Response to Referee 1

We would like to thank the referee for their kind comments on the clarity and readability of the paper and also for their feedback. We note the referees comment regarding the need to motivate the use of LIF to detect SO_2 in the introduction and address that in our response to the comment about line 55.

Line 13: add a word after "(PF)" to indicate that this is not a form of mass spectrometry. Perhaps "instrument"?

We have added the word "analyser" after PF (unless referring to PF data, PF signal, PF technique etc.) and the word "instrument" after LIF, for clarity.

Line 17: ppb is used where as elsewhere "pptv" is used. Be consistent please.

We have amended every instance of "pptv" or "ppbv" to "ppt" and "ppb" respectively, in line with the standard convention in this field.

Line 55: I think at the end of this paragraph a short new paragraph would be helpful outlining the properties of SO_2 that make it suitable for detection – it's cross section, stability etc. Here it would be helpful to explain how UV remote sensing instruments are not suitable for estimating remote SO_2 abundances due to their low concentrations, for example, hence motivating the focus of this work. This comment relates to the overall statement: "I found that it lacked a bit in the introduction to motivate what it is about SO_2 that lends LIF as being such a useful approach to its detection".

We have added the following text in the specified location:

"SO_2 absorbs strongly in the ultraviolet region of the electromagnetic spectrum, characterised by a series of absorption bands at wavelengths from 403 to 106 nm (Manatt et al., 1993, Stark et al., 1999; Rufus et al., 2003). Therefore, spectroscopic SO_2 detection techniques typically exploit this UV region.

Fluorescence-based techniques employ excitation in the $\tilde{C}$ band (~190-230 nm), which offers the highest absorption cross sections and fluorescence quantum yields, thus providing the most sensitive detection. Since other atmospheric species also exhibit strong absorption features in this UV region (e.g. the Hartley band of O_3 and the γ system of NO), techniques using broadband excitation sources (e.g. the lamp in the PF analyser used in this work) are susceptible to spectral interferences, thereby reducing their sensitivity and selectivity to SO_2. These interferences can be somewhat overcome by the use of bandpass filters to control the excitation wavelength, even more powerful however is the ability to resolve spectral features of SO_2 by the use of a narrow band light source such as a laser. The combination of such a light source with the large absorption cross section of SO_2 at these wavelengths make LIF a prime candidate for achieving SO_2 detection with both very high sensitivity and specificity.

Remote sensing techniques (e.g. differential optical absorption spectroscopy (DOAS) and UV camera spectral imaging) primarily target the weaker $\tilde{B}$ band (~300-320 nm), which enables the utilisation of commercial UV cameras and detectors and coincides with less interference from other species but also corresponds to smaller SO_2 absorption cross sections than the $\tilde{C}$ band. Such remote detection techniques are, in general, not sufficiently sensitive to detect the low levels of SO_2 seen in clean marine air due to the smaller absorption cross sections at 300-320 nm and because they rely on absorption rather fluorescence detection."

Line 90: Reference Figure S4.

We have added a reference to Figure S4 on line 122 instead, as we deemed this a more appropriate position.

Line 183: "was" should be "were".

We thank the referee for this comment and have amended the text accordingly.

Figure 10: Personally, I think it would be neater to have the gradient and R^2 presented inside the plot like in Figure 8. Also, why is there no uncertainty on the CIMS-SO_2:CO_2 gradient?

We have added the gradient and R^2 values inside the plot for Fig. 10 and have also amended the similar regression plots in the SI. The lack of an uncertainty for the CIMS-SO_2:CO_2 gradient was primarily due to difficulties in accurately estimating an uncertainty for the CIMS. Based on the reviewers comments we realise that this is a significant omission from the work and have therefore estimated a CIMS uncertainty of 102 % based mainly on the fitting of the SO_2 peak. However, due to a large difference in instrument uncertainty between the CIMS SO_2 and CO_2 data, this effectively down-weights the CIMS data to the point that the fitted slope becomes poorly constrained and biased toward zero. Therefore, an uncertainty on the CIMS-SO_2:CO_2 gradient has been estimated from an ordinary least squares (OLS) fit and added to Fig. 10. For consistency, the regression plots in the SI have also been fitted via the OLS method.

Figure 11: ppb used in y axis label whereas ppbv mentioned in the text.

We have amended the text accordingly.

Line 404: Reference for the ACSIS project would be helpful – and possibly a Figure in the supplement of where it was flying.

We have added a reference to the "Data supporting the ACSIS programme" publication by Archibald et al. (2025) on line 437, which includes a flight track plot of ACSIS-7.