

As a general note, there are a large number of changes in the revised manuscript which are **not** in response to the reviewer comments but rather seem to be 'proof reading' changes from the authors. In contrast, many of the reviewers comments were only addressed in the reviews and not in the article. The changes made without reference to reviewer comments were substantial and in some cases change information the reviewers referred to. Therefore, I have decided to send to revised article back to the reviewers to be sure that all their comments are addressed adequately and that they accept the changes made.

In future, the authors should ensure that they conduct proof reading **before** submission of the article. All reviewer comments should result in a change in the manuscript and not just a clarification in the author response – unless there is a clearly stated disagreement with the reviewer comment. Even if the reviewer has simply misunderstood or missed a point, the article should be made more clear so that readers do not face the same problem.

Response to comments from Reviewer 1:

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.

- Please add this information to the revised manuscript.

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).

- Please add this information to the revised manuscript.

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.

- Please add this information to the revised manuscript.

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.

- Please add this information to the revised manuscript. This reviewer asks several times about the different inlet heights: please add more information to the paper to address the multiple concerns of the reviewer and anticipate similar questions in readers.

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 ($\sim 0.17\%$) was about half of what was observed by Kattner et al (2015) for the year 2014. In 2015, mean FSC at PPAO ($\sim 0.047\%$) 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 $\sim 0.25\%$ and the higher mode centered around $\sim 0.9\%$.

- Please add this information to the revised manuscript.

Response to comments from Reviewer 2:

3. p5, line 20: in Figure 4, the sector with few mixing ratios over 0.5 ppb in 2015 is southwest, not southeast.

There were indeed few occurrences > 0.5 ppb in 2015 for the southwest sector. Though here we were making the point that the distribution shifted towards lower SO₂ mixing ratios in the southeast.

- Please clarify this point in the revised article.

5. p7, line 9: Liss and Slater (1974) state that both the hydration and the subsequent oxidation of SO₂ are rapid, but they gave no reference to the oxidation kinetics. It may well be that rapid hydration is enough to justify the assumption of near zero concentration of dissolved SO₂. This point should be discussed more fully. Rate constants for the kinetics of sulphite oxidation can be found in Zhang and Millero (Geochim. Cosmochim. Acta, 57, 1705-1718, 1993).

Thanks for the comment. As summarized by Schwartz (1992), SO₂ dissociates almost instantaneously to form HSO₃⁻ and then sulfite in seawater. The effective solubility of SO₂ in seawater (pH ~8) due to this chemical enhancement is very large (dimensionless water:air solubility of about 5e8), which means that air-sea SO₂ exchange should be gas phase controlled. Oxidation to sulfate permanently removes sulfite from the ocean with a time scale of minutes to hours. This results in low sulfite concentration in the surface ocean (e.g. a few μM or less, Campanella et al 1995; Hayes et al 2006). Even a large dissolved SO₂ concentration of 10 μM would only equate to an equilibrium atmospheric SO₂ concentration of 2e-5 nmole/L of air (equivalent to 0.5 ppt, 2-3 orders of magnitude lower than typical atmospheric mixing ratio). From these calculations, we see that the air-sea concentration gradient in SO₂ resides essentially all in the atmosphere.

- Clarify this response in detail in the revised article.

6. p11, line 1: It is not clear what reductions are being discussed, in particular how does a reduction in 2014 arise?

Sorry for the confusion. "Reduction" here refers to the difference in the computed mean SO₂ mixing ratio caused by excluding the very coastal ship plumes.

- Please clarify this in the revised article.

Additional comments:

- P1 L29: Change oxidations back to oxidation