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Interactive comment on "Measuring SO_2 ship emissions with an ultra-violet imaging camera" *by* A. J. Prata

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General comments:

The submitted manuscript describes the use of ultra-violet imaging cameras for measuring SO2 emissions from seagoing ships. This measurement technique relies on the quantification of the absorption of scattered solar UV radiation by SO2 molecules in the ship plumes. The method has been applied to volcanic plumes with considerable success in the past, although emission rates from volcanoes are typically orders of magnitude higher than those from ships. The manuscript is well written, easy to follow, and the data and results are clearly presented. The reader is nicely introduced to the topic, appropriate references to previous studies are given, and new and original

material is then presented.

I do have one major concern with regard to the measurements: It is standard procedure to measure SO2 in plumes using its differential absorption signature. This means that the absorption of SO2 is measured at a wavelength at which it is prominent (typically close to 310 nm), then normalized by the absorption measured at a second off-band wavelength. The differential method is used to make the measurement selective towards SO2. In the absence of a normalization wavelength, the measurement is influenced not only by SO2, but also by any species that absorbs radiation at the measurement wavelength. Even more problematically, absolute absorption measurements are also influenced by scattering and absorption of aerosols (such as soot particles or sulfate aerosol) in the plume. It is well known that soot is often a major component of ship exhaust plumes. Therefore, it is very unfortunate that this study was conducted using only a single wavelength. This is likely to be a very significant source of error. The author recognizes this fact and states it repeatedly, yet without additional measurements, it is difficult to quantify. Perhaps some of the suggestions given below can help to this extent. In any case, this appears to be a crucial issue, and while the manuscript is clearly a feasibility study, the issue needs to be dealt with in some way and careful resolution of the matter in future measurements should be a central conclusion of the investigation.

Specific comments

P9469 L15 – Perhaps also mention that these imaging techniques have also been applied to industrial stacks and cite this pioneering study: Harold B. McElhoe & William D. Conner (1986): Remote Measurement of Sulfur Dioxide Emissions Using an Ultraviolet Light Sensitive Video System, Journal of the Air Pollution Control Association, 36:1, 42-47, http://dx.doi.org/10.1080/00022470.1986.10466043

P9470 L2 – "Preference is for S content at 0.1 % or 0.5% for ships in harbours or at berth". Please clarify. Surely the preference is not for a higher S content when in the harbor?

P9471 L5 – Please consider changing the ordering of the different sections of the manuscript. The description of the measurement campaigns might better supersede the methods section. This way, the campaign descriptions would be closer to the results obtained in each. Alternatively, the paragraph between P9472 L3 and P9473 L5 could be moved to the results section. As it stands, this paragraph presents results obtained using methods that have yet to be described.

P9471 L20 – Perhaps it is better to refer to "SO2 emission rates from ships" rather than "SO2 ship fluxes".

P9471 L26 and Figure 1- The figure looks quite nice but is not quantitative. This image might be left out of the manuscript considering that other, more quantitative images follow. If it is to be kept in as an example of a raw absorption measurement (pre-calibration), a color scale should be added to the bottom image giving the magnitude of decrease in intensity relative to the background (obtained outside the plume). Be sure to give the 'cutoff value' below which the pixels are not assigned a color. In the current image (bottom), it looks as though the plume actually expands somewhat beyond the area that is highlighted in color.

P9473 L1 and elsewhere – It is mentioned several times in the manuscript that plume velocities are measured by tracking plume features within a sequence of images. Please explain how this was done. Was the tracking performed manually (this is often possible), or was an automated tracking method used? If so, which one?

P9473 L23 – If the SIRENAS-G campaign is not covered in this manuscript, it doesn't really need to be introduced, but perhaps could just be mentioned in the outlook on future work.

P9474 L12 – In table 1, please state that the quantum efficiency is 30% AT 300nm.

P9474 L13 - You refer to an unidentified figure here. The reference should be to Figure

6.

P9474 L15 – It is not clear why a camera with a frame rate of better than 100Hz is needed to measure a weak absorption signal. The signal to noise ratio would appear to be the deciding factor here, and this is governed by the quantum efficiency rather than the frame rate.

P9475 L9 – I would not recommend the term 'backscattered UV light' here, as this term is usually used when active light source is used to illuminate the area of interest, and radiation is scattered back in the direction from which it originated. Instead, perhaps use 'scattered solar UV radiation'.

P9475 L13 – You state that a range of 5 km provides acceptable signal to noise. Please explain how and why the range affects the signal to noise ratio. Also, perhaps it is better to use a distance shorter than 5km as an example in your calculation of pixel size, considering that most of your measurements appear to be made at ranges of just a few hundred meters.

