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Atmospheric Measurement Techniques Discussions

Interactive comment on "Ship emissions of SO₂ and NO₂: DOAS measurements from airborne platforms" *by* N. Berg et al.

N. Berg et al.

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Anonymous Referee #2 Received and published: 6 December 2011

General impression: It's a pity the authors did not finish the AMF simulation, before submitting. The authors spend a lot of time and money on the performance of very interesting measurements. Also the discussion seems to be well done including the modeling of the ship emissions for individual ships. However, they did not spend an adequate time on the radiative transfer simulation. Instead they tried to estimate the geometric light path by assuming a reflection of the light path at the water surface and

included additional information about the water waves. Indirectly they thereby assumed that the incoming radiation is completely homogenous – no angle dependency. I am afraid, that this is a big mistake, as the incoming radiation is dominated by the sun's position.

Therefore I recommend to estimate the light path by using a radiative transfer model, at least for some selected cases. Cases such as "Maas Viking" or "Saalitius" showing a high discrepancy between DOAS measurement and model simulations might be good examples perhaps the difference can be reduced by using a better AMF. Another example might be the measurement in the plume of "Stena Hollandica" as shown in figure 12, it seems the SCD depends on the viewing direction.

Answer: We agree with the referee that there is need to carry out radiative transfer modelling but in our mind this has been pointed out very clearly in the paper, discussing the impact of waves and direct scattering in the plume as main error sources and the fact that we don't entirely understand these. For instance in the abstract, section 3.2, section 6.1, 6.2 and section 7. We clearly state that our assumption corresponds to wind free conditions and then the discussion is a discussion about the magnitude of the uncertainties. We realize that the discussion has caveats and simplifications AND MAYBE SHOULD SIMPLY REMOVE MOST OF THE DISCUSSION IT AT THIS STAGE, NOT TO PRETEND WE UNDERSTAND THE PROBLEM, PLEASE ADVICE.

Ray trace modeling for our application is a complex problem with the largest uncertainty corresponding to the reflection in the water, (specular or diffuse on water particles) combined with plume scattering properties taking into account the solar position and its relative strength compared to the diffuse sky. To our knowledge there are no good models working directly on this today and therefore it will not be possible to carry out any descent modeling within the next 6-8 months. In an ongoing project we have just started preparing for this type of work by collecting particle content and refractive index

of ship plumes through particle measurements. We hope in the next year be able to write a second improved paper on this topic.

However, in our mind, this paper describes a feasibility study with several qualities that makes it suitable for publishing, even though we have not theoretically solved the uncertainty problem.

First of all, this is the first time the DOAS method has been used in this type of application, measuring gas emissions from a ship by using ocean scattered solar light. The results shows that the sensitivity is enough to detect SO2 and NO2 observing the plume only for a few seconds. The trick here is that we have optimized the system for photons by using a 30o angle of the telescope and a very large liquid guide fiber (5 mm) together with a UV sensitive spectrometer.

Secondly, the paper also, for the first time describes an approach how to obtain gas fluxes from a travelling ships by optical measurements from the air. Important here is that one need to calculate the apparent wind (vector sum of wind speed and ships speed and their direction).

Thirdly, the paper shows a validation study, comparing on board measurements with the one obtained by DOAS measurements from a helicopter, table 4. It shows an agreement within 30-40% which is consistent with a rough error analysis made in the paper. In our mind this replaces some of the lack with the uncertainty budget calculation.

Fourthly we also show a comparison of a ship emission model and DOAS measurements onboard an airplane, Figure 14, 15. The comparison shows that the so2 emission measurements and SO2 model results correlate reasonably well. By combing SO2 measurements and modeling of CO2 emissions it seems feasible to discriminate between ships having 1% sulfur in their fuel from ships having 0.1% (new legislation on the baltic sea 2015). Again, this study indicates that the DOAS approach may roughly indicate whether a ships is a gross polluter or not which is encouraging for the idea that airborne DOAS measurements could be used for first alert measurements. WE WILL

ADD SOME MORE ON THIS.

Lastly, We believe it is somewhat unfair that the reviewer requires we solve all problems at once before publishing, when compared to other studies. Forinstance the DOAS instrument by Schiamachy and MAX DOAS measurements are fighting airmass factor and in the past many papers have emerged without a perfect solution to this problem.

