Comment on egusphere-2023-208
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Saveur Belviso

Referee comment by Saveur Belviso (sauveur.belviso@lsce.ipsl.fr) on "Sources and sinks of carbonyl sulfide inferred from tower and mobile atmospheric observations" by Zanchetta et al., Biogeosciences Discussion, https://doi.org/10.5194/egusphere-2023-208, 2023.

Central to this study is the Stochastic Time-Inverted Lagrangian Transport (STILT) model used in combination with anthropogenic emissions inventories (Zumkehr et al., 2018) and biospheric fluxes (from the SiB4 model) to simulate the COS mole fractions at the Lutjewad (LUT) tall tower for the months of January and February 2018. Moreover, in September and October 2019, the authors carried out their own surveys of local anthropogenic sources of COS in the province where the LUT station belongs. For that purpose, in-situ measurements were made on a mobile van using a quantum cascade laser spectrometer (QCLS). These original approaches are valuable for improving our understanding of the sources and sinks of COS in The Netherlands and in Western Europe in general. However, I cannot recommend the publication of this manuscript in its present form because the study is not sufficiently set in the context of former ones. Major revision is required. The three major elements of context that have been overlooked are the following.

Monitoring of COS over Western Europe. Atmospheric COS has been monitored discontinuously for several years (2014-2018) at the Lutjewad tall tower (LUT) in The Netherlands (NL). The whole dataset is of high scientific value because sites where atmospheric COS has been monitored are too few in Europe. Four among the five European monitoring sites are located in Western Europe (LUT- NL (Kooijmans et al., 2016), MHD-IE (Montzka et al., 2007), GIF-FR and TRN-FR (Belviso et al., 2022a)), all gathered in a latitudinal band extending from 48°N to 53°N. Note that atmospheric COS is also monitored discontinuously in the city of Utrecht (UTR-NL) since October 2020 (Baartman et al., 2022). The following comparison of LUT, UTR, MHD and GIF datasets provides strong indication that The Netherlands are a general net source of COS during autumn and winter. The excess of atmospheric COS in The Netherlands is at least equal to 50 ppt. I recommend the authors to highlight these observations as an introduction to their finer scale approaches to sources and sinks of COS in Western Europe. Please better justify why the STILT approach has been applied only to the 2018 survey (see panel A below) when other records exist in the NL (panels B and C).

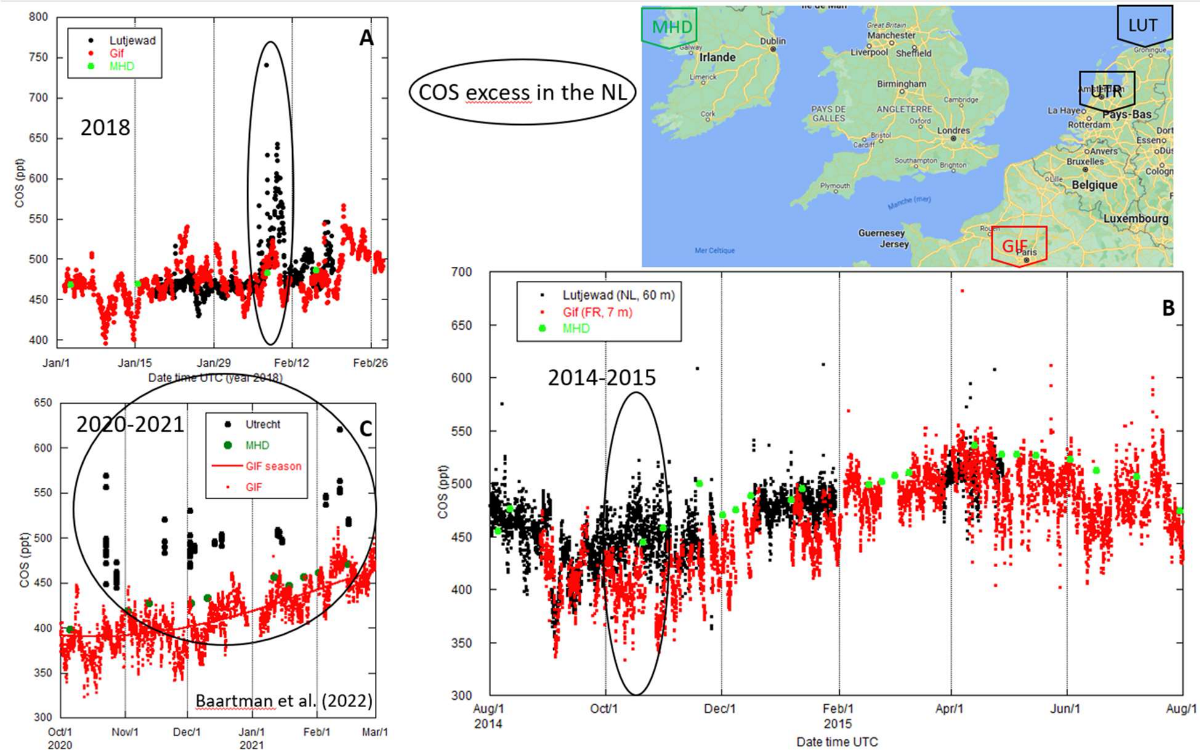
Answer*: the authors thank the referee for this useful insight that had been overlooked. These observations stress even more the importance of a further investigation of local sources at a regional level for the Netherlands. The following paragraph has been added at page 3, lines 21-29 in Section 1 (Introduction):*