P9475 L17 – Here, 'light diminution and enhancement' along the light path are listed as perturbing effects and this is true, but they are actually caused by the other effects in the list. Perhaps rephrase as 'light diminution or enhancement caused by. . .'

P9476 L8 – It is true that multiple scattering invalidates the simple Beer-Bouger-Lambert approach, but so does single scattering if it occurs between the instrument and the region of interest. Please clarify.

P9477 L16 – Please specify the type of filter used. I assume it was a colored glass bandpass filter?

P9479 L6 – The first two sentences in section 3.4 are difficult to understand and perhaps not formulated correctly. For example, it's not clear to me why the light intensity in the vicinity of the plume is assumed to be the main contribution to light entering the plume. Also, I don't think that scattering on SO2 molecules is taken into account. This effect would likely be similar in magnitude as scattering on air molecules, and is explicitly excluded from the Beer-Bouger-Lambert approach. Finally, I also don't understand why the field of view of the camera influences the validity of the assumptions. The radiative transfer of the scene certainly cannot be influenced by the camera.

P9479 L13 – A variation in intensity across the CCD diameter can indeed be caused by vignetting, in which case a reduction in the camera's aperture would improve the behavior. Of course this also reduces the light throughput and therefore negatively impacts the signal to noise. Oftentimes, another effect is also relevant: Because bandpass interference filters shift their transmittance wavelength window towards shorter wavelengths when illuminated in a non-perpendicular direction, they transmit shorter wavelengths to the edge of the image than to the center (depending on the optical setup). Because the UV solar scattered spectrum falls off towards shorter wavelengths, less light is transmitted to the CCD. This effect looks like vignetting, but actually is caused by a change in the filter transmittance wavelength, which unfortunately can also bring a change in calibration with it. Though this effect is probably small in this setup due to the narrow field of view, it is perhaps worth mentioning because if the filter is placed behind the lens in an instrument setup, reducing the aperture will actually increase the magnitude of the unwanted effect (see Lübcke, P., Bobrowski, N., Illing, S., Kern, C., Alvarez Nieves, J. M., Vogel, L., Zielcke, J., Delgado Granados, H., and Platt, U., 2013. On the absolute calibration of SO2 cameras, Atmos. Meas. Tech., 6(3), pp. 677–696, doi:10.5194/amt-6-677-2013 and Kern, C., Werner, C., Elias, T., Sutton, A. J., and Lübcke, P., 2013. Applying UV cameras for SO2 detection to distant or optically thick volcanic plumes, J. Volcanol. Geotherm. Res., 262, pp. 80–89, doi:10.1016/j.jvolgeores.2013.06.009. for a more detailed explanation)

P9479 L15 – It is unclear what the advantage of a fast-sampling camera is when reducing the aperture.

P9480 L17 – Again, how was the tracking performed?

P9481, L11 – Please give the focal length and chip size here, or refer to table 1.

P9481 L15 – Again, consider using a shorter distance.

P9482 L4 – Please clarify: 'results were best only under certain geometries'.

P9482 L13 – Please expand this section by a sentence or two. How was the calibration performed? Was the stack height measured in the imagery?

P9483 L14 – 'It is also assumed that the plume is free of particulates'. In my opinion, this is the single most important (and likely erroneous) assumption that is made. Sea going ships put out significant amounts of black carbon (soot), see e.g. studies by Daniel Lack et al:

Lack, D. A., J. J. Corbett, T. Onasch, B. Lerner, P. Massoli, P. K. Quinn, T. S. Bates, D. S. Covert, D. Coffman, B. Sierau, S. Herndon, J. Allan, T. Baynard, E. Lovejoy, A. R. Ravishankara, and E. Williams (2009), Particulate emissions from commercial shipping: Chemical, physical, and optical properties, J. Geophys. Res., 114, D00F04, doi:10.1029/2008JD011300.

Lack, D., B. Lerner, C. Granier, T. Baynard, E. Lovejoy, P. Massoli, a. R. Ravishankara, and E. Williams (2008), Light absorbing carbon emissions from commercial shipping, Geophys. Res. Lett., 35(13), L13815, doi:10.1029/2008GL033906.

Simply assuming the plume is free of particulates seems like a poor choice. This assumption is only necessary because, contrary to the standard UV SO2 imaging procedure at volcanoes, only a single wavelength channel was used for the measurements presented here. This is quite unfortunate, but of course cannot be changed in retrospect. However, the use of at least 2 wavelength channels to account for particulate absorption in future measurements should be one of the most important recommendations of this feasibility study. Please make sure this comes across – it is currently not emphasized enough.

