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## ***Interactive comment on “Viscosity controls humidity dependence of N<sub>2</sub>O<sub>5</sub> uptake to citric acid aerosol” by G. Gržinić et al.***

**Anonymous Referee #1**

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This paper describes laboratory measurements of the N<sub>2</sub>O<sub>5</sub> reactive uptake coefficient ( $\gamma_{\text{N}_2\text{O}_5}$ ) on citric acid aerosol as a function of relative humidity (RH). The fate of N<sub>2</sub>O<sub>5</sub> is important to understand for constraining the reactive nitrogen budget, and reaction on aerosols is a major component of N<sub>2</sub>O<sub>5</sub> loss (at least in models). Thus, the topic is well aligned for ACP, and the experiments are carefully described and the paper overall is well written. I recommend publication after a few issues are addressed.

–General comments

The measured  $\gamma_{\text{N}_2\text{O}_5}$  is quite low, ranging from 3E-4 to 3E-3 at its maximum. The RH dependence, aside from a couple points, is more of a continuous function than a step-function that would be expected from efflorescence/deliquescence behav-

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ior. The authors use this behavior and some analogous behavior in the viscosity measured independently to conclude that it is viscosity (via diffusion limitations) that controls the observed humidity dependence and even the overall low absolute value for  $\gamma_{\text{N}_2\text{O}_5}$ .

I certainly find the argument reasonable, though the spread in the estimates of diffusivity and that the full behavior of the measured  $\gamma_{\text{N}_2\text{O}_5}$  is not captured by any version does leave a little to be desired. This is the case not only for the citric acid aerosol but also for the application of the model to the previously published malonic acid data from Griffiths et al and Thornton et al. There is a strong fall off for the  $\gamma_{\text{N}_2\text{O}_5}$  on malonic acid aerosol with decreasing RH that the model doesn't capture. Why?

My recollection is that the uptake of  $\text{N}_2\text{O}_5$  to malonic acid at low RH was explained by Bertram and Thornton 2009 as a  $[\text{H}_2\text{O}]$  effect. Is that not treated in model? Or is it that the diffusivity parameterization for  $\text{N}_2\text{O}_5$  in malonic acid needs to be steeper at low RH (or some combination) as it is with the Reid et al and Lienhard versions for citric acid? It seems like the authors are arguing that changes in  $[\text{H}_2\text{O}]$  may affect diffusivity, not just reactivity, and so what is the  $[\text{H}_2\text{O}]$  effect in Bertram and Thornton 2009 explaining (one or both)?

I feel like the authors are on the cusp of being able to reconcile several disparate results and thus demonstrate a comprehensive understanding, but then stop short. For example, I think high viscosity (low diffusivity) can explain why citric acid mixed with ammonium sulfate was an outlier in the Gaston et al work, but again, this connection isn't made very strongly in this paper and is it necessary to also invoke a phase separation between ABS and citric acid as used in that paper to explain the other systems?

In addition, could the authors amend the uptake model to include a surface reaction term so that the different diffusivity parameterizations could be better evaluated against the data? It is certainly reasonable that there is a reaction with surface water as noted

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given the non-zero  $\gamma_{\text{N}_2\text{O}_5}$  measured on solid organics and reported by others previously. Then use the fact that model "works" for citric acid and invert it to put forth a viscosity vs RH curve for malonic acid that would be required to explain the  $\gamma_{\text{N}_2\text{O}_5}$  at low RH, if not just a  $[\text{H}_2\text{O}]$  effect. If it is the latter, why is it not in the model?

–Detailed Comments

Line 6 21993, it seems wall loss was evaluated before and after aerosols were delivered to the flow tube, but please specify.

Line 4 21995, I think the appropriate reference is Griffiths et al, not Thornton et al (at least for the aerosol systems mentioned).

Line 5 21995,  $\gamma_{\text{N}_2\text{O}_5}$  for citric acid, even at its highest ( $3\text{e-}3$ ), is almost an order of magnitude lower than an aqueous sea salt particle.

Figure 1 legend, "N<sub>2</sub>O<sub>5</sub> particle phase" - that isn't really the case right? I assume it is "N<sub>2</sub>O<sub>5</sub> reacted" or "particulate nitrate"

Figure 3, are the lower gammas at 35 and 53% RH evidence of possible efflorescence? Putting a drier inline at 17% RH before the flow tube seems to me to run the risk of causing efflorescence at times.

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