**

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>Arizona test dust</th>
<th>Inner Mongolia desert dust</th>
<th>Xinjiang sierozem</th>
</tr>
</thead>
<tbody>
<tr>
<td>253</td>
<td>1.3 hr–6.6 day</td>
<td>1.0 hr–5.1 day</td>
<td></td>
</tr>
<tr>
<td>268</td>
<td>1.4 hr–7.0 day</td>
<td>1.1 hr–5.8 day</td>
<td></td>
</tr>
<tr>
<td>283</td>
<td>2.0 hr–10.2 day</td>
<td>1.2 hr–6.6 day</td>
<td>4.7 hr–24.7 day</td>
</tr>
<tr>
<td>298</td>
<td>2.3 hr–11.9 day</td>
<td>1.5 hr–7.5 day</td>
<td>6.5 hr–34.2 day</td>
</tr>
<tr>
<td>313</td>
<td>2.4 hr–12.2 day</td>
<td>1.6 hr–8.2 day</td>
<td>7.8 hr–40.7 day</td>
</tr>
</tbody>
</table>

**References**


