

## ***Interactive comment on “Reactive uptake coefficients for heterogeneous reaction of N<sub>2</sub>O<sub>5</sub> with submicron aerosols of NaCl and natural sea salt” by D. J. Stewart and R. A. Cox***

### **Anonymous Referee #2**

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General comments.

Stewart and Cox have presented a concise description of their measurements of the reaction of N<sub>2</sub>O<sub>5</sub> on NaCl aerosol particles. However, it seems that a few details in their methodology need to be addressed. These details may lead to a significant change in their results for the reaction rate coefficient  $k_r$ .

Specific comments.

The author's posit that  $g$  is a function of particle size. In principle, they should be able to show this by showing a dependence of measured  $g$  on particle size distribution. Unfortunately, this was not shown. In this respect it is worth pointing out that the

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weak size effect observed by Thornton et al. was not for the same type of aerosols. Nonetheless, it would help the reader if it is stated that it is assumed that  $g$  is a function of particle size, preferably at the beginning of the analysis section.

Mentioning this early on is important because if the uptake coefficient is  $g(r)$ , then it should be included in the kinetic expression E1. Then E1 would be integrated over the size distribution where  $k_l = \text{Sum\_}d k_l$  where  $d k_l = 0.25g(r)CdSA(r)$ . If the width of the distribution is not too large, ( $\ln \sigma \leq 0.3$ ) E1 can be used but then  $g$  is for a radius of  $r_p \exp(2.5(\ln \sigma)^2)$  where  $r_p$  is the peak of the number distribution (e.g., JGR, v100, 18775, 1995, Appendix.) However, it is not clear that this shortcut is valid here as 'the standard deviation', which really appears to be  $\log_{10} \sigma$ , is quoted at 0.27. The change in procedure detailed here may lead to a larger reacto-diffusive length and thus to significant changes in the extracted rate coefficients.

Yet their discussion of size dependent effects alludes to some sort of correction in this respect but it is hard to tell exactly what was done. If the authors do as is recommended above and start out with the assumption that  $g$  is a function of  $r$ , this may help the paper's clarity. The middle paragraph on p.10 is notably confusing as they are quoting a different value for  $\ell$  than that in Table 2. Furthermore, one should not use the volume approximation E3 when  $\ell$  is comparable to, or less than, the peak radii for the volume distribution. Finally, it is not clear what  $\langle r \rangle$  is: they quote values near 100 nm whereas the peak of the volume distribution is  $\sim 250$  -300 nm.

Finally, it is not clear that  $\text{HNO}_3$  concentrations can be neglected in the droplets. In addition to the nitrate effect, this will affect the pH of the droplets. As the authors indicate, a few hundred ppbv  $[\text{N}_2\text{O}_5]$  converted to  $\text{HNO}_3$  and fully taken up and distributed throughout the volume of the droplets will result in  $\sim 1$ -2 M  $[\text{NO}_3^-]$ . One may need to consider that  $[\text{nitrate}]$  is larger near the surface of the droplets as  $\text{N}_2\text{O}_5$  (and  $\text{HNO}_3$ ) uptake is ongoing: the time constant for equilibration of  $[\text{NO}_3^-]$  within the droplets is on the order  $10^{-4}$  s (compare to the hydrolysis rate of  $\sim 1$ -to- $10 \times 10^4$  s $^{-1}$  for  $\text{N}_2\text{O}_5$ .)

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