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Interactive Comment

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

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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 #3 are below.

1)'Lagrangian air mass or control volume' needs to be defined at the initial time. Some description of its time-evolution might be useful, especially in terms of its volume.

–In a Lagrangian model the "air masses" do not have a precisely defined volume. The trajectories represent the path of an infinitesimal "particle" moving with the wind. Any set of particles surrounding a volume at initial time describes a surface that is moved

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and deformed by the wind. The mass contained within the envelope would be constant, even if the advecting winds are those resolved in analyses (because the winds obey the continuity equation), as discussed in Methven et al (2003) for example. In this sense, it is appropriate to call it an air mass even though material can be exchanged between trajectories or across the envelope by diffusive mixing (or unresolved turbulent motions). In the experiments shown here the initial trajectory points lie along flight tracks where there are measurements. Although these are lines, rather than envelopes about an initial volume, it is still relevant to describe the ensemble (or portions of it with similar composition) as an "air mass".

2)More details (or a schematic diagram) of mixing processes between the Lagrangian air mass and adjacent background (shadow trajectory) make the manuscript readable. In addition, the terminology 'mixing' should be clarified in the manuscript. Does it mean 'vertical turbulent mixing'? please clarify and use it consistently through the manuscript.

–Yes, the mixing scheme represents turbulent diffusive mixing in the vertical. Full details of the mixing scheme in the model are contained in section 4.2 of Pugh et al, GMD, 2012, which also contains a schematic diagram, shadow trajectories and background profiles in figures 7, 8 and 9. Rather than including these diagrams again, the manuscript has been amended to refer more specifically to Pugh et al (2012), and references to mixing have been defined more carefully. The definition of mixing has been clarified, with the phrase "This term is referred to in the manuscript as 'mixing'" being added after equation 8.

3)According to "mixing" process explained in section 3, atmospheric boundary layer height might be an important parameter in the model.

–Yes, the BL height (taken from the ECMWF model analysis) will be important for the background profile, as this is the depth over which the background profile is well mixed.

4)The Lagrangian trajectories of chemical plumes studied were calculated using the ECMWF which has coarse spatial (1.125 deg) and temporal (6-hr) resolutions. Is it enough to track accurate position of plume ensemble members? The plumes captured by aircraft are much narrower and shorter in time as compared.

–There are several papers (including Methven and Hoskins, JAS, 1999) where it has been shown that tracer features can be represented accurately on scales 6-10 times narrower than the smallest resolved scale in the advecting wind field. The main error is usually a sideways displacement of tracer filaments. So we expect to represent the transport of features of the order 10-20km in chemical tracers which is of the order of 100s along a flight track. There is variability observed on shorter space and time scales and data is used every 10s to obtain representative spread in initial composition.

The Lagrangian matches were identified by Methven et al (2006), using two trajectory models alongside hydrocarbon fingerprinting and the match was assessed using the independent tracer equivalent potential temperature.

5)Clear explanations of 'reference trajectory' and 'shadow trajectory' might be necessary. What does 'the centroid of the trajectory ensemble (P3031)' mean?

–The centroid is in general terms defined as the "centre of mass" of a feature. In this manuscript, the "reference trajectory" is defined as the trajectory that passes most closely through the downstream matching flight segments identified by Methven et al (2006). We refer loosely to this (and similar trajectories) as the centroid of the ensemble. The manuscript has been changed to make this more clear.

6)Based on the previous observational study, the atmospheric thermal structure over the North Atlantic can be characterized by strong stable condition. In the strong stable marine boundary layer condition, turbulent mixing might be significantly suppressed, happen only sporadically. In addition, the atmospheric boundary layer heights are quite low (100 m), or sometimes it does not well defined. So, it is expected that a turbulent mixing has a minor impact. This condition can be included in the interpretation of the author's model simulations.

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–Under such conditions, the value used for the diffusion coefficient (kappa) should reflect the stability of the atmosphere, and would generate a small amount of turbulent mixing. Often, if the plume of interest is well above the boundary layer, it can be completely disconnected from the surface. If such a scenario were modelled in this framework, the influence of mixing on the plume of interest would be small.

7)Several studies on dry deposition velocity of O3 shows it ranges from 0.006-0.15 cm/s, where high end can be measured over unstable marine conditions such as Gulf of Mexico in USA. In the reviewer's viewpoint, the value of 0.2 cm/s used in the manuscript for case 3 is a quite large value if we consider commonly very stable marine condition over the North Atlantic.

–The experiment with a dry deposition velocity of 0.2cm/s was intended to be a sensitivity experiment to text the model's response to high deposition velocities, and we agree that this is too high compared to what would be expected. It is noted in the manuscript that observed values were a maximum of 0.1cm/s during the field campaign with an average of 0.044cm/s (0.03cm/s was used here in the control).

8)Spatial distribution of the ensemble trajectories is not shown in Figs. 1-4. Wide spread of the trajectories is expected after long-range transport because of strong wind shear in the vertical direction over the North Atlantic.

–This is true, the trajectory ensembles do spread out in the horizontal as well as the vertical. Since they have already been shown in Methven et al (2006) they have not been repeated here to keep the paper shorter. The ensembles used here do differ slightly from Methven et al, because the initialisation windows are longer to include aircraft vertical profiles.

9)Model-observation discrepancy in Figs. 1-4 might be also attributed to both the poor initialization and the poor representation of the plume by the aircraft. Inclusion of analysis and explanation regarding this might be necessary.

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–This is a risk of this method, and has been highlighted in the manuscript in section 4.2, although the same error could potentially affect all simulations.

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