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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Satellite measurements of formaldehyde from shipping emissions" *by* T. Marbach et al.

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First of all we want to thank this reviewer for the positive assessment of our manuscript and the interesting comments.

Major changes have been applied to the manuscript. Before we address the reviewer comments in detail, we give a short overview of the two major changes of our manuscript.

a) The discussion of the origin of the enhanced HCHO values is completely changed. We now discuss in detail (especially in section 3) the findings of this study compared to those from the previous study on NO2 emissions from ship traffic (Beirle et al. 2004). By comparing the observed absolute values and the peak shapes and distances from

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the ship route, it becomes obvious that neither direct emissions of HCHO, nor direct emissions of NMHC can explain the observed HCHO values. Instead, we conclude that increased degradation of CH4 is the most probable cause for the enhanced HCHO values. Increased CH4 degradation is caused by increased OH concentration inside the ship plume, and the intermediate oxidation products (with lifetimes of several hours) are then transported away from the ship route. In addition, also outside the emission plume enhanced OH concentrations might occur due to increased ozone concentrations. Besides in section 3, we added these findings and conclusions to the abstract and conclusions. We also changed the title to reflect the fact that the observed increase of HCHO is very probable not caused by direct emissions.

b) We investigated the potential influence of aerosols on the satellite observations in more detail and discuss the related findings in the manuscript (section 4). First we carried out additional radiative transfer simulations using various aerosol properties. It turned out that even for rather high aerosol optical depths the change of the AMFs are < 50% and could not explain the observed HCHO increase assuming typical background concentrations of HCHO. It is interesting, however, to note that large enhancements of the AMF occur only for scattering aerosols (single scattering albedo close to unity) which are probably not adequate to describe aerosols from ship emissions (Coakley and Walsh: Limits to the Aerosol Indirect Radiative Effect Derived from Observations of Ship Tracks. Journal of the Atmospheric Sciences, 59, 668–680, 2001). Nevertheless, even if aerosols from ship emissions were purely scattering aerosols with high optical depth, the resulting high top of the atmosphere radiance should be clearly visible, e.g. in aerosol products from satellite observations. Second, we analysed the simultaneous observed O2 absorption in the satellite spectra and applied the same averaging and filtering as for the HCHO data. The resulting maps did not show a similar pattern as found in the HCHO satellite data indicating that the enhanced HCHO values are not an artefact caused by modifications of the tropospheric AMF. We added this information to section 4.

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Detailed replies:

Reviewer comment: “This paper presents satellite retrievals of formaldehyde, in particular over the shipping lanes between Sri Lanka and Sumatra. The Method section is well-written, describing the retrievals, as well as presenting some validation results. However, the modelling component of this work does not seem to add much to the analysis; many questions remain about the cause of the differences between the model and the observations. The question of whether model resolution or emission inventories cause the lower modelled amount might be addressed by comparing the model NO/NO2 with GOME NO2 observed along this track.”

Author reply: We agree that due to the coarse spatial resolution of the model, the comparison of our results to model simulations is of limited value. Especially, the complex and non-linear in-plume chemistry can not be well represented in such models, which leads to systematic deviations of the determined trace gas distributions. We made this clearer in the revised version of our manuscript (beginning of section 4). Nevertheless, we still think it is interesting to perform such a comparison, especially because these models are often used to investigate the impact of emission sources on the global trace gas concentrations. Such models also allow to study the effect resulting from switching on or off specific emission sources (see Hoor et al., 2009). As stated above, in the revised version of the manuscript, we discuss the chemical implications of our measurement results in more detail, especially in relation to the simultaneously retrieved NO2 distributions. We added this information to section 3, to the abstract and conclusions.

Reviewer comment: “Also, some model sensitivity studies, such as where the ship emissions of HCs are turned off, would be interesting. More examination of the model results of other compounds, illustrating the perturbation to OH, etc., along the ship track would provide useful justification of the discussion on p. 10498.”

Author reply: We agree and we added a reference to a recent paper (Hoor et al.,

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2009), where such sensitivity studies were carried out. Indeed they found that ship emissions have a large effect (increase) on the global distribution of the OH radical in the boundary layer. We added this information to the manuscript (section 3). From the EMAC model we also calculated the change in the CH4 degradation due to ship emissions (see below). It clearly shows increased CH4 degradation along shipping routes, especially over the area investigated in our study. We added the resulting map (Change in the CH4 degradation due to ship emissions derived from QUANTIFY), below, showing a CH4+OH yield up to 0.7 ppbv/day. Because of the limitations due to the coarse model resolution, we are a little bit hesitative of including this map in the revised version of our manuscript.