In the absence of any additional information, one approach to dealing with this problem could be to perform a sensitivity study based on measurements by others. The references given above e.g. include information on the typical abundances and optical properties of black carbon in sea going ship emissions. Using that data, an estimate of the errors associated with assuming a particulate-free plume could be made.

But I wonder if there might be some information available in the visible imagery. Though the author states that there were 'no visible signs of particulates in the plume', some of the visible-light RGB images reported in the manuscript appear to indicate that there might be some visible absorption. While it could just be an artifact of our printer, Fig 1 (top) and Fig 15 (top) both seem to show a very faint visible plume coming off of the ship stacks. It is quite difficult to grasp in the RGB image, but an attempt at getting some quantitative measure of particle absorption might be obtained by analyzing a monochrome version of the image. Preferably, the blue channel alone would be extracted from the photograph and analyzed in much the same way that the UV images are analyzed (i.e. drawing a cross-section through the plume and deriving the absorbance relative to the background). This channel is not as close to the employed UV channel as would be ideal but aerosol scattering and absorption cross-sections tend to vary only slowly with wavelength and it might at least give some quantitative information about aerosol absorption.

P9484 L16 – Here the presence of particulates is acknowledged. See above comments.

P9485 L8 – Really 'no visible signs'? See above comments.

P9485 L9 – It is unclear how a large distance to the plume could lead to an overestimation of the emissions. Please clarify.

P9487 L21 – It is currently standard practice in volcano applications to use 2 UV imagers, each measuring in a different wavelength. Please cite some literature to this extent.

P9487 L24 – Please clarify what you mean by 'minimizing coincidence'. Wouldn't you want to maximize coincidence?

P9493 Table 3 – According to this table, the visibility has the largest impact of all error sources. This is probably true, but there is no mention of how the numbers (10 – 20 counts etc) were derived, or of what conditions these are representative. Please clarify and explain what is meant by 'good visibility'.

P9495 Figure 1 – Please include a color scale for the lower image (intensity decrease relative to background) and specifically state which threshold was used for coloration. Some of the plume in the lower image does not appear to have a color assigned to it. Perhaps lower the threshold?

P9496 Figure 2 – The length scale is difficult to read because the numbers are very small, but it appears to show that the images have been distorted (compressed in the vertical direction). Though both vertical and horizontal axes appear to be of similar lengths, the vertical axes represents 200m while the horizontal one only represents 100m. Were the images really distorted? If so, why? It doesn't seem necessary.

P9497 Figure 3 – See above comment.

P9503 Figure 9 – It seems a bit odd that an example of a cell measurement by DOAS is shown but this very cell is not included in the plot in Figure 8. The cell appears to contain 200 ppmm of SO2, yet Figure 8 does not have a point at 200 ppmm. Perhaps shown an example from a cell that was actually used for calibration, or include this cell in the calibration curve?

P9504 Figure 10 – Please indicate how much SO2 is in this cell. Is it about 1,000 ppmm?

P9506 Figure 12 – Please increase the font size for the top x axis label. Also, there is a parenthesis missing in the caption.

P9508 Figure 14 – Please increase the font size for the top x axis label. Also, there is a parenthesis missing in the caption.

P9509 Figure 15 – Here, you state the UV image is of the SO2 path concentration. Is this true? If so, please be sure to add a color scale. Also, there is a parenthesis missing in the caption.

P9510 Figure 16 – Please increase the font size for the top x axis label. Also, there is a parenthesis missing in the caption.

Final Remarks

The submitted manuscript represents a feasibility study for applying UV imaging cameras to measure SO2 in sea going ship emissions. The study concludes that there is some promise to the technique, but that a number of issues need to be sorted out before quantitative operational use is possible. The paper is written in a clear and concise manner, and the presented data is fairly easily understood.

The one main issue that I believe is not discussed sufficiently is the problem of using only a single wavelength channel. This makes it impossible to account for scattering and absorption of particulate matter in the ship plumes. However, I hope that the above comments may help the author in finding an adequate approach to dealing with the issue. Only then will it be proven that the measured UV extinction does indeed stem predominantly from SO2 absorption, thus giving the feasibility study value and enabling valid conclusions about the applicability of UV imaging systems to ship emissions. Once this issue has been addressed, I recommend this manuscript be published in Atmospheric Measurement Techniques.

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 9467, 2013.