

Author Comment with regard to:

“Attribution of Atmospheric Sulfur Dioxide over the English Channel to Dimethylsulfide and Changing Ship Emissions”
by M. Yang et al.

14 March, 2016

Many thanks for the thoughtful *comments and suggestions from Anonymous Referee #1*. We are very glad to hear that the referee found our contribution valuable. Below are our replies to the specific comments, which are in *italic*.

Anonymous Referee #1

General comments

This is a well written and novel paper which uses observations from the Point Penlee Atmospheric Observatory to explore the importance of ship and biogenic sources to SO₂ over the English Channel. The measurements reported are a valuable indicator of the effectiveness of Sulfur emission reduction schemes. The scientific questions explored are well within the scope of ACP and I recommend publication after the following minor recommendations are addressed.

Specific comments

Page 2 line 17 suggest ‘source to’ replaced with ‘contributor to’
Suggestion accepted.

Line 20 “ aerosols resulting from ship emissions contribute to tens of thousands of cases of premature mortality” Suggest insert ‘cases of’
Suggestion accepted.

Page 4 line 2 “SO₂ and O₃ were blanked simultaneously”- blanked could be mistaken for capped off. Suggest ‘blank measurements of SO₂ and O₃ were made simultaneously.’
Suggestion accepted.

Line 4 “We checked the calibration of the SO₂ instrument occasionally” – be more specific about frequency – whether monthly, yearly etc
We change the word occasionally to “twice a year.”

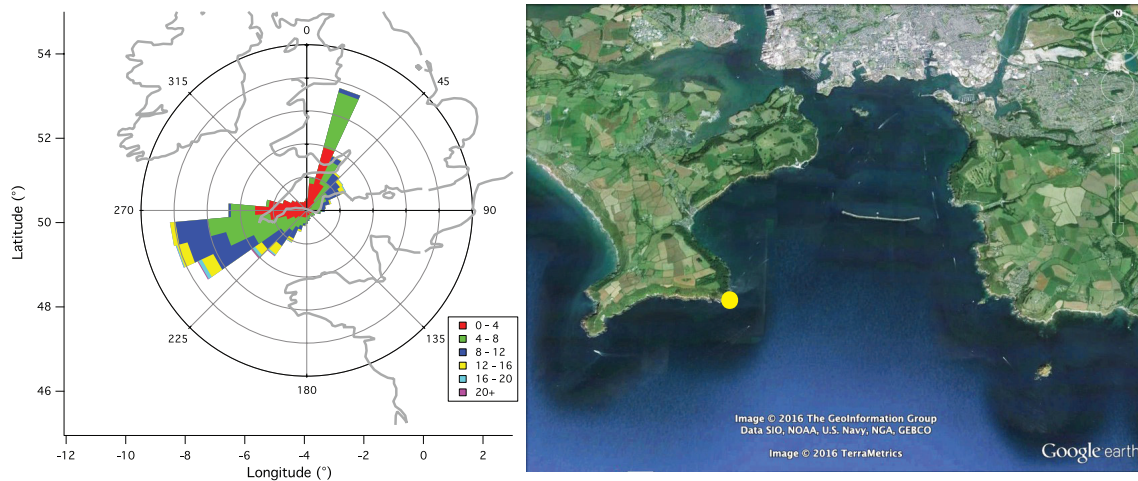
Page 4 line 8. SO₂ measured at a height of 2m and CO₂ at 18m. This is a significant height difference and I wonder might lead to de-coupling of air masses sampled at different heights? (particularly important for FSC calculations). Is there a measure of wind direction, speed at these two heights for comparison?

Thanks for the comment. The ground level of the observatory is about 11 m above mean sea level. SO₂ was measured at ~13 m above mean sea level (2 m above ground) while CO₂ was generally measured at ~18 m above sea level. We did not have simultaneous wind measurements at these two heights. However, a significant difference in wind

direction between 13 and 18 m (both within the surface layer of the atmosphere) is generally not expected.

Page 4 Results A map of the Southern part of UK with the location of the observatory is needed to give perspective to where the observatory sits in relation to Plymouth Sound and the English Channel. This is provided in Yang et al 2016 but would also be useful here. Currently statements such as Page 4 line 24-25 “The wind sector between 110 and 250 deg is completely unobstructed by land” is not obvious looking at Fig 1.

Thanks for the comment. We’ve added the close up map below. The yellow circle indicates the location of the observatory. The Plymouth Sound spans ~4 km east of PPAO.



Page 4 line 22 The wind rose shows 2 distinct dominant wind directions (SW and NNE). Suggest add a sentence describing land use/potential sources to the NNE.

We will add that the NNE sector is likely influenced by emissions from terrestrial anthropogenic sources.

