

***Interactive comment on “A Lagrangian analysis of the impact of transport and transformation on the ozone stratification observed in the free troposphere during the ESCOMPTE campaign” by A. Colette et al.***

**Anonymous Referee #2**

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The paper describes a multiple-tool analysis of enhanced ozone concentrations observed in the free troposphere (FT) during the ESCOMPTE experiment. High-resolution back-trajectories are first computed using mesoscale simulations of the MesoNH model to identify the origin of the air masses and it is found that the enhanced ozone concentrations originated from the Iberian boundary layer. In a second step, the chemical transformation (e.g. ozone production/loss) during the transport from Spain to the south of France is quantified using a chemical box model travelling along Lagrangian trajectories with the initial concentrations being provided by a

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mesoscale chemical transport model. The paper addresses important topics (e.g., chemical transformation in air masses travelling over long distances) with an original suite of tools. I believe the paper should be published in ACP given that the following comments are addressed.

1) Section 3.1.2. The Authors characterized the origin of air masses (e.g. from the boundary layer) using simulated value for TKE. The criteria they use is however not entirely clear to me and appears to be somewhat arbitrary. Could the Authors justify a little bit more their criteria?

2) Section 3.3.2 and 3.3.3. I find these sections a little bit hard to follow. For example the Authors give the results for the enhanced  $\text{NO}_x$  scenario before saying why they think the model may underestimate observed  $\text{NO}_x$  levels. In fact, reading section 3.3.2, one already wonders about the validity of initial conditions and the possible validation of the CHIMERE results with EMEP observations. This discussion (as well as that on the MONA observations) should come earlier in the manuscript. I would recommend rephrasing these sections to avoid repetition and to increase the “readability” of the paper.

3) Section 3.3.2. It is mentioned that ozone is “degraded” in 31% of the trajectories. This is a rather substantial portion of the cases; however these are not discussed at all. How do those trajectories differ from the remaining portion in which net ozone production occurs? Are the export processes (e.g. scavenging efficiency for  $\text{NO}_x$ ) or initial conditions different?

4) Section 3.4.1. It is not so clear to me why the Authors used climatological values for the background concentrations rather than the actual 3-D fields provided by CHIMERE.

5) I do not think this is correct to say that the simulations identified by the Authors are “optimal”. There are several parameters that could be important for ozone production in the travelling air masses (and thus concentrations at the end of the trajectories) that are not discussed at all. For example, water content and photolysis rates in the

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travelling air masses are key factors for determining the ozone chemical tendencies [e.g. Reeves *et al.*, 2002]. I think that a discussion on the sensitivity of the results to these parameters should be added in the paper. This should be done at least for water vapour as this appears to be a particularly important parameter [e.g. Reeves *et al.*, 2002]. The Authors should make an attempt to quantify to what extent the final concentrations of ozone are sensitive to mixing and  $\text{NO}_x$  rather than to water vapour, for example. Would that be possible to assess the validity of the water content used in the box model simulations?

6) Section 3.5. The originality of the present paper lays in the use of a suite of tools that provides insights in terms of ozone chemical terms in air masses recently exported from the planetary boundary layer and experiencing subsequent transport over medium-range distances. These kinds of calculations are usually performed with 3-D models such as the one used to prescribe initial concentrations. In Section 3.5, the Authors compare their estimate of free tropospheric ozone production with those provided by a 3D global model, which is clearly not a fair comparison, as the smaller scale processes that the Authors attempt to account for in their paper would not be included in such a global model. I think that would be much more interesting to examine whether the CHIMERE model i) suggests similar processes (e.g. export from the Spanish PBL and transport into the ESCOMPTE area, ii) is capable to reproduce the observed variability in ozone in the FT during the IOP, and iii) agrees with the estimated ozone production.

## References

Reeves C. E., et al., Potential for photochemical ozone formation in the troposphere over the North Atlantic as derived from aircraft observations during ACSOE, *J. Geophys. Res.*, 107 (D23), 4707, doi:10.1029/2002JD002415, 2002.

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