

Blue: Referee comment

Black: Our comment

Red: The sentences in our manuscript

Overall evaluation and general comments:

The preprint by Kazuki Kamezaki on a “*compact continuous measurement system for atmospheric carbonyl sulfide*” describes and evaluates modifications to improve a commercially available OCS instrument. In that respect, the term “*Development of...*” in the title is clearly an overstatement. While this could be amended by changing the title, I ’m afraid that the paper in its present form contains too little substance and too many errors and ambiguities to be publishable in AMT. The necessary modifications go beyond normal revisions, which is why I recommend not to accept this paper for publication. Below, I explain my main concerns. Then I list some specific and technical issues to underline my criticism and to provide recommendations should the authors chose to rewrite the article for resubmission at a later stage.

Reply: Thank you for your comment. We have removed the “development of” from the title. We apologise for the poor quality of our preprints. We have considered your comments seriously and revised the manuscript accordingly. Thank you for your comments.

The methods section (Section 2) falls short of providing anything close to a comprehensive instrument description. Neither is the overall measurement and data analysis concept of the original MIRO analyzer fully described, nor are shortcomings or problems that motivated the adaptations made to the analyzer in this work properly characterized. In a methodological study such as this one, I expect, for example, the following details to be given and explained: What are the dimensions of the optical cell and what is the absorption path length? What light source and detector are being used? At what wavelength is OCS being measured? Does the analyzer measure an absorption spectrum that is analyzed by spectral fitting, or does it measure absorption only at one or more distinct wavelengths? Which water absorption band is used as a reference peak, and how does this referencing work? How is the temperature of the light source stabilized, and how is wavelength and baseline stability ensured? What are the main sources of noise? Without such methodological details, it is impossible to put the applied modifications into context and evaluate their benefits as well as the overall performance measures.

Reply: Thank you for your comment. The manufacturers were contacted for further information and provided as much as they could about the MIRA Pico without revealing some of their trade secrets, according to them. We have added the following information to Section 2.1: **COS concentrations were measured using a MIRA Pico analyser (Figure S1a). The MIRA Pico analyser is a portable, field-deployable instrument designed for the simultaneous measurement of**

COS, CO₂, and water vapour concentrations with one Hz. The MIRA Pico analyser is ultracompact (30 cm × 20 cm × 10 cm), and lightweight (3 kg), uses a small cell (60 mL), and has an optical path length of 13 m. The upstream filter is a 0.01-micron fluorocarbon borosilicate glass microfiber element, and the tubing upstream from the optical cell is made of polyurethane tubing. The flow rate was adjusted using the pump voltage, which was set at approximately 210 mL min⁻¹. The pressure inside the optical cell was 140 mbar. The analyser incorporates a mid-infrared tuneable diode laser and a HgCdTe (MCT) photovoltaic detector for signal acquisition. The optical cell package contained an internal thermoelectric cooler (TEC) that controlled the temperature of the laser diode (Figure S1b). The laser TEC output (the laser temperature) was fed into a proportional-integral-derivative (PID) loop to maintain the water line absorption peak in place. The determination of CO₂ and COS spectra by the MIRA Pico analyser relies on referring to the position of the water spectral peak in the ~4 μm region and calculates each concentration by spectral fitting individually. Consequently, the water vapour concentration (>1000 ppm) in the air is mandatory for COS measurements, making it difficult to measure COS concentrations in low-temperature or high-altitude regions without humidification. Specifically, it is difficult to use MIRA Pico when the temperature is below -20 °C and the relative humidity is below 8% at 10 °C. The COS concentration was output from the MIRA Pico analyser in the dry mole fraction with a 1-point linear correction determined by the manufacture. Equipped with two inlets, the analyser enabled switching back and forth between the unaltered air and COS-scrubbed samples, mitigating temperature-induced drifts, and the guaranteed number of switches was 100 million. The main sources of measurement noise are due to electrical equipment, the quality of optical coatings, and purity of the laser beam. However, under the original settings, this system exhibits a significant signal drift (see section 3.1), with a standard deviation (1σ) of ± 50 ppt on a 10 min time scale, and its accuracy is also sensitive to ambient temperature. In addition, introducing high water vapour concentrations into the MIRA Pico analyser may cause condensation in the cell, making measurements impossible.

With respect to performance measures (given in Section 3) some of the numbers are neither well rationalized nor convincing. The terms accuracy, precision, and uncertainty are not always used in a correct and consistent manner, and it is not at all clear how the results of the various measurement series performed on known standards are used to validate or correct COS concentrations measured in the field.

Reply: We apologise for not providing sufficient explanation. We have added the experimental methods to the Methods section of Section 2.4.2. We have also added the results of our tests on the stability of the reference gas in Section 3.2, as we thought you were probably concerned that the reference gas was not stable. Sections 2 and 3 describe each analysis and its use.

The description of the field measurements in Section 4 is not very meaningful in my opinion. The explanations and interpretations are largely rudimentary and speculative. They go too far for a purely technical paper, but not far enough in order to be of scientific value. Given the time period reported and the region covered, the results are only of (rather limited) local interest, and the case that more such measurements could be a game- changer in understanding the OCS budget and cycling is not convincing.

