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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-range transport of ozone and related pollutants over the North Atlantic in spring and summer" *by* S. A. Penkett et al.

Anonymous Referee #2

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Review report manuscript Long-range transport of ozone and related pollutants over the North Atlantic in spring and summer by Penkett et al.

In this paper airborne tracers have been analyzed over the Atlantic to understand the origin of middle tropospheric ozone. The authors have a long-standing experience in airborne measurements and its interpretation. The measured species are believed to be of high quality. The data are very interesting, and the middle tropospheric ozone budget remains an important issue, both for air quality as well as for climate issues. I therefore find the subject and analysis relevant for publication in ACP.

I nevertheless think that the data interpretation miss some vital tools in order to justify the important conclusions made in the manuscript concerning long-range transport

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and origin of the polluted air. In order for the paper to be published I recommend the authors to extend their data analysis with more precise trajectory calculations, the use satellite cloud data images, and ECWMF analyses. It is well known that trajectories are a powerful tool for data analysis, but their weaknesses are also known. Some of the uncertainties can be covered by using the above mentioned additional meteorological information that is easily accessible to many in the atmospheric community. An extension of the trajectory analysis is needed to address the uncertainties, inherently present because of the underlying spatial resolution and quality of the wind data. In the specific comments below you will find suggestions how to include these extensions.

Specific comments

Page 4409 Abstract and Conclusions The abstract and conclusions are not adjusted to each other sufficiently (PAN in NOy, long-range transport).

Page 4410, line 5-8 I suggest you add Waibel et al. Chemosphere Global Change Sci., 1, 233-248, 1999, who examined airborne observations of large CO concentrations in the lowermost stratosphere, attributed to long-range transport with forest fires over northern America as a possible source.

Page 4412, lines23-24 There are nine flights shown in Figure 1, not ten.

Page 4417, section 3.7 What trajectories were calculated; from 3D winds or isentropic? What is the significance of the trajectories with starting points every 100 m using a grid that has a 1x1 degree resolution? A similar question concerns how 30 km filaments can be calculated or traced with the trajectory model if the underlying source grid is 1x1 degree? Or were these calculations performed by a reverse domain filling technique? If so please specify.

Page 4419, line 1 This shows I do not understand to what this refers to, or what this has to do with trajectory precision.

Pages 4419/4420, section 4.1 Please refer to figure 2 at the start of the section. I find

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the Figure 2 as evidence for high trajectory precision not convincing. To my opinion, a test of the trajectory precision can only be achieved if a large set of trajectories is calculated along the whole flight route and the end locations correlated with the chemical data. Then one could see if the trajectories follow the chemical variability, which is to my opinion a more proper evaluation of their precision.

I further cannot find 16:30 UT. I guess you refer to a situation that is not shown in Figure 2? If so, please add this information.

I dont see how the trajectories in the right panel of Figure 2 give clear evidence of continental pollution from the boundary layer. The trajectories end well above the boundary layer and the uplift is located over a remote area in northern Canada with I presume hardly any pollution in the boundary layer?

Page 4422, lines 7-9 How do you know the ship sources are significant? Is this conclusion based on the measured profiles or on earlier performed model studies?

Page 4422, lines 17-19. The majority of the trajectories at the top of the profile no. 3 (red colored) seem to originate from the upper troposphere, rather than from the boundary layer (except for a few orange and yellow) trajectories. The starting points from the orange trajectories cannot be traced in the color plot and the yellow trajectories start at and below 4 km. Therefore, a more likely interpretation of Figure 5 and figure 4 is uplifted boundary layer air into the upper troposphere above the continent through convection. To examine this I suggest to add satellite images at the time the air parcels cross the continent to trace possible convection. Important additional information can be gained from operational ECWMF analyses of convection indicators (for example, CAPE or convective precipitation) at the same time.

Why dont you perform a similar analysis for profile 9? I find the big enhancement at 6 km much more spectacular than in profile 3. See comment below.

Page 4422, line 28 What do you mean with a total anticorrelation?

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Page 4423, lines 1-13 I would like to see more analysis and interpretation of the right panel (profile 9) in Figure 4. At line 1 it seems that an interpretation of this profile started, but I cannot find the stated positive correlation between water vapor and ozone between 3.4-5.2 km in this profile. Then it is concluded that a negative correlation between water vapor and ozone should not be used to indicate stratospheric air. The text gives the impression that this conclusion is based on profile 9. But I cannot find the structures in this profile that support these statements. Please specify more clearly to which profile you refer.

Page 4424, section 4.3 I suggest to add a figure with the profiles, like was done in the previous section. I find this section difficult to read without any profile visualization.

Page 4425, lines 15-17 The fact that convection may be less likely in spring is not convincing. As suggested above for the other flight, I believe a more thorough analysis of possible convection is needed using satellite images and ECWMF analyses at the time the trajectories cross the continent. This is especially required to justify the important conclusion that ultra-long-range transport occurred.

Page 4427, line 4. I dont understand the word even. What seasonal dependence in marked layering should we expect?

Page 4427, line 25. You mention that the low ozone near the surface is partly due to higher photolysis rates (photochemical ozone production) in summer. But since the measurements are taken in September (solar zenith angles in September approximately equal those in April), Im not sure how valid this conclusion is to explain the differences between April and September, unless the lifetime of ozone close to the marine surface is sufficiently long to have a summer memory, which I doubt is true. Could you clarify this?

Page 4428, line 25 Referring to my earlier comments, Im not convinced that the trajectory data is really clear on the origin of the air. I think such statements can only be made after more thorough analysis (as outlined above).

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Page 4429, Discussion section I think that this section will gain significantly when addressing the convection issue more thoroughly.

Page 4429, line10 Could you be more specifically concerning the transport processes? Do your observations and analysis support mainly upward transport associated with cyclogenesis, or convection over the main land? Or is it impossible to distinguish between both? Does ultra-long range transport play a role of importance in the overall transport?

Page 4429, line 26. I think the word therefore is somewhat misplaced here. The general picture (given that you refer to figure 10) does not evidently show long-range transport.

Page 4430, line 2 A possible explanation. Explanation for what?

Page 4432 You conclude that the major constituent of NOy is PAN. Earlier you stated that ship emissions provide a major NOx source in the lower atmosphere. Are these statements in conflict with each other? Could you clarify this?

Typos page 4412, line 8: In all => All

page 4419, line 5. Rephrase

Figure 3 The profile number 14 should be 12.

Page 4422, line 26: rephrase.

Page 4422, line 28, There is A total anticorrelation ...

Page 4427, line12. I suggest changing 1348 profile to profile sampled at 13:48 (UTC?)

Page 4428, line 24, I would just mention middle troposphere instead of layers between 4-8 km, consistent with lines 9-13 on page 4413.

Figure 10. You do not mention which species belong to which colors, either in the text or in the figure caption. I understand that the color legend of this figure is similar to the

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previous ones, but for clarity I suggest to add the color legend here too.

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Interactive comment on Atmos. Chem. Phys. Discuss., 4, 4407, 2004.