

Interactive comment on “Distribution and origin of ozone in the eastern Mediterranean free troposphere during MINOS (August 2001)” by G.-J. Roelofs et al.

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Answer to the comments and suggestions by the reviewers.

We thank both reviewers for their valuable comments and suggestions. Based on their comments we have revised the paper as follows:

General comments Both reviewers asked for a discussion of the uncertainties associated with the simulation results, more specifically with the separate contributions from different regions and from lightning. The largest uncertainties are associated with inaccuracies in the downward transport of ozone from the stratosphere (derived from previous STE studies with this model) and from the large range in estimates of NO_x emission from lightning. In section 5 we have added the following discussion:

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“Although ozone concentrations in the mid-troposphere, i.e., where O_3 s maximizes, appear to be representative, the model may overestimate mixing of intrusion air, especially downward into the lower troposphere and the boundary layer (Figure 1). Therefore, the calculated contribution by stratospheric ozone is probably an upper limit, with an estimated uncertainty of several DU. The present estimate of NO emissions from lightning ranges between 3 and 13 Tg yr⁻¹ (Prather et al., 2001), so that the actual contribution from lightning may differ by a few DU from the model estimate. We remark that the study by Scheeren et al. (2003) suggests that, for MINOS, the model underestimates enhanced tropospheric NO_y originating from lightning. The contribution from Europe may be a lower limit because biomass burning emissions in Eastern Europe during MINOS were not considered by the model (Traub et al., this issue). Additional errors are associated with other inaccuracies in the ozone precursor emission inventory, in the representation of transport and mixing processes, e.g., associated with convective clouds or synoptic disturbances, and in chemical transformation rates, but we estimate these to be relatively small. A more detailed sensitivity study is necessary to quantify the uncertainties more precisely.”

and in section 3.3 we note: “Ozone in the mid-troposphere is overestimated by 10–15 ppbv, representing about 2–3 Dobson Units (DU; 1 DU = 2.69 × 10¹⁶ molecules O₃ cm²),...”

Reviewer 2 misses a sufficient validation of the model, and reviewer 1 remarks that a more vigorous evaluation of the precursor concentrations would be required to make the results more robust. In section 2 (model description) we refer to previous studies with the chemistry-ECHAM, in which simulated transport and chemistry are validated and evaluated. These studies, which focussed not only on the climatology of tropospheric ozone but also on the ozone distribution during specific meteorological events and/or measurement campaigns, have lead to a better understanding of the model performance. The study indicates that the largest discrepancies between measured and modelled ozone are due to inaccuracies in the simulated transport associated with the

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model resolution. We note that the paper by Scheeren et al (2003) in this issue compares modeled and observed NO_y , CO and alkanes. They conclude that ozone and CO are simulated realistically. NO_y and the alkanes are underestimated by the model, probably due to too strong mixing between air from stratospheric and from lower tropospheric North American origin within the synoptic disturbances travelling over the Atlantic Ocean. A possible underestimation of NO emissions from lightning may add to this. Nevertheless, the relatively good agreement for ozone during flight 3 indicates that the photochemical part of the ozone (and, hence, the ozone precursor chemistry) during MINOS is represented satisfactorily by the model. We have mentioned this in section 3.1.

Reviewer 1, specific comments 1253, I.13: One of the factors that lead to transport inaccuracies is the vertical resolution of the model. This feature affects the model representativity in different ways at different altitudes. The vertical resolution influences the representativity of the exchange between the BL and the free troposphere, for the representation of shallow layers in the middle troposphere (in this case the different "conveyor belts" in synoptic disturbances travelling across the Atlantic), and for a realistic vertical ozone concentration gradient in the tropopause region. So, the limitations of a too coarse vertical resolution affect the whole troposphere, with overestimation of ozone in some cases and an underestimation in other cases as a result. Therefore, the coarse resolution is mentioned at several occasions in our study. We agree with the reviewer that this was somewhat confusing in the original manuscript, and we have explained this in more detail in the revised version.

1254, I.19: We meant here the small scale flow in the BL, which influences venting to the FT, the simulated height and stability of the BL and the representation of land-sea breeze circulations. However, most backward trajectories presented in our study are in the free or upper troposphere and are not affected by this. Generally, however, the occurrence of turbulent or convective events in the FT may be misrepresented in the ECMWF data so that the accuracy of the trajectories decreases with time. This may

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be also the case with flight 08, described in section 3.3. We altered part of this section to: "Backward trajectories (not shown) indicate that the air was advected from the west across southern France, northern Italy and Greece at an altitude between 600 and 750 hPa (2–4 km altitude). It is possible that as a result of shallow convection the air mass has been affected by boundary layer pollution, which also explains the relatively high concentrations of organic trace species observed in this air mass, up to ~ 3 ppb acetone, ~ 5 ppb methanol, and 700 ppt acetaldehyde (De Gouw et al., this issue), but this has not been simulated by the model in which convection is a highly parameterized and sub-rid scale process."

1255, I.9: See our remark above. We have mentioned now that the ozone plume is anti-correlated with CO, indicating that air is sampled with a significant stratospheric contribution.

1256, I.15: This was indeed confusing. Although the atmosphere between 10 and 15 km is located in the UT during most of the MINOS period, during these few days the lower stratosphere is found at these altitudes. We have changed the sentence to "is resulted in a lower tropopause height over the eastern Mediterranean region and, hence, higher ozone levels, 80–200 ppbv, between 10 and 15 km altitude."

Following the reviewer' suggestions we added information on the simulation of NO_x emissions from lightning and on the budget of the ozone cross-tropopause ozone flux. We have added a Figure 7 displaying the regional distribution of tropospheric ozone during MINOS.

Reviewer 2

As remarked above, we added a discussion on the representativity and uncertainty of the model results, and placed more emphasis on the fact that the model results are estimates. We changed the paper title to "model study of ozone in the eastern Mediterranean free troposphere during MINOS (August 2001)".

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S4, I.10: We rephrased this part to: "The concentration variability at these altitudes is mainly associated with ozone of stratospheric origin (O_3s), while ozone concentrations from photochemical origin (O_3t) are less variable between 45 and 55 ppbv, as shown in Figures 4b and 4c."

Figure 5 now displays the latitudinal distribution for August 8, corresponding with the model-observation comparison presented in Figure 1a and the backward trajectories presented in Figure 2.

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