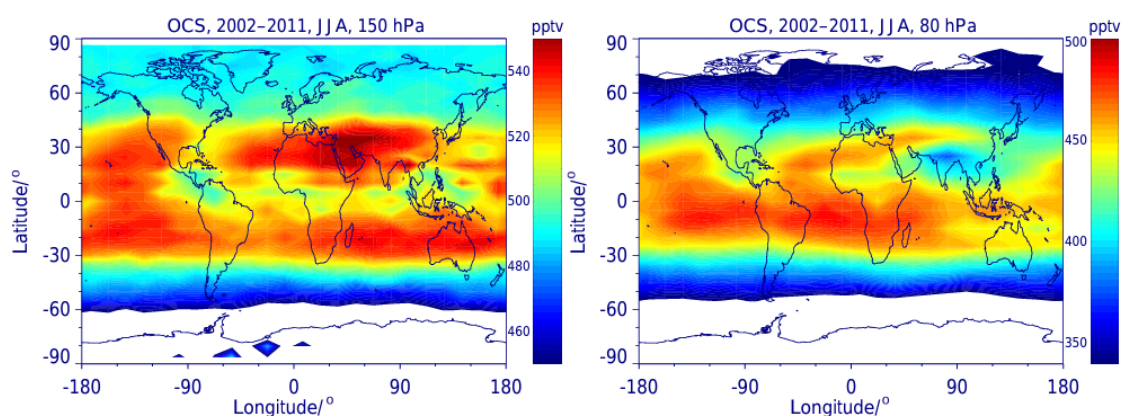
Reply: The conclusion has been overstated. We have removed the results regarding the COS concentrations observed in cars. However, we have discussed the results of 10 days of observation at the Tsukuba site based on meteorological data. Because we wanted to show that reasonable COS concentrations could be observed with the MIRA Pico system in this field observation, we changed our conclusions as follows: **The MIRA Pico system was used to perform atmospheric observations and provide reasonable results, indicating that this analytical method can be used to observe variations in atmospheric COS concentrations.**

Specific comments:

Line 27 – 30: In my opinion, these two sentences are misleading. First, there are no “*local contributions to SSA production*”, which happens in the stratosphere and is not directly connected to the near-surface OCS cycling. What you mean is contributions to the budget, which in turn plays a role for how much OCS reaches the stratosphere, where SSA is produced. More importantly, the statement that “*tropospheric OCS sources and sinks entail great uncertainty due to the limited number COS observation sites*” is far too simplistic. Arguably, a few more sites with OCS observations in well chosen locations could help to better quantify the regional distribution of certain sources and sinks. But overall, a great deal of understanding tropospheric OCS cycling has been achieved with data from available networks and satellites, and I expect a bit more in terms of strategy to address remaining uncertainties than simply calling for more OCS observations.

Reply: Thank you for your comment. The concentration of COS in the stratosphere is not uniform, as reported by Glatthor et al. (2017). The reason for this nonuniformity is thought to be the contribution of local COS emissions. Therefore, it is important to understand the distribution of local COS concentrations. As you have pointed out, increasing the number of observation points is too simple. To accurately understand the dynamics of COS, it is essential to observe ground-level concentrations of COS. However, long-term ground-based observations of COS concentrations are limited to the Americas, Europe, and Antarctica, and the long-term observation sites are biased. Models and satellite observation results cannot capture short-term fluctuations, and ground observations are considered essential for clarifying the causes of COS fluctuations

occurring at the site. Thus, we have changed the sentences in lines 29-34 as follows: COS concentrations have been monitored over the long term by National Oceanic and Atmospheric Administration (NOAA) (Montzka et al., 2007). The dynamics of the global COS are beginning to be understood through satellite observations and models (Kuai et al., 2015; Glatthor et al., 2017; Ma et al., 2023; von Hobe et al., 2023). However, the observation sites are highly biased, and there is no continuous observation data exist for Asia, South America, Africa, and the Atlantic Ocean. Observing COS at locations with no such measurement points and analysing COS variation factors results in a more accurate understanding of the COS budget. We added new references to the revised manuscript.



(From Figure 12 in Glatthor et al. (2017))

References

Glatthor, N., Höpfner, M., Leyser, A., Stiller, G. P., von Clarmann, T., Grabowski, U., Kellmann, S., Linden, A., Sinnhuber, B.-M., Krysztofiak, G., and Walker, K. A.: Global carbonyl sulfide (OCS) measured by MIPAS/Envisat during 2002–2012, *Atmos. Chem. Phys.*, 17, 2631–2652, <https://doi.org/10.5194/acp-17-2631-2017>, 2017.

Kuai, L., Worden, J. R., Campbell, J. E., Kulawik, S. S., Li, K.-F., Lee, M., Weidner, R. J., Montzka, S. A., Moore, F. L., Berry, J. A., Baker, I., Scott Denning, A., Bian, H., Bowman, K. W. Liu, J., and Yung, Y. L. :Estimate of carbonyl sulfide tropical oceanic surface fluxes using Aura Tropospheric Emission Spectrometer observations, *J. Geophys. Res. Atmos.*, 120, 11,012–11,023, doi:10.1002/2015JD023493, 2015.

Ma, J., Remaud, M., Peylin, P., Patra, P., Niwa, Y., Rodenbeck, C., Cartwright, M., Harrison, J. J., Chipperfield, M. P., Pope, R. J., Wilson, C., Belviso, S., Montzka, S. A., Vimont, I., Moore, F.,

Atlas, E. L., Schwartz, E., and Krol M. C.: Intercomparison of atmospheric carbonyl sulfide (TransCom-COS): 2. Evaluation of optimized fluxes using ground-based and aircraft observations. *Journal of Geophysical Research: Atmospheres*, 128, e2023JD039198. <https://doi.org/10.1029/2023JD039198>, 2023.

von Hobe, M., Taraborrelli, D., Alber, S., Bohn, B., Dorn, H.-P., Fuchs, H., Li, Y., Qiu, C., Rohrer, F., Sommariva, R., Stroh, F., Tan, Z., Wedel, S., and Novelli, A.: Measurement report: Carbonyl sulfide production during dimethyl sulfide oxidation in the atmospheric simulation chamber SAPHIR, *Atmos. Chem. Phys.*, 23, 10609–10623, <https://doi.org/10.5194/acp-23-10609-2023>, 2023.

