Supplementary Information (SI)

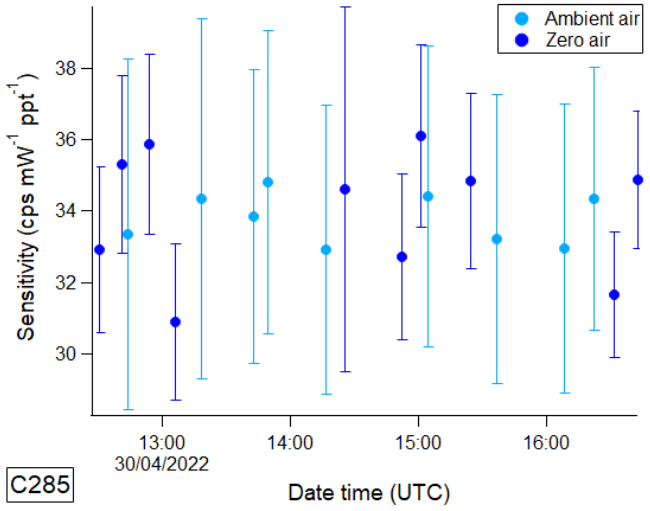

Figure S1. LIF sensitivity variation over time during flight C285 for calibrations in ambient air and zero air. Error bars are given to a 2σ confidence interval.

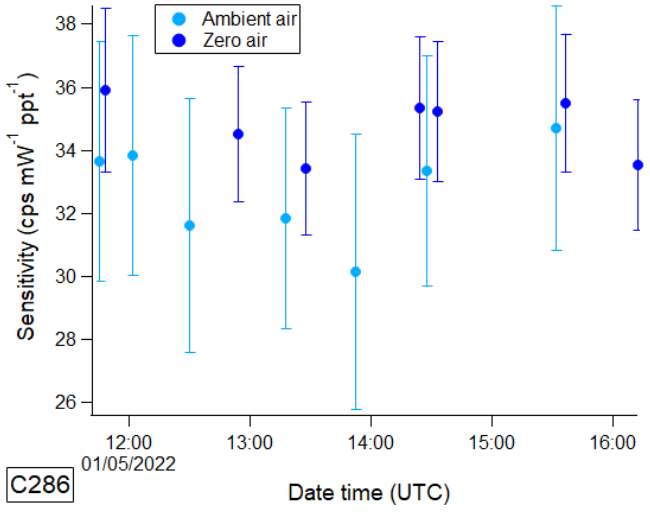

Figure S2. LIF sensitivity variation over time during flight C286 for calibrations in ambient air and zero air. Error bars are given to a 2σ confidence interval.

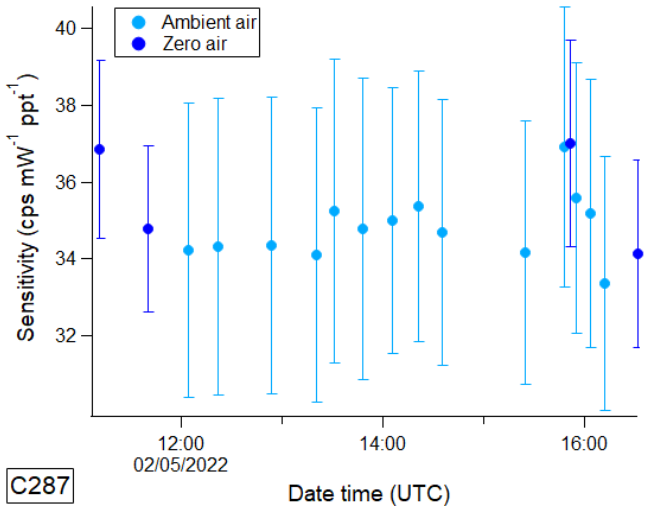

Figure S3. LIF sensitivity variation over time during flight C287 for calibrations in ambient air and zero air. Error bars are given to a 2σ confidence interval.