Tropospheric COS molar fraction is only monitored in a few sites in Europe. Among these, four monitoring sites are located in Western Europe, within 48°N and 53°N: Mace Head, Ireland (Montzka et al., 2007), Gif-sur-Yvette and Trainou, France (Belviso et al., 2022) and Lutjewad, the Netherlands (Kooijmans et al., 2016). Moreover, COS has been recently monitored discontinuously in Utrecht, the Netherlands (Baartman et al., 2022). The observations in these studies show higher autumn and winter COS molar fractions in the Netherlands than those at Gif-sur-Yvette and Trainou, France. This calls for a more thorough investigation of possible local sources in the Netherlands at a local and regional scale.

Concerning the STILT application, the choice was led mainly by the unusuality of the observations in 2018 and we mostly aimed to disentangle the influence of local and regional sources on these observations. In particular, given the extremely high molar fractions *registered in 2018 and the initial suspects on ploughing activities as a COS source, the authors chose to focus on this particular period alone.*

The sentence starting at page 7, line 43 was modified as follows:

The simulation covers the period from January to February 2018, given the availability for both observations and models data <u>and unusually high COS molar fractions (see Section 3.3)</u> for this period. This analysis aims to disentangle the influence of local and regional sources on these observations.



In section 4.1, the authors concluded that the largest excess of COS recorded at the LUT station between February 5-10 could not be ascribed to air transport from anthropogenic sources inventoried by Zumkehr et al. (2018). Only smaller enhancements measured between February 14-15 were ascribed to known European industrial areas including the Ruhr and the Antwerp-Rotterdam-Amsterdam areas. It would be very interesting to apply the STILT approach to the second large episode of COS accumulation in LUT's atmosphere dated October 2014. Moreover, because the UTR station is located closer than LUT to the potentially important Belgian-Dutch sources of anthropogenic COS, I would recommend the authors to apply the STILT approach to the UTR area too.

***Answer**: the authors thank the referee for this remark and agree it could be very interesting to apply a thorough analysis to the UTR area as well as to other periods when continuous measurements were realized in LUT. However, the authors believe this addition would better fit within a further study aiming to compare regional and/or continental influences on COS monitoring sites and would therefore be beyond the purpose of the study presented in this paper. A combined effort to assess areas that bias COS measurements in different monitoring sites would surely provide a relevant addition to the understanding of COS sources.*

COS seasonal cycle amplitudes over Western Europe. LUT data is also used to investigate the amplitude of the seasonal variations at this site (cf. Fig. S1 copied below). In the legend of Fig. S1, the authors state that “The seasonal cycle shows a peak-to-peak amplitude of 87 ppt, which was estimated to be 96 ppt by Kooijmans et al. (2016) when no flask measurements were included.” I recommend the authors to compare their observations during the period 2014-2018 with the atmospheric seasonal cycle amplitudes (SCA) assessed over MHD (Montzka et al., 2007) and GIF (Belviso et al., 2022b).

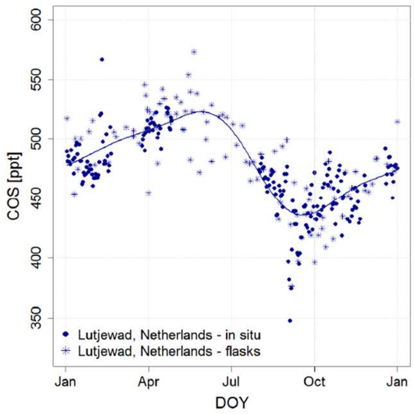
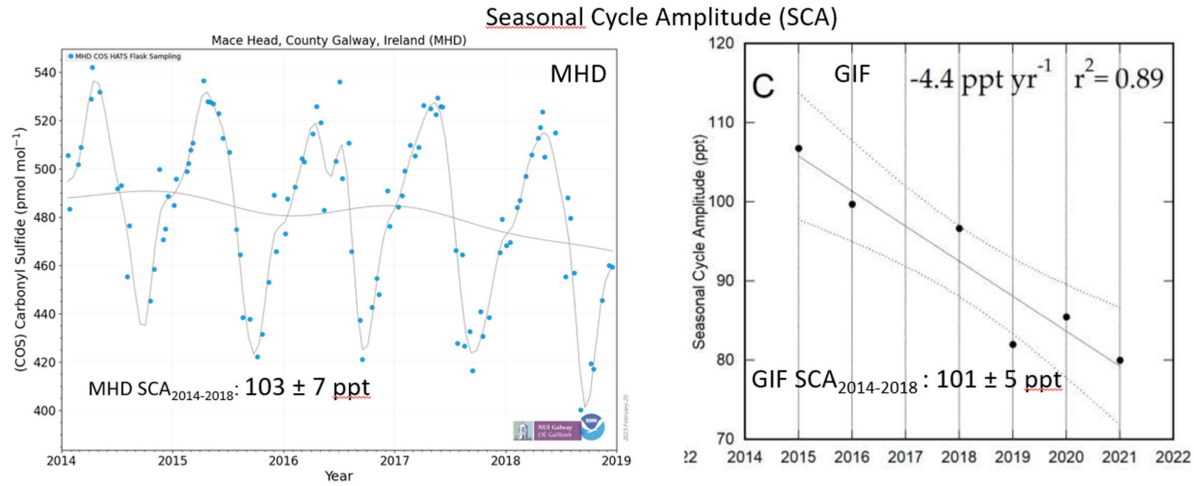