Lines 39 + 47: The statement “*However, these devices are large, expensive, and costly to maintain.*” is too unspecific as these adjectives are somewhat relative. Also, a few lines below, you define a “*good precision*” out of the blue. I would like to see more compelling arguments what the precision needs to be to tackle relevant science questions, and what the exact problems are with the size and costs of existing analyzers. In other words, where are the real problems that need to be solved? Later, it is stated that the MIRO is “*less than half the price*” of other analyzers and “*small*”. Again, please be more specific here (at least make some reference to the details given in Section 2) and clearly state why the differences matter.

Reply: We apologise for the lack of specific information. We have rewritten the sentence in line 44 as follows: *However, the problem is that these devices are large and consume much electricity. The original Aerodyne COS analyser uses 250 W (without a pump), and the original LGR COS analyser uses 400 W. These must be installed at locations where commercial power is available. However, when observing remote locations (such as forest sites in Southeast Asia and remote islands), the power supply may not always be sufficient.*

In addition, we have added the following in line 56: *Furthermore, the MIRA Pico analyser consumes only 15 W, making it suitable for use in remote areas where the power supply is insufficient.*

Line 58 – 59: *If > 5000 ppm of water vapour are needed for the OCS measurement to work, then the instrument will be useless in cold or high-altitude environments (unless you go through the not simple efforts to moisten the sampled air). This should be clearly stated.*

Reply: We have rewritten the sentence in line 76 as follows: *Consequently, the water vapour concentration (>1000 ppm) in the air is mandatory for COS measurements, making it difficult to measure COS concentrations in low-temperature or high-altitude regions without humidification. Specifically, it is difficult to use MIRA Pico when the temperature is below -20 °C, and the relative humidity is below 8% at 10 °C.*

By asking the manufacturer, they answered that they could confirm a water peak up to approximately 1000 ppm. Thus, we changed the value from 5000 to 1000 ppm.

Line 66 – 69: From the text and Figure 1, I find it difficult to understand how the temperature stabilization was really done. Can you give details on the “*commercial refrigerator*”? And was the entire MIRA instrument put into the refrigerator or just the optical cell? I also don’t understand why you would set the refrigerator to 15 °C and the Peltier cooler to 29 °C? What is the target temperature inside the optical cell, and is temperature monitored inside the cell or just inside the refrigerator or at the Peltier cooler? And what is the “*cushioning material*” and what is its purpose? If it is thermal insulation, then “*cushioning*” is the wrong word.

Reply: We have added photos to clarify temperature stabilization and the actual temperature changes to Figure S1. We have also changed the “cushioning” to an aerogel sheet. We have added information regarding the refrigerator and cushioning material in line 93. **Inside the refrigerator, the optical cell package was surrounded by thermal insulation, which is an aerogel sheet on top of blue Styrofoam (Figure S1c), and the temperature was controlled using a Peltier cooler set at 29 °C placed at the bottom (Figure 1). The refrigerator releases heat from inside to outside, and the Peltier cooler under the cell regulates this heat. Double insulation with refrigerators and insulation minimises temperature fluctuations, and an aerogel sheet is interposed between the Peltier cooler and optical cell to further reduce temperature variations.**

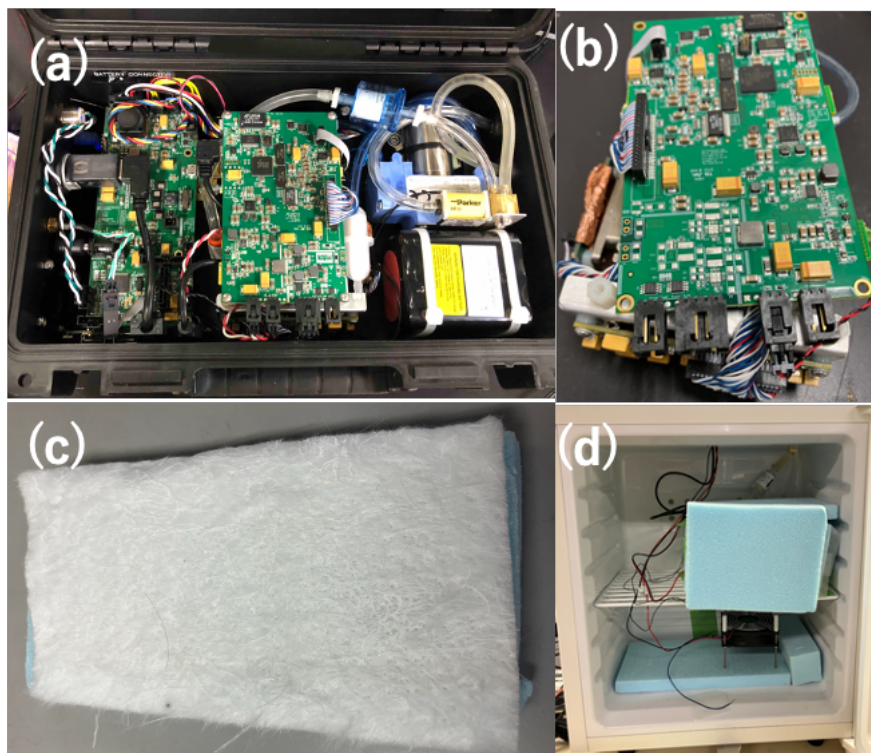


Figure S1. (a) MIRA Pico analyser, (b) optical cell package, (c) thermal insulation, and (d) MIRA Pico system in the refrigerator. The optional cell package consists of the optical cell and the upper and front base. Thermal insulation is an aerogel sheet on top of blue Styrofoam. The optical cell package is covered with insulation. The fan columns are stuck onto the blue Styrofoam.

