**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Quantification of chemical and physical processes influencing ozone during long-range transport using a trajectory ensemble" *by* M. Cain et al.

Anonymous Referee #2

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General comments:

The Lagrangian modelling framework is a very useful tool for modelling structured layers in the troposphere and for the interpretation of aircraft observations of atmospheric composition. As pointed out by the authors, it also has many advantages over the Eulerian approach to composition modelling such as effective resolution and computational cost. However, it has suffered from two major drawbacks: poor or no representation of mixing and convective transport. In the submitted manuscript by Cain et al., they have extended an existing photochemical trajectory model to explicitly include a robust representation of mixing so that photochemistry and mixing during long-range transport

of air masses can be modelled simultaneously. In addition to mixing, they have partitioned accumulated changes in ozone by process, thereby separating the chemical and physical processes during long-range transport. They have then applied this model to a number of case studies from the ITCT-Lagrangian 2004 aircraft campaign and performed a number of sensitivity experiments. As a result of the model extension and its success in simulating the observed magnitude and variability of the aircraft observations, this work represents a major contribution to Lagrangian modelling, improves our understanding of the chemical and physical processes which affect atmospheric composition during long-range transport and should be accepted for publication in ACP when the specific comments below are addressed.

Specific comments:

1. You often refer to photochemical production of ozone. Strictly speaking, this should be net photochemical production.

2. Can you comment on the suitability of shadow trajectories that do not move in the vertical? To what extent do you think this underestimates the variability in the vertical in your background profile? This occurs to some extent in Case 1 but how important do you think it might be in the other cases?

3. In section 4.1, you mention that there is chemical loss of ozone during the night. Is this really correct?

4. In Case 1, the model overestimates OH and CO, with the implication that mixing is too slow in the model and/or that the ensemble doesn't include the cleaner air above the air mass of interest. If this is the case, can you then comment on why there is great similarity between the observed and modelled rate of decay of ethane, acetylene, and propane?

5. Case 5 represents low-level transport behind a cold front, and unsurprisingly, wet deposition of NOy is an important process in this case. You mention that modelled

losses by wet deposition are overestimated. I'd like to see some discussion on the impact of only using surface precipitation and an additional sensitivity test in which three-dimensional precipitation from ECMWF analyses is used in the model (if available).

6. In Section 5, Ridley and Arnold (2009) have explored the uncertainty in chemical reaction rate co-efficients for these cases ... Add co-efficients to this sentence.

7. In Section 5, change "where the air masses is well" to "where the air masses are well".

8. Given the number of cases considered here and the number of sensitivity tests, I found some of the sections a little bit clumsy, jumping from one case to another. However, the summary of all cases in Section 7.2 was coherent with some very clear messages. It would improve the manuscript if the authors could somehow make Sections 5 and 7.1 more coherent.

9. Why was the basic chemistry used in the sensitivity tests for cases 1, 2 and 5? If you think that the full chemistry scheme would give greater overlap with the observations, why not use it?