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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Formaldehyde over the eastern Mediterranean during MINOS: Comparison of airborne in-situ measurements with 3D-model results" *by* R. Kormann et al.

Anonymous Referee #1

Received and published: 5 May 2003

This is a very nice paper that represents a fairly thorough analysis of HCHO measurements during the MINOS campaign and their relationship with model results. These results are important for furthering our understanding of atmospheric chemistry, and this paper should be published after the authors address a number of issues that are detailed below.

Page 1304, Abstract: The authors should indicate that the 42-pptv limit of detection is at the 1s level.

Page 1305, lines 11 & 12: The authors should indicate that the ~ 4 hour lifetime is near the surface and the lifetime is even lower with increasing altitude due to enhanced photolysis rates.

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Page 1306, line 8: The Wagner et al. actually achieved very good measurement-model agreement on average and the indication of a model overestimation for this study needs to be changed. Two lines down the statement that in the middle to upper free troposphere "models tend to systematically underestimate the HCHO concentrations" needs to be modified slightly to read "models generally tend to systematically...". This revised statement accounts for the fact that Fried et al. [2003] found on average very good agreement during TOPSE.

Page 1307, 3rd line from bottom regarding the forward facing inlet tube: The authors should comment on what precautions if any were taken to avoid sampling liquid water into their instrument. Liquid water can either take up HCHO or re-emit this gas depending upon ambient and inlet temperatures and other factors.

Page 1308, first several lines regarding calibration: The authors should indicate where in the system the liquid standards and the 1 in-flight calibration were added. The optimum approach is to add the standard near the inlet tip in front of the pump. Was this done? What tests were carried out to insure that there was no loss of HCHO in the pump nor did the pump act to temporarily soak up HCHO and re-release it later? Why was only 1 in-flight calibration carried out? Ideally, one should add gas phase standards frequently throughout each flight on both zero air and ambient air to constantly insure no inlet loses. Like the calibration, the authors need to comment on where the zero air was injected. Was the entire inlet zeroed or just a portion? How was zero air generated? Was the output flow from the Hopcalit (check on the spelling of Hopcalit) trap re-introduced back into the inlet or was a zero air cylinder used? In some cases zero air cylinders may have as much as 500 pptv of HCHO in them and this would cause a systematic error in the reduced HCHO measurements. How frequently was zero air introduced into the system? The particulars of zeroing and calibration can be very critical for highly accurate airborne measurements of HCHO, particularly when comparing measurements and models at very low ambient concentrations. These issues should not deter publication of this paper, but the authors need to acknowledge

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wherever applicable the potential problems that may be present in their data due to their zeroing and calibration methods.

Page 1309, in the discussion of the O3 effect: This discussion is very nice and the authors are to be commended for taking care in assessing the affects of O3 on their measurements. I would suggest in the 10th line from the bottom of the page adding "on the inlet walls" after "due to reactions with O3..." In the 5th line up from the bottom I would add "in the laboratory studies" after "during the O3 production process" to reaffirm the belief that the intercept is most likely an artifact of the laboratory tests.

Page 1311 end of 1st paragraph: The authors should mention that the 280-pptv concentration reported by Fried et al. [2002] is where the air mass showed clear evidence of anthropogenic inputs; all the measurements both clean and polluted produced an a combined average of 265 pptv.

Table 2: There is a typo in the altitude bin; change 8-97 to 8-9.

Figure 3: Since this figure is so important and gives the essentials of this paper, I would advise making this figure a little bigger for readability.

Page 1313, 11th line from bottom: The acetone effect on HCHO will also depend upon the NO2/NO ratio and not just the NOx level since the amount of PAN formation and the competing CH3 formation (removal of CO2) from the CH3C(O)OO radical will depend upon the NO2/NO ratio. This should be added in the text.

Were model results run with a simple box model using measurements of NMHCs as input to eliminate the effects of incorrect emission inventories used in the 3D model, and more importantly, the effects of vertical convection? Somewhere in the model description on page 1311 the authors should indicate the inputs to the model.

It is also interesting to note in Fig. 4 the high NO concentrations and its disparity with model results. Since the reaction of NO + CH3O2 governs the rate of HCHO production, the elevated NO levels might very well be playing a role in the extra HCHO

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production. Were measured or modeled NO values used in the model calculation of HCHO? Perhaps the authors should comment on the elevated NO levels. Also a time series of HCHO along with NO would be very convincing in showing in situ production from some VOC. Perhaps the authors should consider adding such a figure.

Discussion, 1st sentence: Referring to comment on Page 1306 line 8 above, the statement needs to be modified slightly to read "state-of-the-art model predictions are generally consistently lower than observations"

Table 3 caption: Should include here the fact that acetone was increased in the 3D model.

Page 1314, 6th line from bottom: Change "during the whole period..." to "during the early deployments at low light levels...". Correct the spelling of artifacts in last line on this page and in lines 9 and 12 on page 1315.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 1303, 2003.

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