Section 2.2: There are several things that I don't understand in this section and that need further explanation and rationale: What is the purpose of the ECU and cooling the sample to 2 °C? How do you know/ensure that the fraction of OCS removed by the activated charcoal is constant? With the Nafion dryer, how do you manage to keep the > 5000 ppm water to make the MIRO work (cf. Section 2.1)?

Reply: ECU was installed to prevent moisture from condensing inside the cells. We have added the following sentence in line 103: **The ECU prevents water from condensing inside the cell.**

To check the fraction of COS removed by the activated charcoal, we conducted an additional experiment in Sections 2.4.2 and 3.2.

We did not use a Nafion dryer to remove water but used it to make similar water vapour concentrations of the reference and sample. We have added these sentences in line 121. **The Nafion dryer facilitates the transfer of water molecules from a gas with a high moisture content to a gas with low moisture content via a Nafion membrane. Dry air is commonly used in counter current flows to remove water from samples. However, using a parallel flow, the water contents of the reference and sample gases were maintained at similar levels. Throughout the measurement period, water vapour concentration remained constant. Therefore, no additional measures were necessary to maintain a constant water vapour level. However, depending on water vapour variations, adjustments to the ECU temperature settings may be necessary.**

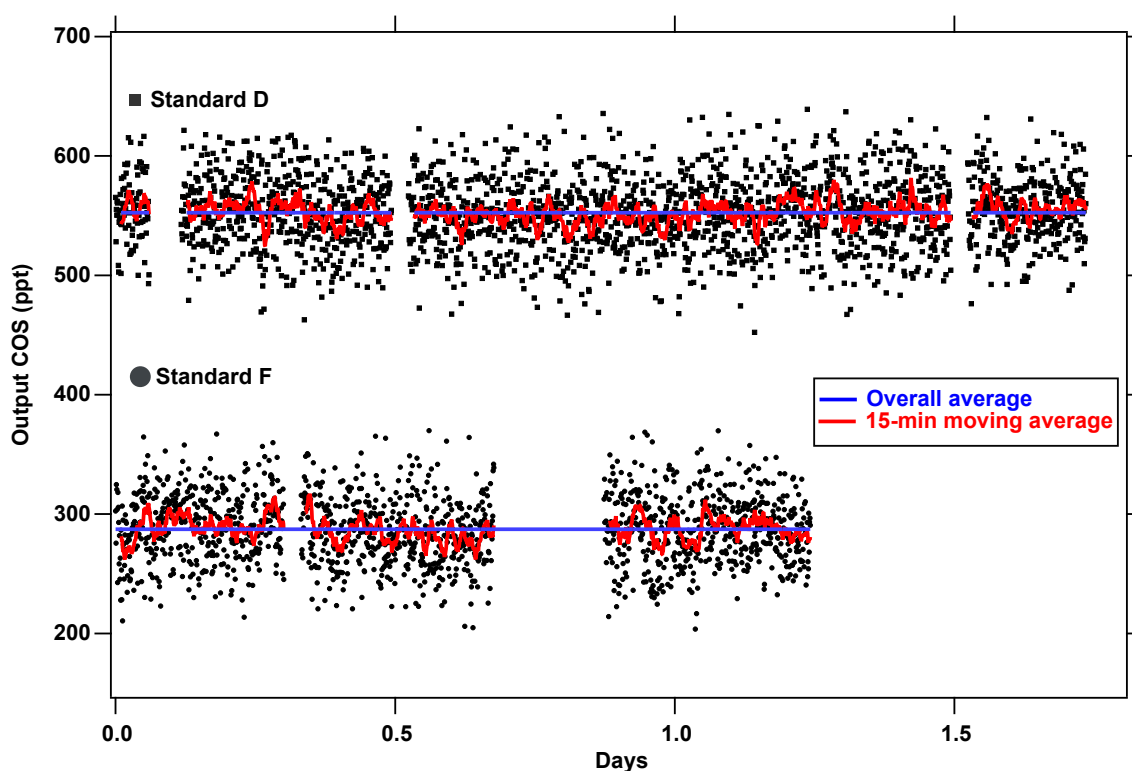


Figure 3. Evaluation of repeatability of COS concentration using standard D or F as a sample and outside air as a reference gas. The blue lines show the overall average, and the red lines show the 15-min moving average COS concentration for standards D and F. The plot is approximately every 1 min cycle. The results for the calibration periods in which standards A to C were injected were removed.

Line 92: How do you “humidify” the dry standard gases by passing them through and ECU set to 2 °C? If there is any amount of water present, cooling the gas would either remove water due to condensation or increase the relative humidity. If no water is removed, the absolute amount or mole fraction should remain constant.