15

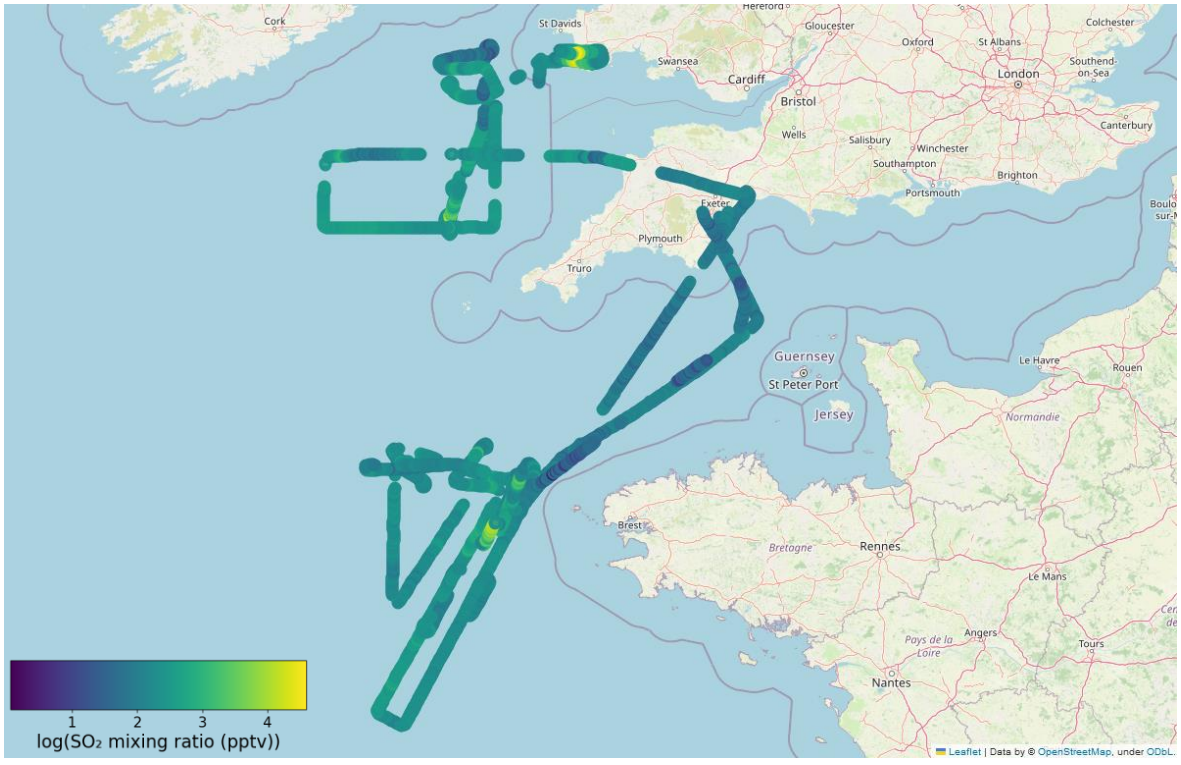

Figure S4. Colour map of 10 s LIF SO_2 mixing ratios along all three ACRUISE-3 flight tracks.

