Response to Referee 2

We thank the referee for their positive remarks regarding this work particularly the characterisation of the LIF instrument and measurement protocols. We also feel that their detailed comments, particularly regarding the altitude plots, have enabled us to improve the manuscript, for which we are very grateful. Our responses to the referee's specific comments are as follows:

Line 174: "and to capture instrument drift": Did the authors notice any consistent drift for the LIF instrument over the course of a given flight? If so, what is the possible cause?

We have added the following text to explain the cause of the LIF instrumental drift:

"The observed drift resulted from variations in laser linewidth caused by external temperature fluctuations within the aircraft cabin, which directly impacts instrument sensitivity."

Line 181: "As a result of inconsistencies in the laser linewidth": How was this determined that the laser linewidth was the cause of variations in the sensitivity as opposed to errors in the calibration system, etc.? There is a lot of overlap in the error bars in Figs S1-S3, so it doesn't appear that the sensitivities are statistically different.

We determined the laser linewidth to be the cause of variations in sensitivity due to corresponding reference cell signal changes. We agree that the sensitivities are not statistically different and so have replaced the text "As a result of inconsistencies in the laser linewidth, the sensitivities were seen to vary slightly during the course of a flight" with:

"As the sensitivities during the course of a flight largely agree within errors"

Line 242: "larger interfering peak": What are some possible compounds that would correspond to the interfering peak in Figure 4(a) at m/z 190.899311? After seeing that figure, it is understandable why obtaining SO_2 from that m/z is so challenging.

We have added the following text to suggest a possible compound that would correspond to the interfering peak in Figure 4(a) at m/z 190.899311.

"likely an isotope of nitric acid"

Figure 8: I assume these are York regression fits being reported? Also, could the authors report the error on the slope and intercept for the fits (particularly Fig 8a)?

York regression was not used in Fig. 8 as we were unable to accurately quantify the large uncertainty on the CIMS observations. As the CIMS measurement uncertainty is significantly larger than the LIF, for the reasons explained in the paper, we felt a standard OLS was sufficient. In responding to the reviewers comments we have estimated a CIMS uncertainty of 102 % and attempted a York regression for this plot. However, due to the large difference in instrument uncertainty associated with the I^-CIMS cps data compared with the LIF, this effectively down-weights the CIMS data to the point that the fitted slope becomes poorly constrained and biased toward zero. As the primary aim of these plots is to a) obtain a suitable gradient linking the CIMS cps data to the LIF mixing ratios for estimating the CIMS mixing ratios, and b) compare how well the PF and LIF mixing ratios agree, we feel a simple ordinary least squares (OLS) regression method is adequate and has been used for both plots for consistency. We have stated the fit technique in the caption of Fig. 8 and have added the errors on the slopes and intercepts from the OLS fits, as requested.

Lines 401-404: A comparison is made between SO_2 LIF data between the ACRUISE and ACSIS-7 campaigns, but no further analysis is done other than just showing the vertical profile of SO_2 in Fig S9. The authors should either develop this analysis further or consider cutting ACSIS-7 and Fig S9 from the manuscript.

We thank the referee for this comment and appreciate their perspective. While an in-depth analysis of SO_2 vertical profiles is outside the scope of this paper, we believe that including these ACSIS-7 data provides a valuable contribution to demonstrating the capabilities of the LIF instrument and for presenting rare observations of SO_2 vertical structure in the remote marine environment. We have therefore retained the reference to ACSIS-7 and Fig. S9 (as supported by Referee 1) and have added the following text comparing our observations with the only other reported vertical SO_2 data over a similar region in the North Atlantic, to our knowledge. However, if the editor agrees we should remove this material from the paper we will.

“It is evident that the marine environment sampled during ACSIS-7 was cleaner at the sea surface, likely reflecting the reduced influence of ship emissions compared to ACRUISE-3. SO_2 mixing ratios, however, become increasingly comparable to those measured during ACRUISE-3 at higher altitudes and display a similar decreasing trend with altitude up to 2000 m due to vertical mixing and diffusion from marine SO_2 sources. The ACSIS-7 profile exhibits a notable mixing ratio inversion at 2000 m, coincident with the marine boundary layer height.

While reported observations of SO_2 altitude profiles over the ocean are scarce, relevant comparisons can be drawn from measurements in the Atlantic between 30N – 54N during the ATom campaigns (Bian et al., 2024). Considering differences in sampling strategies, time of year and location etc., the ATom profiles are consistent with our observations, showing SO_2

mixing ratios of a similar order of magnitude, albeit slightly lower, and a comparable decreasing trend with altitude (up to 3.8 km).”

Figure S10: Why is this figure included if not mentioned in the main text?

We thank the referee for this comment and have removed Figure S10 from the SI.

Figure 4 was hard to read since it appeared more pixelated than other figures.

We thank the referee for this comment and have improved the resolution of Figure 4.