

## Referee 2

<p>Overall: This paper presents laboratory measurements of the reactive uptake coefficient of N<sub>2</sub>O<sub>5</sub> on mineral dust particles, which is important for understanding the NO<sub>x</sub> removal rate and evaluating its impacts. Comparison of the N<sub>2</sub>O<sub>5</sub> uptake coefficient of two kinds of mineral dust particles shows that illite has much larger coefficient than Arizona Test Dust (ATD) particles and has stronger dependence on RH, which may be explained by the difference in chemical composition of illite and ATD. The paper is generally well written and the interpretation is plausible. I suggest publication on ACP after the following comments are addressed.</p>	<p>We thank referee 2 for the detailed review and positive comments on our manuscript.</p>
<p>In the abstract, it is stated that the uptake coefficient of ATD is independent on RH. This is not what shows in Figure 6. For ATD, it is clear that the uptake coefficients at RH = 0 and RH = 30% are 1.5 times (Table 1) larger than the coefficient at RH = 50% and RH = 70%. I believe a statistical test (T-test) will result in significant difference between them at 95% confidence level. The measurement at 20% is lower than those at RH = 0 and RH = 30% but has the largest uncertainty, so could that measurement be an outlier?</p> <p>A related question is that it is not clear how many repeated experiments were conducted at each RH and, how are the error bars calculated in Figure 6?</p> <p>In comparison, for illite, the uptake coefficients at RH = 0 and RH = 20% are 2.3 times greater than the coefficient at RH = 70%. Therefore, a more accurate way to present the difference is “the uptake coefficient of ATD has a weaker dependence than that of illite”.</p>	<p>We concur and indeed already state (p24866) that the uptake coefficient is at most weakly dependent on RH...” In the revised abstract we write:</p> <p>In contrast, the uptake coefficient derived for ATD is much lower (~0.006) and displays a weaker (if any) dependence on relative humidity (0-67%).</p> <p>Experiments were repeated at least three times at each RH for both dust, and the errors are the standard deviation (1 <math>\sigma</math>). In the revised version, the text now reads:</p> <p>“The heterogeneous reaction of N<sub>2</sub>O<sub>5</sub> with illite aerosol particles was investigated at five different relative humidities with initial N<sub>2</sub>O<sub>5</sub> concentrations in the range of (11-21)×10<sup>12</sup> molecule cm<sup>-3</sup>. Uptake experiments were repeated at least three times at each different RH for both illite and ATD.”</p> <p>and another sentence has been added to the header of Table 1:</p> <p>“Errors are statistical only (1<math>\sigma</math>), derived from a minimum of three individual results.”</p>

<p>2. The uptake coefficient of illite does not decrease until RH &gt; 20%, whereas the ratio of the mass of absorbed water to the mass of illite increased significantly for RH of 0-20%. This does not support the explanation that the adsorbed water “deactivates” the reactivity of illite. More interpretation is needed.</p>	<p>True. Unfortunately, we cannot focus in on small sections of the dataset as experimental scatter in the data is too large to make more than qualitative statements. We are wary of over-interpretation of the RH dependence. For example, <math>\gamma</math> (RH 0%) being <math>(91\pm39)\times10^{-3}</math> and <math>\gamma</math>(RH 17%) being <math>(93\pm8)\times10^{-3}</math> does not preclude a significant change in <math>\gamma</math>. Indeed, Figure 6 shows that a linear dependence from RH0% to RH69% would still be within error bounds.</p>
<p>3. An interesting point for Figure 6 is that the uptake coefficient of ATD starts to decrease when RH &gt; 30%, while the uptake coefficient of illite starts to decrease for RH &gt; 20% – there seems to be some threshold of the RH dependence, which is worth mentioning.</p>	<p>See comment above. The data is too scattered to accommodate such an interpretation.</p>
<p>4. A minor point is to label the two panels of Figure 6 as (a) and (b).</p>	<p>The Figures are labelled with the dust type. It’s not obvious why lettering with (a) and (b) should increase clarity.</p>
<p>5. On P24867, it says quartz does not have surface OH. In fact, quartz has surface OH, and that explains the dependence of uptake coefficient on RH for ATD. The low hygroscopicity of ATD can account for its weaker dependence on RH.</p>	<p>We agree. See comment above to referee 1. The text now reads:  The results for ATD reveal a rather different picture, with lower uptake coefficients (factor ~10 lower than illite at 0 % RH) and (at most) a weak dependence on RH. The lower uptake coefficients may be related to the mineral composition of ATD which mainly consist of feldspar and quartz (Broadley et al., 2012), which may have less (and/or less reactive) surface OH groups. The weak dependence on RH is probably related to the fact that the hygroscopic growth of ATD particles is very small (Gustafsson et al., 2005; Vlasenko et al., 2005) and therefore even at high RH, the amount of adsorbed water on the surface does not contribute significantly to N<sub>2</sub>O<sub>5</sub> solvation/ionisation but may still result in deactivation of surface OH groups as shown by Goodman et al. (2001). Surface sensitive methods would be required to confirm this postulate.</p>

