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Atmospheric Chemistry and Physics Discussions



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

**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 very constructive suggestions. We followed most of them (see replies below) and give detailed explanation in cases where we disagree.

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

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

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. 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.

Detailed replies:

The manuscript describes an investigation using GOME to examine HCHO from ship-



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ping emissions. It includes a detailed description of the HCHO GOME retrieval. A brief comparison is presented with MAX-DOAS observations in Milano. High pass filtering is used to investigate HCHO over the ship corridor. The results are compared with a model. The manuscript would be more appropriate for AMT as it mostly describes a measurement investigation. Several issues need to be addressed before publication in either ACP or AMT.

Author Reply: Of course, publication in AMT would be an option. However, we feel that the most important outcome of our study is the results on the amount and distribution of atmospheric HCHO mixing ratios related to ship emissions. These results should be of interest for the broader scientific community. Therefore we think publication in ACP is an appropriate choice.

A high-pass filter is applied to GOME data to look for HCHO from shipping emissions. The technique of Beirle et al. 2004 was valuable at the time, but is less relevant now that high-resolution satellite data are available. Why is OMI not mentioned? The OMI pixel size is much better suited to look for a small feature like a ship track. The OMI data might eliminate the need for a high-pass filter.

Author Reply: In principle we agree that similar studies should be made also for data sets from other satellite instruments, in particular for OMI data. Besides the better spatial resolution, OMI observations also have a better (daily) coverage. However, we think that HCHO observations from GOME are also a good choice: a) The GOME HCHO data set was directly available from DOAS analyses made in our group and covers a long time span. b) Because of the orientation of the GOME-pixel, the resolution in north-south direction is already rather high (40km) for the GOME dataset. From the results shown in Fig. 4 it becomes obvious that the width of the HCHO peaks is much larger (in the order of 100 to 200km). Thus GOME data seem to be very well suited for studying HCHO from ship emissions on that ship track with the world highest ship density on open Ocean.



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By the application of the high-pass filter it is possible to discriminate the weak HCHO signal associated with the ship tracks from the rather strong HCHO signals from other sources (in our case strong continental sources of HCHO). Even in future studies using satellite observations with better spatial resolution, we would recommend the application of a similar high-pass filter.

We added this information to section 3 of the revised version of our manuscript. We also included a hint to OMI (and SCIAMACHY) at the end of the conclusions.

Figure 5. The high-pass filter reduces HCHO VCDs over the ship corridor in EMAC from 5.5x1015 in c) to 4x1014 in d). Why? The change in GOME VCDs from a) to b) is much smaller over the ship corridor. An explanation is needed to justify the conclusion that the modelled HCHO values over the ship corridor are lower than in the GOME high-pass filtered data.

Author Reply: Many thanks for this hint! The difference can be easily explained by the different 'background' values in both data sets. The GOME data have been normalized over a reference region (see chapter 2). Thus the 'background' SCD values are close to zero, while the model 'background' value over the same selected region (central Pacific) as for the GOME normalization is about 3.6e15 molec/cm2. The scales of the GOME data (Fig. 5a) and model data (Fig. 5c) have been thus chosen such that in both cases the 'background' values appear in the same colour. Nevertheless, the enhancement over the ship tracks is not so different in both data sets: about 3.5e15 molec/cm2 for the satellite data, and about 2e15 molec/cm2 in the model data. After high-pass filtering, the respective values are about 7e14 molec/cm2 for the satellite data (Fig. 5b), and about 4e14 molec/cm2 for the model data (Fig. 5d). This ratio is similar to that of the enhancements over the 'background' values. We added more information in section 3 and in the figure caption of Fig. 5 to avoid these misunderstandings.

How do aerosols from ship emissions affect the air mass factors? This should be discussed. Could the signal in the SCDs arise from increased aerosol scattering, rather



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than from HCHO? Do satellite observations of aerosol (such as MODIS) show a signal along the ship corridor?

Author Reply: This is an important point. We addressed it in two ways: a) First we made sensitivity studies using different assumed aerosol properties. The respective changes of the HCHO AMF could not explain the observed enhancements in HCHO. b) Second, we investigated the observed radiance and O2 absorptions over the region of interest. As proxy for the radiance we used the HICRU cloud fraction which is proportional to the top of the atmosphere radiance, but no systematic was found. From the investigation of the O2 absorption, also no systematic feature was found, indicating no systematic aerosol effect on the tropospheric AMF.

We added this information to the revised version of our manuscript (section 4).

The authors speculate in the abstract and conclusion about reasons that the modelled HCHO values over the ship corridor are lower than in the GOME data. The reasons stated are that the emission inventories are too low and/or that there is in-plume chemistry not represented at the model resolution. Two other possibilities that should be noted are the effect of the high-pass filter, and errors in the GOME retrieval.

Author Reply: We agree that both aspects can have an effect on the comparison. However, as stated above, the effect of the high-pass filtering should be rather small. In contrast, uncertainties of the GOME retrieval might cause at least part of the discrepancies. We made this clearer in the revised version of the manuscript. However, we think that the most probable reason for the differences is largely related to deficiencies of the representation of the in-plume chemistry. Due to the coarse model resolution, the complex in-plume chemistry can not well be represented and can cause systematic errors of the resulting trace gas distributions. Similar findings were reported also from other studies (e.g. Wang et al., Environmental Science & Technology “Improving Spatial Representation of Global Ship Emissions Inventories”, 2008). This study shows that the ship activity patterns from ICOADS and AMVER show discrepancies that could



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significantly affect the accuracy of the ship emissions inventories. Especially the paper shows an underreporting to ICOADS and AMVER by ships near coastlines. We added this information to the revised version of our manuscript.

The phrase good agreement in the validation needs to be better justified. Statistics such as correlation or RMS difference are needed. The statement about clouds is not very convincing. The difference on 21 August is about a factor of two. The cloud fraction on that day is 0.15 which is smaller than the threshold referred to elsewhere as cloud-free.

Author Reply: We are aware about the fact that our validation exercise is only based on very few observations. Thus, adding statistics on the correlation or RMS difference might not give much more insight. However, we calculated these quantities (R2 of about 0.6 between the MAX-DOAS and the GOME data) and added this information to the revised version of our manuscript (in the text and in the legend of figure 2). In our opinion, the most important conclusion of our validation exercise is that the HCHO VCDs from both data sets show rather similar absolute values. Besides cloud effects, the disagreement between individual data points could also be related to possible spatial gradients of the HCHO concentration, which are not resolved by the coarse spatial resolution of the GOME pixels. We added this information to the revised version of our manuscript.

The last paragraph of section 3 speculates on the difference between NO2 and HCHO. One proposed explanation is the difference in grid sizes. This effect should be checked.

Author Reply: The grid size has only a negligible effect because the HCHO peaks are much wider than the grid resolution. We added this information to the end of section 3. In the revised version of our manuscript we discuss the differences between NO2 and HCHO in more detail. Besides the differences in the absolute amplitudes, also the distance of the peaks from the location of the ship tracks holds interesting information on the formation process of HCHO (see above).



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