Reviewer comment: "A more technical point, I found the description of the model output that was used rather confusing. It is stated that output is archived as 5-hourly instantaneous fields and later, that the monthly mean is filtered for local time between 10 and 11am. Does this mean the monthly mean does not include a value for each day of the month (because of the 5 hour output)? It would be much better if the output could be coincident in time with each satellite overpass. If this is the case, it should be stated more clearly."

Author Reply: Concerning the model there is no output every day (4 consecutively output days, with 6 days without output) for the time between 10 and 11 am because of the 5-hourly instantaneous fields archive from the EMAC model. EMAC has also the possibility to deliver hourly data but only after 2002 which is out of range from our study. Nevertheless, before using the 10 and 11 am output, we worked with daylight outputs of the EMAC model which did not show a perceptible difference with the output time between 10 and 11 am (low diurnal cycle in the model). This consistence with the daylight output makes us confident to use the more restricted output which also somehow reflects the gaps of the GOME daily coverage (every 3 day at the studied latitude). We added the following sentences to section 4: 'Note that this filtering, together with the 5-hourly read-out of the model data leads to gaps in the retrieved tome series. We

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checked that these sampling effects had no significant influence on the comparison.'

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 10487, 2009.

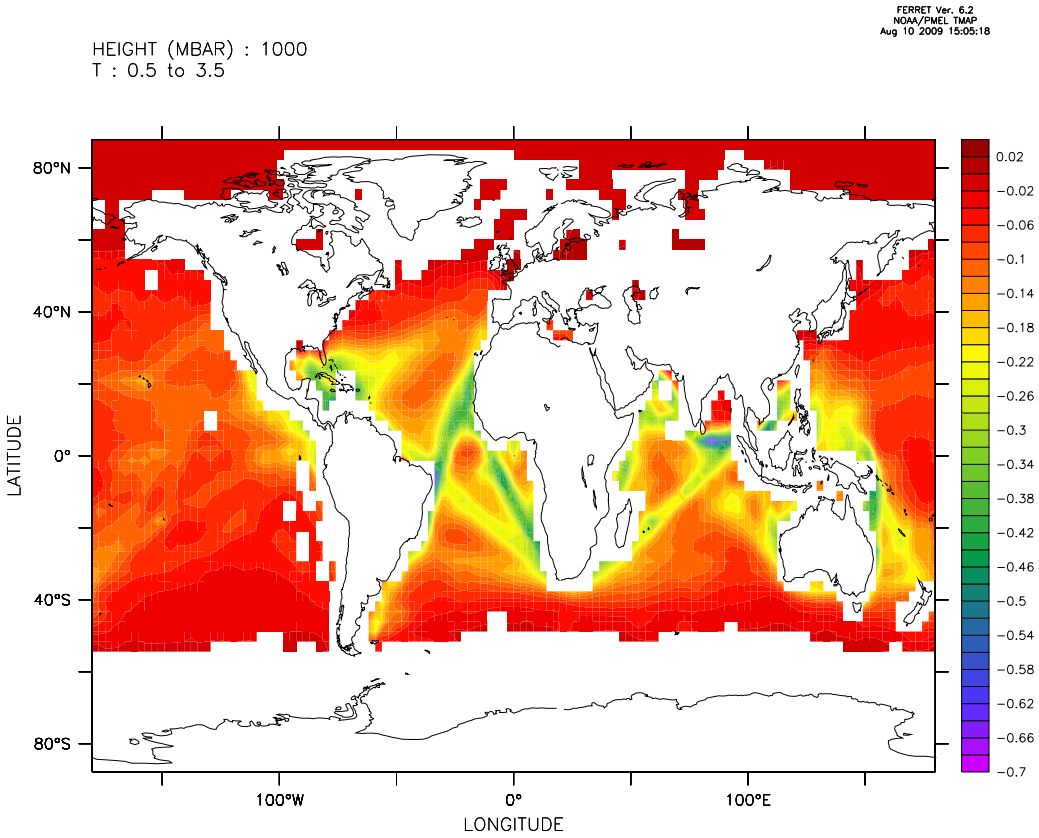

Fig. 1.

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