<p>6. In Figure 3, it is better to show surface area concentration, which is a more related parameter than number concentration.</p>	<p>The surface area is indeed more related parameter than the number concentration. However, through the paper we derived the uptake coefficient from the dependence of <math>N_2O_5</math> concentration on aerosol number concentration, as shown in Figure 4. In order to be consistent we prefer to plot the aerosol number concentration in Figure 3.</p>
<p>7. P24864, Line 17, the formula of the slope shouldn't include Nd.</p>	<p><a href="#">Correction made.</a></p>
<p>8. <a href="#">P24866</a>, L6-7, "decreased by a factor of 2-3 as RH was increased from 0 to 67%." Again, there is no decrease in uptake coefficient between RH = 0 and RH = 20% for illite.</p>	<p>The decrease by a factor of 2-3 clearly refers to the overall effect when RH increased to 67% from 0%. See related comment above.</p>
<p>9. P24866, Line 10–14, "One possible explanation for the decrease in <math>\gamma(N_2O_5)</math> with increasing RH is the competitive adsorption between <math>H_2O</math> and <math>N_2O_5</math>, whereby the increased coverage of <math>H_2O</math> at high RH may result in blocking of particularly reactive surface sites, which are then unavailable for <math>N_2O_5</math> uptake, yet have insufficient water to support solvation/ionization of <math>N_2O_5</math> to <math>NO_2</math> and <math>NO_3</math>." I don't follow the reasoning here. How does increased coverage of <math>H_2O</math> at high RH result in insufficient water? Please clarify.</p>	<p>We agree, this was convoluted. What we try to say is that the two potentially competing processes (hydrolysis of <math>N_2O_5</math> or deactivation of surface OH) will have different dependencies on RH. We now write:</p> <p><a href="#">"One possible explanation for the decrease in <math>\gamma(N_2O_5)</math> with increasing RH is an increasing rate of deactivation of surface groups which are reactive towards <math>N_2O_5</math> (e.g. OH, see below) by physi-sorbed <math>H_2O</math>. For <math>\gamma</math> to decrease with increasing RH requires that the deactivation effect must outrun the rate of increase of surface hydrolysis of <math>N_2O_5</math> as RH increases. However, as hydrolysis (solvation/ionization of <math>N_2O_5</math> to <math>NO_2^+</math> and <math>NO_3^-</math>) will require more than one adjacent, surface-adsorbed <math>H_2O</math> molecule, the latter will likely only take place under conditions of multi-layer adsorption, with an unknown RH threshold."</a></p>
<p>1. Abstract: ATD is not defined.</p>	<p>done</p>
<p>2. P24861, L18, "(Wagner et al., 2008)" -&gt; Wagner et al. (2008)</p>	<p>done</p>
<p>3. P24864, L17, "According to Eqs. (2) and (3) The" -&gt; "<a href="#">According</a> to Eqs. (2) and (3), the"</p>	<p>done</p>