

## ***Interactive comment on “Lagrangian analysis of low level anthropogenic plume processing across the North Atlantic” by E. Real et al.***

### **Anonymous Referee #2**

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This paper deals with the processing and transport of photo-pollutants between continents through the use of case study. The ICARTT experiment provided an ideal opportunity for such a study. The Lagrangian modelling tool used in this study is highly suitable for this kind of study. The paper is structurally well constructed, although there are minor typographic issues. The major conclusions, that wet deposition of HNO<sub>3</sub> has a significant impact on the O<sub>3</sub> budget and that photochemical loss of CO plays an important role in determining the CO to O<sub>3</sub> ratio, are significant and run somewhat counter to the perceived wisdom. I have some detailed comments below and the paper requires some modification but with these changes I believe the paper is suitable for publication.

Major issues

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## N<sub>2</sub>O<sub>5</sub> hydrolysis

On page 7521 the authors discuss the lack of inclusion of N<sub>2</sub>O<sub>5</sub> hydrolysis on aerosols in their model. They justify their lack of inclusion through a &#8216;two wrongs&#8217; making a right argument - as the lack of inclusion in some of the simulations leads to an underestimate in NO<sub>x</sub> they should not include it. I don't find this argument convincing. Firstly, given the weight placed on HNO<sub>3</sub>, NO<sub>y</sub> and NO<sub>x</sub> in the paper, missing the sink of roughly 50% (annual global mean) of the NO<sub>x</sub> seems untenable. Secondly in some of the simulations (chemistry only) the NO and NO<sub>2</sub> concentrations are overestimated compared to the observations. The inclusion of N<sub>2</sub>O<sub>5</sub> hydrolysis would presumably improve these simulations. Thirdly, the rate limiting step for the N<sub>2</sub>O<sub>5</sub> hydrolysis system is the production of NO<sub>3</sub> at night. Thus a detailed knowledge of the aerosol surface area and type are not really needed to incorporate this processes; the presence of the reaction at a reasonable rate is all that is required. I would suggest that an N<sub>2</sub>O<sub>5</sub> hydrolysis reaction is included in the model and the impacts assessed.

## HNO<sub>3</sub> and O<sub>x</sub>

The importance of HNO<sub>3</sub> as a sink and source of NO<sub>x</sub> and of O<sub>3</sub> is a central theme of the paper. How HNO<sub>3</sub> has this impact is not discussed in great detail. I can imagine two possibilities. One is that it impacts the NO<sub>x</sub> concentration and thus the rates of production and loss of O<sub>3</sub>. The other is that it impacts the O<sub>x</sub> budget (as HNO<sub>3</sub> is one component of O<sub>x</sub>) and in that way directly impacts the O<sub>3</sub> budget. Given the HNO<sub>3</sub> concentrations are so high losses of HNO<sub>3</sub> may impact O<sub>x</sub> directly. The authors should investigate this point.

The use of NO It is understandable that the authors have used NO as this is a primary observation. However, its concentration changes rapidly with time of day, temperature and O<sub>3</sub> concentrations. Using the NO/CO ratio as in page 7517 is fraught with problems due to the changing values of JNO<sub>3</sub>, O<sub>3</sub> and T. Using measured (NO+NO<sub>2</sub>) or deriving an NO<sub>2</sub> concentration from steady state would remove these problems. The

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authors should show that they have investigated this point or remove the NO/CO ratio from the paper.

Wet-deposition Although I agree that the dominant impact of wet deposition is to remove HNO<sub>3</sub> the authors have not shown that the impact is entirely due to this. The wet deposition of other species (H<sub>2</sub>O<sub>2</sub>, MHP, CH<sub>2</sub>O etc) may also have an impact on the photochemistry. Runs where only HNO<sub>3</sub> is wet deposited would quantify the significance of HNO<sub>3</sub> wet deposition robustly.

Quite a lot of details are given of wet deposition schemes which are in the end not used in the modelling. This section should be shortened and tightened up.

Minor comments Abstract &#8216;Chemical and wet deposition processing dominated&#8230;&#8217; Dominated over what?

Page 7528 The background concentrations are given in Table 3 for some of the species. What about other species? Hydrocarbons, oxygenates? Are they mixed to a zero background? How were these background conditions derived?

Page 7529 How do the mixing rates here compare to those derived by Arnold et al.?

Page 7530 The comparison with the measured VOCs is problematic as they appear to increase in many cases between interceptions which is beyond the model to simulate. There should be some comment about this. Presumably this is something to do with observational techniques, not sampling the sample plume, emissions etc.

The authors should specify that there is an &#8216;indirect&#8217; effect of wet-deposition on O<sub>3</sub>. It appears at times in the text that they are suggesting that the wet-deposition is directly responsible for O<sub>3</sub> loss.

## Conclusion

The discussion of the problems with July 22nd and July 26th Lagrangians seems overtly concrete given the evidence presented in the text which is more flimsy that

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the conclusions suggest. Also the discussion of ship emissions and London in the conclusions is not included in the text and thus seems inappropriate for the conclusions. Both these items should be strengthened in the text or removed from the conclusions.

Tables and Figures ————— The figure and table captions could be improved. The caption for Table 2 should be expanded to explain what alpha spec actually is. Presumably this should be alpha HNO<sub>3</sub>. Table 4 shows measured NO<sub>y</sub>/CO and NO/CO ratio as well as O<sub>3</sub>/CO indicated in the caption.

Some of the figures are rather small and hard to read (Fig 2, Fig 4) although this may be due to the ACPD page format.

There are many typographical errors, missing words, spelling mistakes etc in the text. For example: 'O<sub>3</sub> levels'; should be replaced by O<sub>3</sub> concentrations as level may be interpreted to mean altitude. Phrasing such as 'Concerning the S-CLOUD'; does not read well in English. It would be advantageous if a more careful check was made of the language by a native speaker.

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 7509, 2008.

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