Reviewer 1

Comment
This manuscript describes a new approach to the measurement of the relative uptake coefficients of NO3 and N2O5 to aerosol particles. In this approach the losses of NO3 and N2O5 to the same aerosol sample are measured using cavity ringdown spectroscopy obviating the need to characterize the particle sizes, morphologies or surface area. Measurements of the ratio of uptake coefficients on Saharan dust are presented, and from these data the uptake coefficient of NO3 is inferred to be an order of magnitude smaller than the only other value reported. These experiments serve to illustrate the approach and how it can be used to constrain uncertainty on uptake coefficients. A large measured ratio of NO_3 and N_2O_5 uptake coefficients from experiments on ambient urban aerosols implies that these particles contained a substantial amount of organic species or a coating of organic species. Additional experiments with soot aerosol demonstrated a range of ratios, presumably from the variation in the soot source conditions.
Reply
None required

Comment
Overall, this manuscript is very well written and focused on describing this new approach to measuring ratios of uptake coefficients on complex aerosol. Since it eliminates the need to determine the aerosol surface area, this approach should prove useful for the study of a wide range of aerosols, and other researchers may benefit by using this approach. The content is appropriate to Atmospheric Chemistry and Physics, the length is commensurate with the content, and the data are interpreted well and in such a way as to support the conclusions. This manuscript should be accepted for publication as is, but the following comments are provided for the authors' consideration.
Reply
None required

Comment
Does the measured ratio of uptake coefficients change if one reactant is in large excess to the other? For example, inhibited uptake of one species may be observed if the other species occupies a substantial number of surface sites. Were any experiments conducted with a large [N2O5]/[NO3] ratio (or vice versa)?
Reply
The ratio of NO_3 to N_2O_5 was varied over a factor of three without inducing a measurable change in the relative uptake coefficient. The use of low concentrations of NO_3 and N_2O_5 should help prevent significant surface saturation of the sample, at least at short exposure times. The following text has been added: "We also found no significant change in $\gamma(NO_3)$ / $\gamma(N_2O_5)$ upon varying the absolute mixing ratios of NO_3 and N_2O_5 within the range indicated in Table 1. The ratio of NO_3 to N_2O_5 concentration was also varied by a factor of ≈ three, with no measurable change in the relative uptake coefficient. This is related to the use of relatively low concentrations of both reactants, which (at least at low exposure times) will not modify the surface reactivity significantly."

Comment

It is mentioned that the measurements of Saathoff et al., 2001 and Karagulian and Rossi, 2007 support a large uptake coefficient for NO_3 than for N_2O_5 on soot (section 3.3, page 11). It would be helpful if these results are quoted so as to allow comparison to the present work.

Reply

We feel there is little to be gained by being quantitative regarding the soot experiments. As described in the text, the soot types used by us and by Saathoff / Karagulian are vastly different and a detailed comparison is not warranted. The point of the soot (and ambient aerosol measurements) was not to derive accurate data, but to illustrate the range (and limitations) of this method.