Atmos. Chem. Phys. Discuss., 6, S708–S712, 2006
www.atmos-chem-phys.org/acpd/6/S708/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

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 #1

Received and published: 2 May 2006

The paper describes LIDAR observations of free tropospheric ozone measured during the ESCOMPTE IOP2b intensive measurement period. In particular, the origin of layers with enhanced ozone concentrations at altitudes from 1.5 and 4 km between June 23 and 26 is studied in great detail. To this end, the authors use high resolution trajectories to deduce the airmass origin for these layers. Trace gas distributions in the identified source area, the planetary boundary layer of the Iberian peninsula, are simulated with a 3D CTM. To differentiate advected O3 from free-tropospheric in-situ

production a Lagrangian box model is used to calculate net O3 production along the airmass-trajectories. Sensitivity studies are performed on the influence of NOx initial conditions and mixing with free tropospheric background air.

The paper addresses an important question of atmospheric research by an elegant combination of different modelling tools. This combination of Lagrangian and Eulerian models is a particular strength of the paper. Unfortunately, the restriction of the observations to LIDAR O3 data limits the ability of the authors to unambiguously identify the cause of the enhanced O3 layers in the free troposphere. The train of arguments for a planetary boundary layer origin of these O3 layers is tempting, but I would prefer a more rigorous exclusion of other sources, e.g. downward transport from the middle or upper troposphere. Nevertheless, the paper should be published in ACP after a few specific points have been addresses:

1 The authors mention, that Dufour et al., 2005 propose a different explanation for the origin of the O3 rich layers, namely downdraft from the tropopause region. A way to differentiate between these two explanations - stratospheric origin vs. polluted boundary layer export followed by in-situ O3 formation - could be the consideration of other boundary layer tracers. The authors mention the availability of simultaneous aerosol backscatter ratios from the ALTO instrument. These data are used exclusively to correct the O3 retrivals, although they should also provide some information on the airmass origin. If the air in the O3 layers stems from the continental boundary layer, I would expect to find enhanced aerosol loadings (at least to some extent). On the contrary, upper tropospheric air or even stratospheric air should have a very low aerosol loading. Therefore I recommend that the authors check whether the aerosol data can be used to strengthen their argument for a boundary layer origin of the ozone layers. On the same topic, in the paper by Ancellet and Ravetta, 2005 reference is made to a measurement flight of the Dornier-128 aircraft in the afternoon of June 24 above the ALTO station, showing good agreement between LIDAR and in-situ O3 measurements at 2800 m. According to the overview paper on ESCOMPTE by Cros et al., 2004 this

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aircraft was equipped with an in-situ CO monitor based on VUV-spectroscopy. If CO data from this flight are available, they should be used to look for a simultaneous enhancement of the pollution tracer CO in the O3 plumes, which would also strengthen the argument for a planetary boundary layer origin of the airmasses and unambiguously exclude a stratospheric origin.

2 The authors nicely show that the calculation of the net O3 tendency in the free troposphere strongly depends on the NOx mixing ratios exported from the Spanish boundary layer into the free troposphere. They mention average free tropospheric (background ?) NOx measurements from the MONA instrument on board the ARAT aircraft, that are much higher than modelled background and pollution layer NOx concentration. Unfortunately, the reference given in the paper (Said et al., 2005) only discusses the data quality, but not the measurements itself and no other reference is made to the observations. Since NOx levels in the background air and in the plumes have such a strong influence on the conclusions from this study, it would be nice to get either an appropriate reference for the NOx data obtained during this IOP2b or include the data itself (vertical profiles) in the present paper.

Minor points:

Page 1920, lines 8-14: According to FLEXPART 33% of the free troposphere below 5 km correspond to airmasses recently extracted from the PBL. I assume that FLEXPART does not resolve subgrid scale processes such as shallow convection or orographic circulations. So what is the mechanism that is responsible for the transport of boundary layer air into the free troposphere in these trajectory calculations? Are these subgrid-scale processes included or accounted for in the MesoNH model and is this the reason, why trajectories from this model are regarded as more reliable (Page 1923, line 18)?

Page 1924, line 28: Here it is indicated that the airmasses according to the trajectory analysis are coming from a wide area all over Spain, but the authors state that nevertheless this is not contradictory with the homogenous O3 concentrations measured.

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This conclusion is not obvious and should be explained in more detail.

Page 1931, Discussion of in-situ O3 production: From an inspection of Fig 5a and 6 I got the impression that significant free tropospheric O3 production occurs only in those layers with high NOx concentrations. An additional plot showing the O3-difference between these two figures, would yield a clear view for which airmasses in-situ O3 production is a significant contribution.

Page 1932, lines 17-20: The average CHIMERE NOx concentration is lower than EMEP observations by approx. a factor of 2. To judge on the significance of this observation, standard deviations for both observations and simulations should be given. In addition, it is not clear to me how the average in the model was calculated, in particular whether NOx concentrations at the stations' altitudes are used or boundary layer average concentrations, similar to those used in the initialisation of the CiTTyCAT model.

Page 1935, line 26-27: In what sense are the enhanced NOx runs more realistic than the reference run? Figure 7e demonstrates that neither the enhanced NOx nor the reference run can simulate observed NOx (background) levels and both runs overestimate O3 concentrations in the plume, which could alternatively be the results of lower NOx, e.g. due to low export efficiency for NOx emissions from the continental boundary layer as discussed e.g. in Parrish et al. (JGR, Vol 109, D09302, doi:10.1029/2003JD004226, 2004).

Page 1936, lines 22-23 : Why is the O3 concentration in cluster 23 decreasing with increasing NOx ?

Page 1937, lines 6-23: In the discussion of the different contributions to the O3 build-up in the layers only average concentrations are given. Again to judge on the significance of the interpretation, standard deviations should also be given. My feeling is that although there is a tendency, it is not statistically significant (e.g. O3 from the Iberian PBL is 68.6 +- 12.8 ppbv, while the simulated O3 in the plume is 74.7 +- 6 ppbv, which is not significant).

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Technical comments:

Page 1924, line 3: remain in the FT.

Page 1924, line 8: I guess it should read: clear air turbulence.

Page 1924, line 14: Figure 3 is discussed before Figure 2.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 1915, 2006.