Reply: We have added a more detailed explanation of the ECU in line 103. The ECU prevented water from condensing inside the cell. Urethane tubes were used in the flow path up to the pump, and a Teflon tube was used downstream of the pump. The ECU, featuring a Peltier cooler, facilitated air cooling and disposal of condensed water into the drain. The positive pressure in the ECU forced the collected water to drain from the bottom of the ECU along with the air. Despite maintaining the ECU’s internal temperature at 2 °C, water release was infrequent. Consequently, during periods of elevated humidity, open air passing through the ECU occasionally exceeded 2 °C. Conversely, the dry air flowing through the ECU was humidified by the water retained inside.

Section 2.4: This Section completely lacks the necessary detail. For the little information given here, I can only guess that the MIRO gives out some offset OCS concentration even for samples that do not contain any OCS, and that you assume the reference gas to be OCS free so that it can be used to quantify this offset. But I'm missing proof that this is really the case (see next comment).

Reply: We apologise for the lack of clarity regarding this information. We added Figure S4 to the revised manuscript. The COS concentration in the sample was offset by the reference value, as shown in Figure S4. The value of the reference gas did not change significantly in the short and long terms. We have added this explanation in Section 3.2.

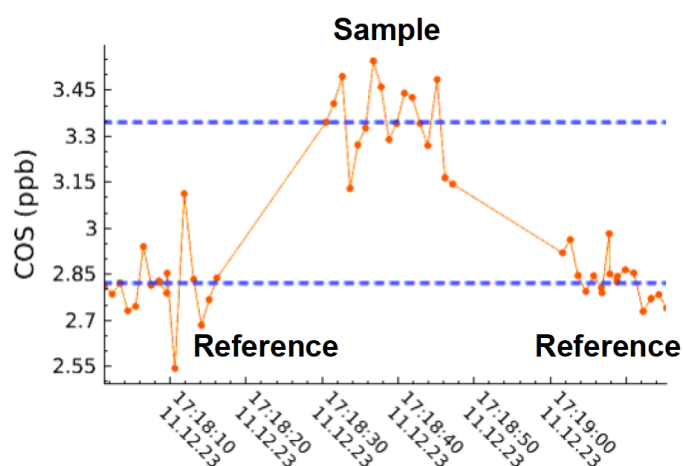


Figure S4. The measured value of COS using MIRA Pico. The COS concentration of the sample was obtained by subtracting the average value of the preceding and subsequent reference gases injected into port 0 from the average value of the gas injected into port 1.

Section 3.1: I have several issues with this section: (i) Why are Alan deviations of the original and modified systems compared for different time constants (5s/160s vs. 40s/180s)? (ii) If I understand it correctly, the comparison was not made for the same sampling procedure, i. e. there were no reference gas injections with the original instrument. The rationale behind this and why you don't think that the results are affected by this should be explained. (iii) When using room air with an activated charcoal filter that does not remove all the OCS, how can you rule out OCS variability in your reference gas? In other words: how can a gas with a low but unknown OCS concentration serve as a reference? (iv) When looking at the Alan deviation plot in Figure 2, it becomes evident that the original instrument performs significantly better in terms of precision at time constants below 10 seconds. This needs to be discussed! When looking at the Alan deviation for longer time constants and considering your modifications, the drifts in the original system appear to arise from temperature instability. This should be discussed. If the red curve is correct,

one take home message of your work is that the original MIRO has severe problems with temperature stabilization that render it useless for long term measurements!

Reply: We apologise for the inadequate explanation. For (ii) and (iii), standard C was used as the sample gas to measure the Allan deviation. No reference gas was injected. Therefore, the Allan deviation results were not affected by the reference, and the original and modified MIRA Pico could be compared. Regarding (i), we described the time of the lowest Allan variance and the time of the highest drift effect. However, because it is difficult to understand, we have revised the statements in Section 3.1 as follows: **The MIRA Pico analyser was dominated by white noise up to an integration time of 5 s and started to drift in an approximately non-linear manner after approximately 30 s. The Allan variance was the smallest at 5 s, with a value of 680 ppt². After modification, the MIRA Pico system exhibited white noise for up to 20 s and started to drift in an approximately non-linear manner after approximately 40 s. The Allan deviation was the smallest at 40 s, at 274 ppt². Regarding (iv), the increase in white noise observed in the modified MIRA Pico is discussed below. The Allan deviation was lower in the MIRA Pico analyser than in the MIRA Pico system at an integration time of less than 5 s. The increase in white noise in the MIRA Pico system was due to vibrations from the refrigerator or fan or the effects of cable extensions.**

As you mentioned, the MIRA Pico is strongly influenced by drift (Figure S3), which affects the precision of the measurement. MIRA Pico is marketed for the observation of atmospheric COS concentrations; however, we recommend that at least some effort be made to stabilise the temperature.

Section 3.2: In this Section, you demonstrate good linearity (Figure 5) and the absence of significant long-term drifts (Figure 4) but that doesn't necessarily translate to good accuracy. It is evident from Figure 4 that, on average, the MIRO tends to underestimate the concentrations of all three standards, which is also reflected by the slope of the calibration curve being 0.90 rather than 1 and the intercept being negative rather zero. I'm missing a discussion why this is the case, and a description if and how the calibration curve is used to correct observed concentrations.