More generally, the wind rose shows there is a low frequency of winds from the SE, with data from this direction an important focus of the paper. Could the authors describe how many hours of data they have used from the SE and SW directions respectively in their analyses?

On average, winds came from our SW sector 7.5 days a month, and came from the SE sector 2.1 days a month.

Line 26 “Winds from between 50 and 110 deg face the eastern side of the Plymouth Sound, which is busy with ship traffic.” Please provide an estimate of the volume of ship traffic for both the English Channel and Plymouth Sound.

According to the Devonport Naval Base Ship Movement Report, the total number of ships in the Plymouth Sound varies from about 4000 per month in winter to 6000 per month in summer. The volume of ship traffic in the English Channel is about 15000 per month (Maritime and Coastguard Agency, 2007).

Page 5, line 5 “The lowest SO2 mixing ratios were observed in the western, terrestrially-

influenced wind sector in both years” It’s not clear from Fig 1 that western direction is terrestrially influenced, hence need for a more regionally-scaled map
See map above.

Page 5 line 13 “A lower, broader peak in SO₂ can also be observed between about 18:30 to 20:00 UTC.” This SO₂ peak seems to correspond with a decrease in O₃, but not an increase in CO₂ which would be expected to be enhanced alongside SO₂. Please comment on this.

Due to the high background mixing ratio in CO₂ (~400 ppm), ship plumes tend to result in much smaller (additional) signal:background ratios in CO₂ than in SO₂. As a result, CO₂ emitted from point sources tends to quickly become indistinguishable from the background with increasing distance (i.e. greater air dilution/dispersion). This is why we only focused on sharp, low plumes in the calculations of the fuel sulfur content.

Line 19 “. . .as well as the busiest part of the shipping lanes.” Could a figure be included which shows the shipping density in the SE versus the SW direction?

We will refer to the ship’s AIS (Automatic Identification System) maps shown by Jalkanen et al ACP 2016.

Page 6, line 14 “Recently-calibrated transmission efficiencies from the manufacturer (Ionicon, Austria) and kinetic reaction rates from Zhao and Zhang (2004) were used to derive the DMS mixing ratio.” As DMS was not specifically calibrated during measurements, please provide an estimate of measurement uncertainty. Please comment on how this uncertainty impacts the diel amplitude of DMS and the calculated mixing ratio of SO₂ from oxidation of DMS.

Without direct calibration, the uncertainty in the atmospheric DMS mixing ratio by the PTR-MS is $\leq 40\%$. A worst case $\sim 40\%$ lower atmospheric DMS would still be able to account for most of the observed SO₂ from the southwest sector.

Page 8 Line 29 In addition to the horizontal distribution of ship plumes, please comment on the likely vertical distribution of ship plumes observed at PPOA, which may be especially important given the different inlet heights of SO₂ and CO₂.

The difference in height between the SO₂ and CO₂ measurements was only 5 m (18-13 m). Eq. 2 from von Glasow et al (2003) predicts that just 10 seconds after emission, a ship plume will have already expanded vertically to a height of 20 m from the surface. It seems highly unlikely for our SO₂ and CO₂ sensors to be sampling different airmasses under typical meteorological conditions in the marine atmosphere.

Page 9 line 6 “A ship 100 km away would have a plume that is observable for nearly an hour”. Suggest add ‘theoretically’ to this sentence, as significant dilution of plume over 100km would make detecting enhancement of SO₂ and CO₂ very difficult?

Agree. Suggestion accepted.

Page 10 line 13 – how does the absolute FSC % from PPOA compare with recent estimates by Kattner and Beecken 2015?

The mean FSC observed at PPAO (~0.17%) was about half of what was observed by Kattner et al (2015) for the year 2014. In 2015, mean FSC at PPAO (~0.047%) was fairly comparable to observations from Kattner et al (2015). Beecken et al. (2015) observed a bimodal distribution in FSC for year 2011 and 2012; the lower mode centered around ~0.25% and the higher mode centered around ~0.9%.

Page 10 line 18 – please comment on how different inlet heights of SO₂ and CO₂ may add to uncertainty in estimating FSC

We don't think the small height difference (5 m) significantly contributes to the uncertainty in FSC. However, using the same gas inlet for the SO₂ and CO₂ instruments and recording data from both on the same PC would help to better synchronize the two gas measurements.

*Line 22 “Long-term records of another tracer (e.g. nitrogen oxides or particle number)”
Suggest that black carbon would be a better indicator for ship exhaust than particle number, as particle number may be enhanced by local biogenically driven events.
Thanks for the suggestion.*

Typing/technical errors Page 10, Line 20 A higher SO₂' remove apostrophe after SO₂
This is not an error. The apostrophe indicates deviation in SO₂ mixing ratio from the background. See line 17 on p. 9.