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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.

M. Cain et al.

mc692@cam.ac.uk

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The authors would like to thank all the reviewers and the editor for their comments and suggestions for this manuscript. Responses to specific points from reviewer #2 are below. All other points made by reviewer #2 have been amended in the final manuscript exactly as suggested.

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?

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–Shadow trajectories follow the same horizontal path of the reference trajectory and evolve photochemically, but do not move vertically. Their purpose is only to provide suitable far-field mixing ratios to the background profile used in the vertical diffusive-mixing parameterisation. The simplification of these trajectories will introduce an uncertainty in the background composition.

A more comprehensive approach would require calculating 3D trajectories of all air masses that come into contact with the air-mass of interest. However, owing to the vertical wind shear these "neighbouring trajectories" could come from distant origins and it would be necessary to model the evolution in composition (mixing and photochemistry) along each one, which would in turn require knowledge of their neighbouring air masses. This is the major weakness of a Lagrangian approach. As trajectory length increases, the number of trajectories required to fully represent the evolution of an air mass, its neighbours and mixing between them increases rapidly. The problem is made tractable here by making the assumption that vertical mixing across the ensemble dominates the behaviour and the correspondence with observations demonstrates that useful results can be obtained taking this approach.

Sensitivity tests were presented varying the mixing rate and the impact on ozone was not larger than the other tests. Therefore it is likely that the assumptions regarding background mixing ratios will have an impact. Overall, we believe that the scheme represents an improvement compared to a simple relaxation to a static background. Another approach would be to use the results from a low resolution Eulerian chemical transport model to provide the background for the mixing scheme (outside the range of the ensemble) as suggested by referee #1 (as used for example in Methven et al, 2003).

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

–There is a small net loss in O3 the reference trajectory, and those trajectories sur-

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rounding it, during the first night of the simulation. The text has been amended to make this clear.

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?

–The rate of decay for CO and the hydrocarbons in the model is consistent with the fall between the second, third and fourth aircraft interceptions. However, CO and C3H8 are overestimated by the model at the centroid of the plume (the red trajectories). This could be explained by a more rapid dilution of the plume between the first and second Lagrangian matches in the atmosphere, perhaps associated with cleaner air above than in the model background, as the background CO mixing ratio above the low level plume of interest is much lower than the plume, reaching a minimum of 70ppbv aloft.

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).

–The following text has been inserted in line 20 of page 3043:

The precipitation field from the ECMWF model used to create the analysis is the rate of water reaching the ground from the whole column. Although a 3-D field for condensed-phase water mixing ratio (liquid plus ice) is calculated as part of the prognostic cloud scheme in the ECMWF model, the conversion to precipitation (rain and snow) and fall out is treated as an instanteous process in each time-step. The scheme does calculate the fraction of precipitation reaching the ground from each level and a profile

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of evaporation, but this information is not output and therefore not available to use offline. In addition, the scavenging coefficient in the parameterisation of wet deposition used in CiTTyCAT (Penner et al, 1991) is based on the relationship between surface precipitation and regional nitrate budgets.

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.

–Sections 5 and 7.1 were written with the aim of describing the processes and how they interact with each other, therefore these sections do go from case to case. A few changes have been made to the text to try and clarify this and to make it more readable.

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?

–The simulations were originally conducted using the basic chemistry during the lead author's PhD. When the chemistry was updated the control simulations were re-run. For cases 1, 2 and 5 the evolution is similar to the basic scheme control and the main effect is a scaling of ozone change as with all the other sensitivity experiments. Therefore, it seems appropriate to assume that the sensitivity experiments would yield similar results about either control run. However, for case 3 the two chemistry schemes gave quite different evolutions (not a simple scaling) and the entire ensemble was re-run. It would also be difficult to re-run all ensembles because the lead author has changed institution and is working full-time on a different job.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 3019, 2012.