Reply: Thank you for your insightful comment. It does not matter that the slope is not 1, but that the relationship should be a constant linear relationship over a certain concentration range. The fact that the slope is 0.9 does not indicate that the laser has poor detection sensitivity. In addition, with respect to the LGR COS analyser, instead of a straight line with a slope of 1, a correction was made for accuracy, as shown below. Using these four known points, a linear equation of the form, $\text{COS}_{\text{actual}} = m * (\text{COS}_{\text{measured}}) + b$ was applied to the raw data ($b = -43.0 \text{ pmol mol}^{-1}$ and $m = 1.1$) (Berkelhammer et al., 2014).

We have added a detailed explanation of the MIRA Pico calibration in line 232 as follows: **Using the MIRA Pico program, it was possible to calibrate the COS concentration and**

change the slope of the system. However, the short-term COS concentration output was unstable, making the slope calibration using the MIRA Pico program difficult. Therefore, we did not perform the slope calibrations on MIRA Pico. Note that this slope or intercept does not indicate a problem with MIRA Pico detection sensitivity.

Line 137: “*This shows that MIRA Pico programmatically corrects for water content.*” I find it strange that you need to demonstrate this. Whether the instrument makes such an internal correction or not should be information available from the manufacturer. Tests should only be necessary to evaluate it this works or not. Ideally, if there is an internal water vapor correction, information should be given on how exactly this is done!

Reply: We asked the manufacturer about the water vapour correction method. The COS concentration was output from the MIRA Pico analyser in the dry mole fraction with a 1-point linear correction determined by the manufacture. We have added this information in line 80. As you have pointed out, we are now verifying that the MIRA Pico correction is sufficient. We have deleted this sentence accordingly.

Lines 140 – 143: What I see in Figure 6b (and also in Figures 3 and 4) makes it hard to believe the numbers stated in the text. The test period was 6 weeks, so the individual measurement points are hours to days apart, and offsets from the mean or target values for individual points appear to be more on the order of 10 – 30 ppt. I would really like to see the math behind deriving the stated overall uncertainties.

Reply: As you pointed out, some of the deviations for each plot were approximately 30 ppt (Figure 4). To calculate the standard deviation, we constructed a calibration curve using all the plots and calculated the standard deviation for each plot (Figure 5b). The results in Figure 5b also confirm that the maximum shift was approximately 30 ppt. However, if we calculate the standard deviation for these plots, the value was ± 13.7 ppt. Because we adopted the COS concentration values for the last 5 min as one plot, the standard deviation was ± 13.7 ppt for 5-min measurements. Auxiliary lines have been added to Figure 4 to show that the results in Figures 4 and 5 are linked.

Line 151 – 152: Why do you state “*overall uncertainty*” (which I assume to combine accuracy and precision) and “*repeatability*” (with which I assume you mean precision) for different time constants? It is not clear to me what you really want to say.

Reply: There are several ways to evaluate measurement precision, and we thought it would be better to compare the precision obtained by a similar measurement method; therefore, we described it as a preprint. However, this is difficult to understand, and the methods are not exactly the same; therefore, in the end, we only compared the precision used for atmospheric

measurements. Several studies have different methods of evaluating precision measurements, making comparisons difficult. We compared the uncertainty (1σ) of the ambient measurements of COS concentrations. The uncertainty (1σ) was ± 7.3 ppt for 1 min using an Aerodyne COS analyser (Kooijmans et al., 2016) and was approximately ± 7.5 ppt for 1 min using an LGR COS analyser (Berkelhammer et al., 2014).

Line 154: It seems odd that ± 0.5 °C should have such a strong effect on precision. What time scales are we talking about here? And it is laser/detector stability, or is it a temperature effect on the gas concentrations? It could be useful in this context to show the measured temperatures, and possibly try to correlate them with any deviations between observed OCS and the known values from the standards.

Reply: A change of 0.5 °C is a critical change in the MIRA Pico. We measured the cell temperature to prevent it from fluctuating by ± 0.5 °C per day. As Aerodyne's COS analyser also controls at 0.2 °C/30 min, a 0.5 °C change in COS concentration measurement using laser spectroscopy can be a large change. We added Figure S3 to the revised manuscript. As shown in Figure S3, the cell temperature and COS concentration output were significantly correlated. In our measurements, to eliminate this temperature effect, we switched between the reference gas and the sample gas and used the value for the reference as the sample value; however, suppressing temperature changes as much as possible was effective in improving accuracy.

