

We thank Tony Cox for this positive appraisal of our manuscript. Each comment made (black) and our response (blue) is listed below. Red text indicates where changes to the manuscript have been made.

The paper describes a study involving field measurements of nitrogen oxides and atmospheric particulate matter, and analysis aimed at estimating the uptake coefficient ( $\gamma$ ) and yield of nitryl chloride ( $f$ ) in the heterogeneous processing of dinitrogen pentoxide in the reaction:  $\text{N}_2\text{O}_5 + \text{Cl}^- = \text{ClNO}_2 + \text{NO}_3^-$ . The measurement site is at 800m altitude in a rural location in Western Germany, which is influenced by pollution from the adjacent Rhein-Main conurbation, and by long range transport of air of marine origin containing sea salt aerosol. This is the latest of several papers appearing in the literature reporting estimates of  $\text{N}_2\text{O}_5$  uptake coefficients and reaction paths on ambient atmospheric aerosol of more or less defined composition. It is known from laboratory studies that the rates of these heterogeneous reactions are highly dependent on atmospheric conditions as well as the chemical nature of the aerosol. Measurements under 'real world' conditions are necessary to provide confidence that the correct parameters are used. This is a timely study and the measurements have been conducted with a well conceived strategy for advancement of knowledge of this potentially important atmospheric process. Two different methods were used to obtain estimates of  $\gamma$  and  $f$  from the observational data. In the first methodology (discussed in section 4.1) involved determination of the formation rates of  $\text{ClNO}_2$  and  $\text{NO}_3^-$  products for known  $[\text{N}_2\text{O}_5]$  and aerosol surface area. A total of 12 values of  $\gamma$  were obtained during the field campaign. The second methodology used for estimating  $\gamma$  is the so called steady state method, (discussed in section 4.2), which assumes that during nighttime the loss of  $\text{NO}_3$  and  $\text{N}_2\text{O}_5$  is in balance with production from  $\text{NO}_2 + \text{O}_3$ . The  $\gamma$  values can be evaluated from the expressions for inverse steady state lifetimes of  $\text{NO}_3$  and  $\text{N}_2\text{O}_5$ . Periods were selected so that air mass characteristics were consistent with assumptions made for a tractable analysis to obtain the target parameters. Only 3 values of  $\gamma$  were obtained using the steady state method. As in previous studies in which uptake coefficient have been derived from ambient data sets there is large variability in the results over the time period of the observations: ( $0.004 < \gamma < 0.11$ ;  $\gamma(\text{av}) = 0.028 \pm 0.027$ ). Overall the results were consistent with those derived from earlier field studies, as discussed in section 4.4. The factors affecting the  $\gamma$  values derived in this study are discussed in section 4.6 in terms of parameterisations of IUPAC panel (RH), Bertram and Thornton (2009; particulate  $\text{NO}_3^-$ ,  $\text{Cl}^-$  and  $\text{H}_2\text{O}$ ), and Antilla (2006; organic content) derived from the body of information based on laboratory studies. The predictions and the measurements are again in reasonable agreement. However the apparent influence of particulate  $[\text{NO}_3^-]$  is less than predicted by the models and the predicted suppression of uptake by organics is much less if it is assumed that the organic content exists as a hydrophobic coating. There was also a high variability in the values of  $f$  (range  $0.035 \pm 0.027$  to  $1.38 \pm 0.60$ ). The large spread of values is expected from the variability in particulate  $\text{Cl}^-$  as a result of differing air mass origins (discussed in section 4.5). The observation of  $\text{ClNO}_2$  is an unambiguous indication that the particles at the measurement site contain  $\text{Cl}^-$ , which will influence the overall uptake rate in the field situation. In summary the study provides further evidence for a role of active heterogeneous chemical processing of  $\text{N}_2\text{O}_5$  in the lower atmosphere at nighttime, which is strongly influenced by the local physical conditions and chemical composition of the particles. The rates are as a result very variable but current mechanisms based on laboratory studies give a reasonable rationale of the parameters observed

Other comments. The presentation of the work is of a high standard of accuracy and completeness. For such complex data set and depth of analysis it is reasonably easy to read

and the diagrams are informative and not excessively detailed. The conclusions are justified in the context of the body of knowledge on this topic

We thank Tony Cox for this very positive overall assessment of the paper.

Minor queries

P 3, l. 20 insert 'a' after 'in'

Correction made

p 5, l. 1 please use metric units of length paragraph starting l.28

We have added mm lengths to the inch lengths listed.

Fig 2. Please reorder the 3 plots to be consistent with the order they are discussed in the text

We prefer to keep the present order.

p 8, l. 1 Please indicate here that the covariance is illustrated in Fig 5

Fig 5 shows the correlation. The co-variance is apparent from the time series.

p 17, l. 14 please indicate the direction of the dependence of gamma suppression on RH; does it increase or decrease?

We now write “As the particles during PARADE have significant organic content (see Fig. 2) this may reflect the fact that the organic suppression of  $\gamma$  is reduced at high relative humidity as reported by (Gaston et al., 2014).”

Table 1, last row, col. 1 Were the observations made at 650 m or at 825 m, the altitude referred to on p3, l.27?

The observations were at 825 m. This was a typographical error and has been amended.