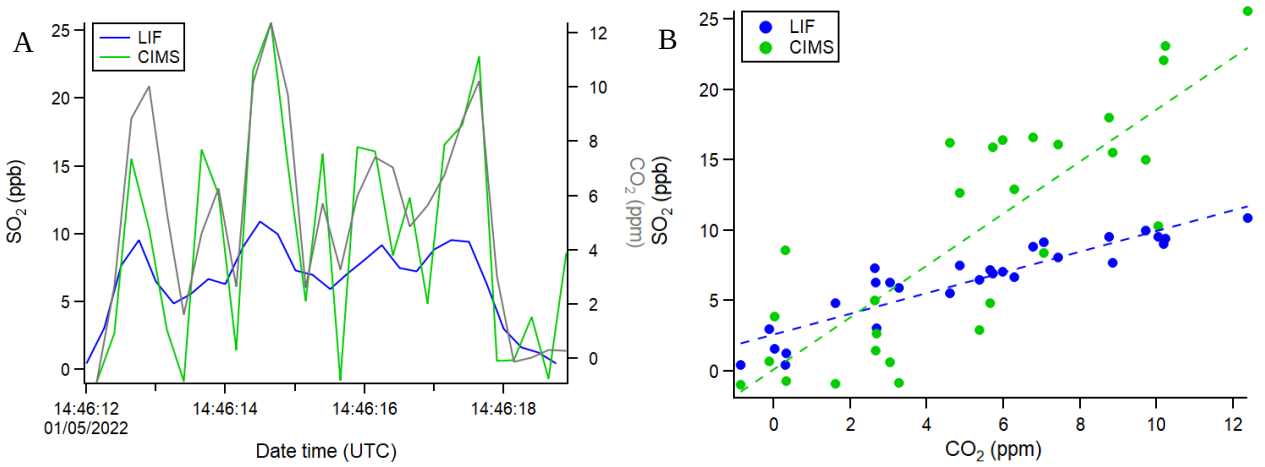

Figure S5. SO_2 and CO_2 mixing ratios of a ship plume during flight C286 showing the data used to calculate an emission ratio via the A) integration method (area under the plume) and B) regression method. The linear fits have gradients and R^2 values of 0.74 ± 0.13 ppb ppm^{-1} and 0.83 (LIF); 1.85 ppb ppm^{-1} and 0.69 (CIMS).

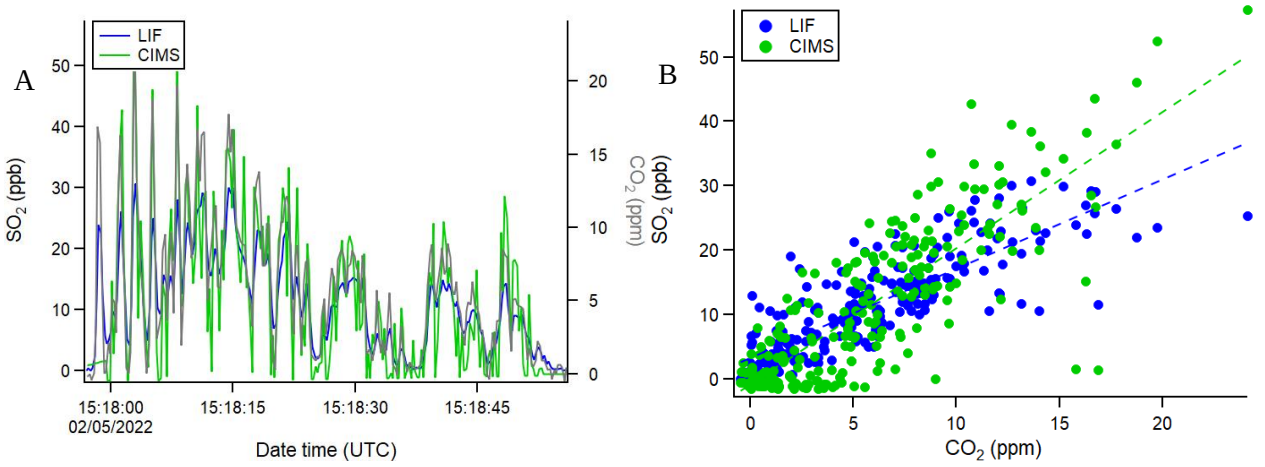

Figure S6. SO_2 and CO_2 mixing ratios of the first ship plume during flight C287 showing the data used to calculate an emission ratio via the A) integration method (area under the plume) and B) regression method. The linear fits have gradients and R^2 values of 1.38 ± 0.12 ppb ppm^{-1} and 0.70 (LIF); 2.11 ppb ppm^{-1} and 0.68 (CIMS).