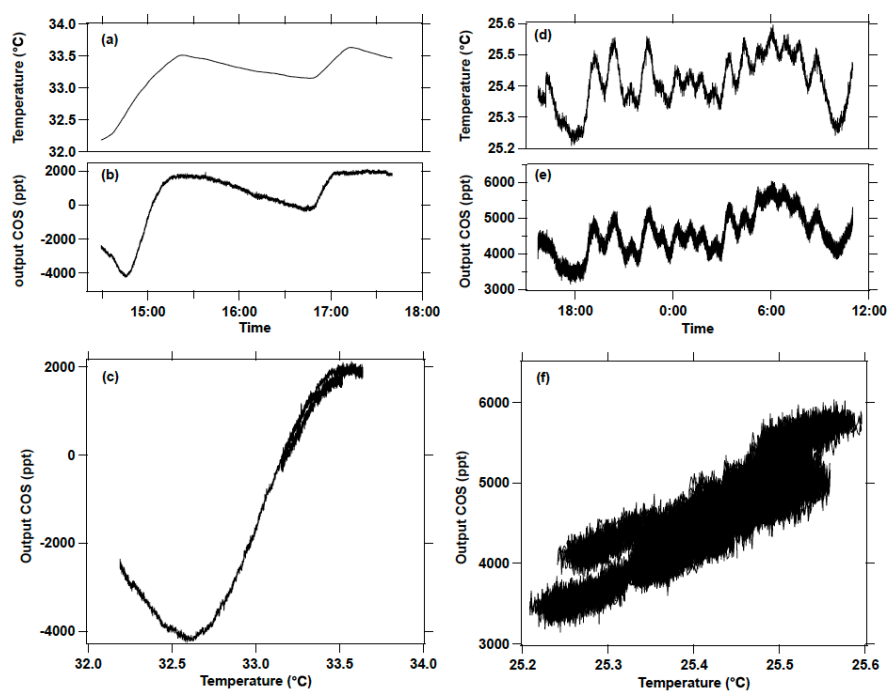


Figure S3. Standard B was injected into the MIRA Pico analyser and the MIRA Pico system for more than 1 h. (a) Temperature variation of MIRA Pico analyser, (b) output COS concentration

of MIRA Pico analyser, (c) relationship between temperature and output COS concentration for MIRA Pico analyser, (d) output COS concentration of MIRA Pico system, (e) temperature variation of MIRA Pico system, and (f) relationship between temperature and output COS concentration for MIRA Pico system.

Line 157 – 164: Are the calibration gas flows used in the cited studies really necessary, or could similar results be achieved with less gas consumption? After all, it is at least possible that the other groups didn't make the strongest efforts to minimize calibration gas flows.

Reply: We agree with this suggestion. We have deleted the comparison between the COS measurement systems for gas consumption.

Section 4.1: The interpretations of the observations sound rather speculative. I'm not saying that the explanations are necessarily incorrect, but it is just not a real interpretation, as is evident from the last sentence (line 196 – 198). I suggest to either do it right (which should not be too difficult for a 10 day period, and you obviously have local weather data at hand) or leave it out of the paper.

Reply: We compared these factors with meteorological data to determine the factors that lead to high COS concentrations. Figure 9 shows the new results of the backward trajectory analysis. These results suggest that the COS concentration in Tsukuba increased due to the influence of anthropogenic COS released from the Keihin Industrial Zone. To connect the conclusion with the content, we have changed the conclusion in line 313 as follows: **Backward trajectory analysis was performed to clarify the relationship between the air mass and COS concentration. Coloured backward trajectories according to COS concentrations are shown in Figure 9. Air masses arriving at the Tsukuba site during periods of high COS concentrations (> 550 ppt) passed southwest. There is an industrial area along the Keihin industrial zone in the southwest at the Tsukuba site. Based on observations conducted from February to April 2001, the Pacific Belt, including the Keihin industrial zone (Figure S2), was reported to be an area of high COS emissions from carbon black production, aluminium production, pigment production, sulphur recovery, and carbon disulphide (CS₂) emissions from rayon production (Blake et al., 2004). Once released, CS₂ is rapidly converted into COS and sulphur dioxide (Chin and Davis, 1993). As described above, there was an influence of diurnal variation; however, the COS concentrations exceeding 550 ppt observed in this study were also likely increased by COS released from the Keihin industrial zone. The MIRA Pico system was used to perform atmospheric observations and provided reasonable results, indicating that this analytical method can be used to observe variations in atmospheric COS concentrations.**

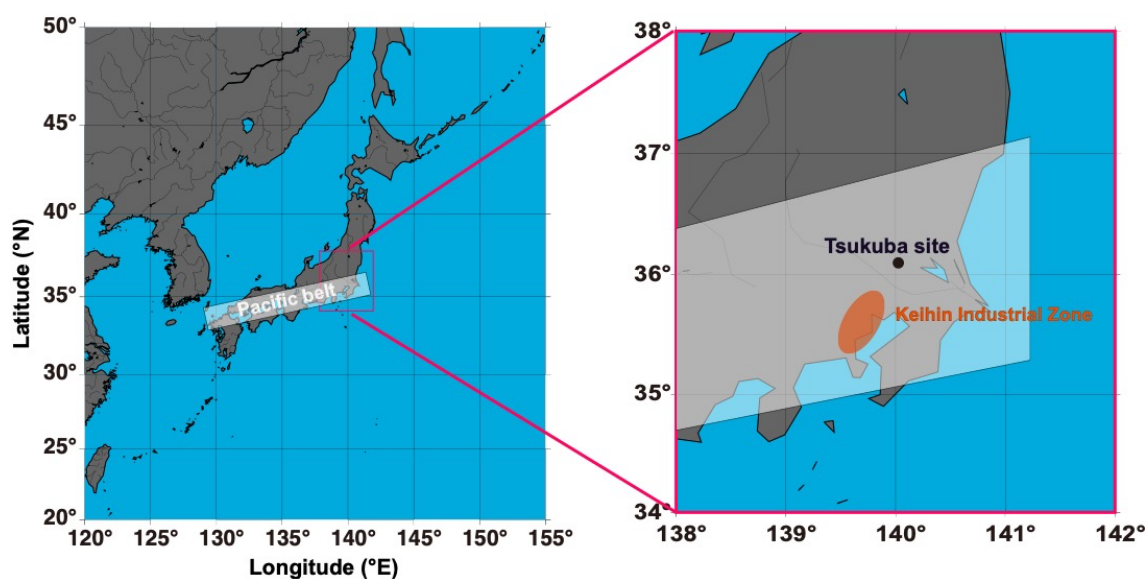


Figure S2. Aerial view of the sampling sites and the location of the Tsukuba site (black). The white area indicates the Pacific Belt of industrial areas in Japan. The brown area indicates the Keihin industrial zone of the Pacific Belt Industrial Zone, the largest industrial zone in Japan.