WE ARE WILLING TO MAKE ADJUSTEMENTS IN THE TEXT TO MAKE CLEAR THAT THIS IS A FEASIBILITY STUDY WITH THE NEED FOR BETTER UNDERSTANDING OF UNCERTAINTIES REGARDING AIRMASSFACTORS BY RAY TRACE MODELLING, WE COULD ALSO REMOVE SOME OF THE DETAILED RESULTS IF REQUIRED. WE HENCE ASK THE REVIEWER FOR MORE SUGGESTIONS ON THIS TOPIC

Regarding some of the reviewers more detailed remarks above: A) , To our knowledge, at 300 nm (Eyvind Aas, Remote Sensing of Environment, 1999) a large part of the irradiance at the ocean comes from the diffuse sky (60-70%) rather than direct sunlight and therefore the impact of solar direction will be less pronounced than what the reviewer claims. This is discussed in some cases in 6.1. but to investigate this properly we need ray trace modelling.

b), We have tried to investigate whether the Stena Hollandica data in Figure 12, and quantitatively in table 4 depends on viewing direction, but we have not found any good correlation, contrary to what the reviewer claims.

In the DOAS analysis the authors included the Ring cross section and the respective trace gas, SO2 or NO2, however it seems no additional trace gases were included. From my experience it is highly dangerous to do a DOAS analysis for SO2 without including ozone. Please re check the analysis, in this case I would not expect a big difference as the time difference between the reference and the measurement is of the

order of a few seconds. However, if the NO2 concentration is high you might observe a reduction in ozone, which might be interesting as well.

Answer: We have previously included also ozone in the retrieval but with no drastic change of results. It was later removed to minimize the degrees freedom in the fitting procedure, to improve the S/N for the measurement. Consider that the plume is only intercepted during a few seconds, and the time difference between the plume measurements and the reference measurements is very short. Hence will there be no change in the background atmospheric column of ozone, although the background ozone inside the ship plume may be lowered due to titration by NO. If one assumes that all ozone (20-30 ppb at sea) is removed and that the path through the plume is 100 m then the negative ozone value will be -2 ppm*m. This corresponds to a differential optical depth of 2e-5 at 310 nm, which is about 20 times lower than the noise level which is 1e-3 due to short integration time. By sniffer measurements we have also observed that titration at sea is surprisingly slow and most of the NOx still remains as NO even several kilometers downwind.

WE WILL ADD SOME MORE DISCUSSION IN TEXT

Comments: P6277L13 The instrument consists of two spectrometers; however to me it is unclear whether one telescope or two telescopes are used. Please clarify: “The spectrometers are connected to a quartz telescope” one telescope for both, or “Each spectrometer is connected to a quartz telescope” two telescopes for two spectrometers.

Answer: There is only one telescope and one spectrometer that is used for either SO2 or NO2, by changing the wavelength region. WE WILL CLARIFY.

P6277L20 “Longest exposure time. . . ” for which of the two spectrometers or for both. Answer: see above.

P6287L1-9 You did not include any other cross section, only Ring and SO2 / NO2?

How about Ozone? See general comment. Answer: see answer above.

P6280L11-13 seems correct but why don’t you use a vector equation: vaw = vship + vwind Answer: The reviewer is correct but we prefer the explicit expression and believe it is a matter of choice one can make.

P6281L13 The position data might be the first thing, of interest, in figure 12 it seems there might be a slight temporal offset between the position and the SCDs, otherwise the enhanced SCD north of the ship track cannot be explained. The roll angle of the plane is not mentioned but surely included in this list, this might be essential for the AMF calculation. Answer: Correct, but during the flights we made certain that the airplane was balanced (not tilted) while transecting the ship plumes. WILL ADD THIS IN THE TEXT. - P6286L1 you removed equation 10, as I recommended, please also remove the reference to the equation.

Answer. Will be changed.

P6288L11 Add “In preparation” to the reference Mellqvist and Berg, 2011 Answer. Will be changed.

P6298 F1 and 2 Please use wavelength in nm for the X-axes. Answer. Will be changed.

P6309 F12 Can you remove the blue background from the picture? Answer. We prefer to keep it is in this way due to software issues.

P6312 F15 The legend is incomplete; the red bar is probably model data as in the previous figures. Answer. Will be changed

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