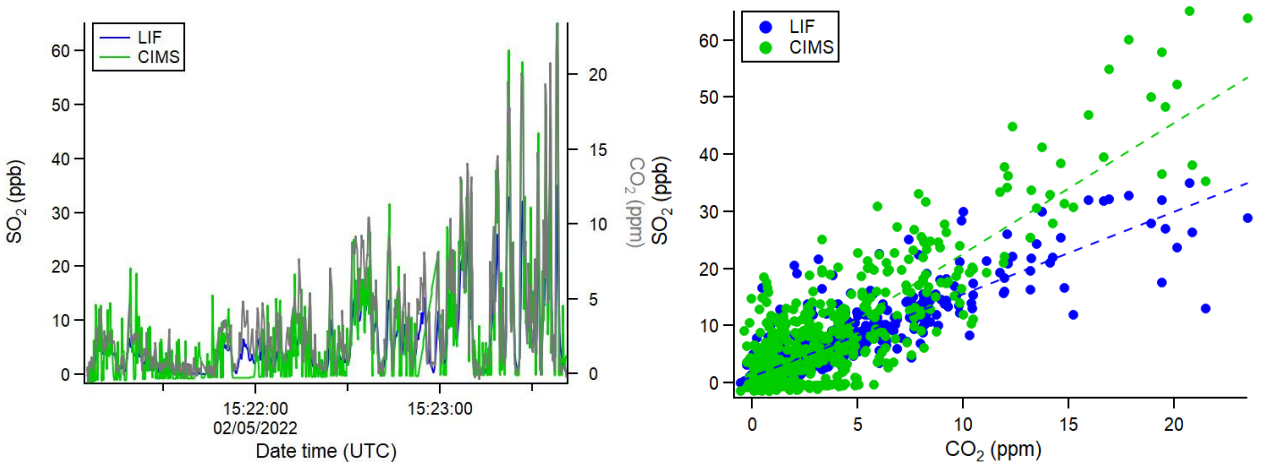

Figure S7. SO_2 and CO_2 mixing ratios of the second ship plume during flight C287 showing the data used to calculate an emission ratio via the A) integration method (area under the plume) and B) regression method. The linear fits have gradients and R^2 values of 1.44 ± 0.07 ppb ppm^{-1} and 0.76 (LIF); 2.29 ppb ppm^{-1} and 0.72 (CIMS).

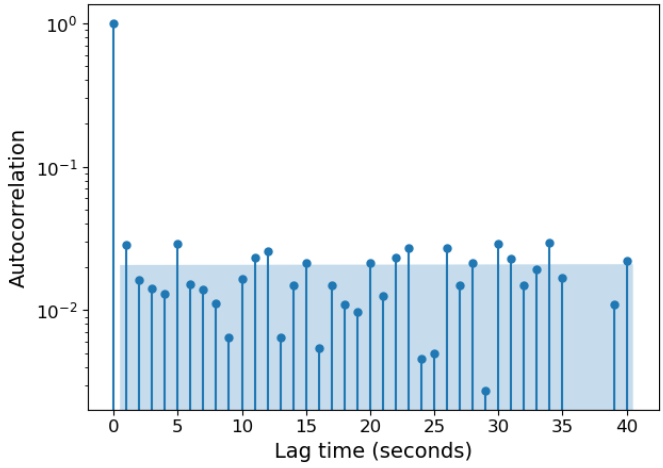

Figure S8. Autocorrelation plot of the LIF data presented in Fig. 11 at 10 Hz.