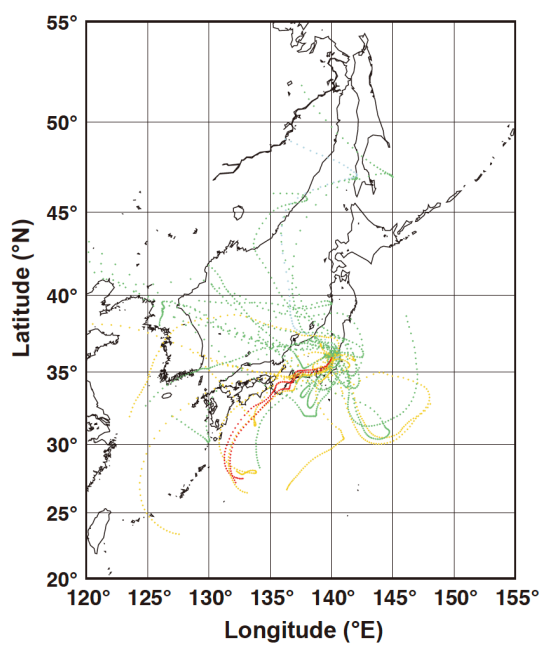


Figure 9. Three-day backward trajectory analysis from 12 to 22 April 2023. Coloured trajectories show COS concentrations observed at Tsukuba every 3:00, 9:00, 15:00, and 21:00 (JST). (Red: ≥ 550 ppt, Yellow: 550–500 ppt, Green: 450–500 ppt, and Aqua: ≤ 450 ppt). Plots show hourly positions.

Line 211 – 215: I wonder what the point is in listing these exact wind speed observations. I suggest to only mention and discuss wind speed in the context of actually trying to explain observed OCS (as attempted in the following paragraph).

Reply: Observing COS concentrations in a car did not allow us to draw a clear conclusion; therefore, we have excluded it from this manuscript.

Line 227 – 229: What does the observed OCS distribution have to do with measurement accuracy?

Reply: We apologise for this ambiguous sentence. This is not related to measurement accuracy and is intended to be accurate in the COS dynamic evaluation. However, we have deleted this section.

Line 230 – 236: There are surely applications where a small, mobile OCS analyzer could be very useful, so efforts to build such a device that produces reliable data are clearly warranted. However, to understand local source and sink processes and overall cycling of OCS, it would be much more useful to measure OCS at the most interesting sites for several days at a time. For a chemically stable gas like OCS, transport and boundary layer dynamics play a key role for local variability, and these are impossible to analyze when taking the instrument from one place to another in a fast moving car.

Reply: We believe that both continuous observations at fixed points and observation by vehicles are important and that different methods should be used depending on the purpose. As you pointed out, long-term fixed-point observations at sites of interest will yield useful information. However, although we were not able to confirm this, we believe it is important to install a measuring device in a car and observe it to confirm the source of a large COS, as in Zanchetta et al. (2023). However, we could not obtain clear results from the COS concentration measurements using a car. Therefore, this information was removed from the manuscript.

Line 239 – 240: The Alan deviation only decreased for time periods larger than 10 seconds.

Reply: We have added the sentence.

Line 249 – 251: The "expected" concentrations do not belong into the conclusions. Summarize important actual observations if there is anything substantial to report.

Reply: We have deleted the inference from the conclusion.

Technical issues:

I always find " pmol mol^{-1} " awkward for mole fractions. Please consider using ppt instead, which is widely used and understood.

Reply: This has been corrected accordingly.

Line 15 and line 247: „...*averaging a standard deviation (1σ) of (505 ± 33) ...*” seems as if 505 is meant to be the standard deviation. The average is 505 and the 1σ standard deviation is 33, and this should be reflected in the wording.

Reply: We have changed the expression to **averaging and a standard deviation (1σ) of (505 ± 33) ppt.**

Line 24: it should be “*ozone depletion*” and not “*ozone depression*”

Reply: This has been corrected accordingly.

Line 24 – 26: the grammar in this sentence appears to be incorrect.

Reply: This has been corrected.

Line 37: The LGR analyzers use off-axis integrated cavity output spectroscopy, not cavity ringdown spectroscopy (there are important differences between the two).

Reply: We appreciate the information. We have changed it accordingly.

Line 62: What is meant by “*standard deviation of 10-min accuracy*”? Accuracy reflects systematic errors or biases and should not depend on any time constant. And what should the standard deviation of the accuracy be relevant for?

Reply: Thank you for the suggestion. We have changed it accordingly.

Line 204: Do you mean 1000 Wh for the batteries?

Reply: Yes. We have changed this and moved it to line 281 for battery information.

Line 212: There is no Supporting info with this preprint!

Reply: We deleted supporting info.

Line 254: The last sentence is obviously incomplete.

Reply: We have changed it.

Figure 3: one zero needs to be removed from the numbers on the y-axis of panel a.

Reply: We do not need to remove one zero on the y-axis of panel a because it is the actual value. The related figure has been moved to Figure S4.

Figure 9: The latitude and longitude panels are not helpful here, as the covered ranges are too small to have any significant effect. To relate the time series to the track on the map, marking a few way points or landmarks by vertical lines in the time series and markers on the map seems the more intuitive solution.

Reply: Thank you for your suggestion. However, we have removed the Figure.