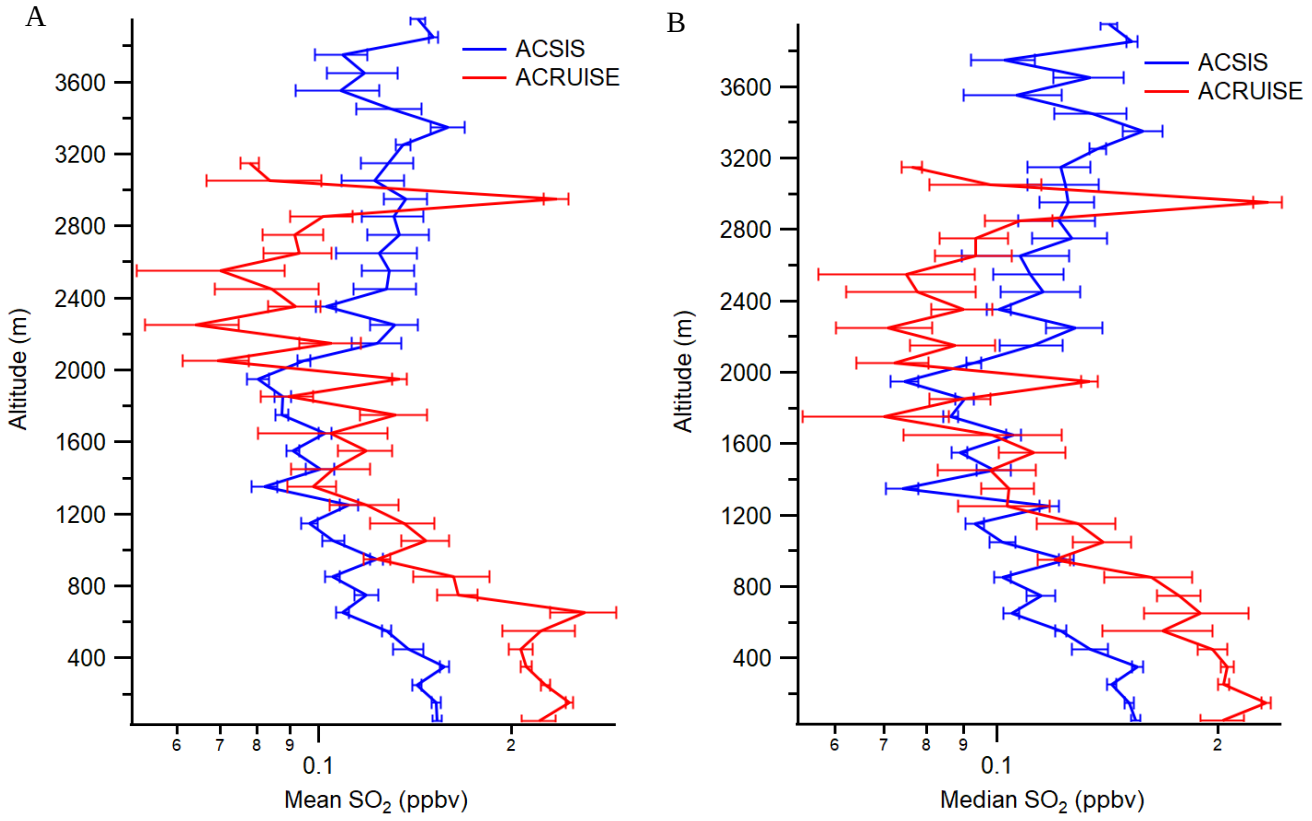

Figure S9. Profiles of mean (A) and median (B) 10 s LIF SO_2 mixing ratios for each 100 m altitude bin, comparing the ACRUISE-3 and ACSIS-7 data. The error bars indicate 2 standard errors.

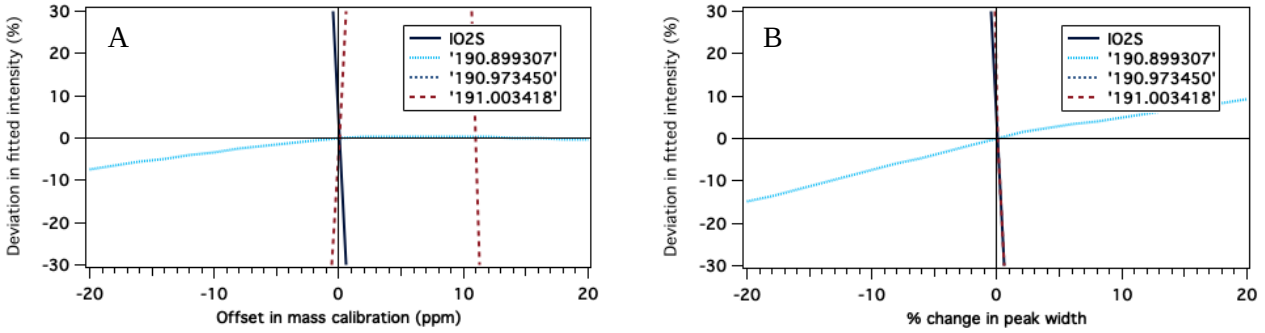

Figure S10. Changes in the intensity of the peaks identified at m/z 191 as a result of A) offset in the mass calibration and B) changes in the peak width.