Figure S1: Seasonal cycle of daytime average COS mole fractions at 60 m in Lutjewad. The data consist of in-situ measurements from August 2014 – April 2015 and January – February 2018 (circles) and flask measurements between December 2013 and February 2016 (stars). The in-situ measurements from August 2014 – April 2015 are an update of the measurements presented in Kooijmans et al. (2016). The seasonal cycle shows a peak-to-peak amplitude of 87 ppt, which was estimated to be 96 ppt by Kooijmans et al. (2016) when no flask measurements were included.



COS SCA is significantly lower (about 15 ppt lower) at LUT than at MHD or GIF. What are the implications for biogenic fluxes of lower SCA in The Netherlands than elsewhere in Western Europe? How does the COS background of Fig. S1 compare with that estimated using the end point of the STILT model trajectories in the analysis domain and the derived 3D concentration fields from the Transport Model 5 – Four-Dimensional Variational model (TM5-4DVAR) inversions (Ma et al., 2021)?

***Answer**: the differences in COS SCA’s have already been mentioned in Kooijmans et al. (2016), where the COS SCA was analyzed for LUT in 2014-2015. This is also the reason why this analysis, which has been realized by adding in-situ observations to the same dataset, was reported as supplementary information. The SCA’s have been depicted in Figure 13 of Kooijmans et al. (2016), which has been copied below.*

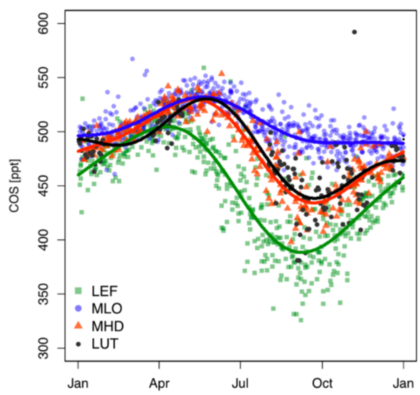


Figure 13. COS seasonal cycle of four sites: Wisconsin, USA (LEF); Mauna Loa, USA (MLO); Mace Head, Ireland (MHD); and Lutjewad, the Netherlands (LUT); the data of the latter site are presented in this study. COS mole fractions for the LEF, MLO and MHD sites were measured from flask samples at a GC-MS by NOAA/ESRL (Montzka et al., 2007). The NOAA/ESRL data are shown as flask pair means from individual sampling events. All NOAA measurements are plotted as function of time of the year and cover a period between 2000 and 2015 for LEF, MLO and MHD. In situ COS measurements with the QCLS at the Lutjewad site during 2014–2015 are shown as daily averages (black). A two-harmonic seasonal cycle is fit through the data.

Citing Kooijmans et al. (2016):

"In Fig. 13 we compare COS mole fractions from Lutjewad with that from three other sites as measured from flask samples with a GC-MS by NOAA/ESRL Montzka et al. (2007). The flask samples cover data between 2000 and 2015 for Wisconsin, USA (LEF), and Mauna Loa, USA (MLO), and between 2001 and 2015 for Mace Head, Ireland (MHD). These data are an update of those presented in Montzka et al. (2007) (data available at ftp://ftp.cmdl.noaa.gov/hats/carbonyl_sulfide/). The flask measurements in Fig. 13 are plotted as function of time of the year. The high-altitude MLO site is less directly influenced by terrestrial ecosystems and therefore shows only small seasonal variation, in contrast to the LEF site, which is largely influenced by (forested) continental air. The Lutjewad COS mole fractions are most consistent with measurements from MHD, which can be expected since both stations are coastal sites and are located at similar latitudes. The seasonal amplitude of COS at MHD and Lutjewad is in between that of LEF and MLO, most likely because both sites are not solely influenced by marine or continental air but by both types of air masses. The COS mole fraction has a minimum in September and October and is a few weeks later than the minimum of the CO2 mole fraction. Montzka et al. (2007) and Blonquist et al. (2011) also observed a COS minimum later than that of CO2. They reasoned that this difference is due to the fact that at the end of the growing season COS mole fractions keep decreasing due to vegetative uptake without at the same time having a source of COS, whereas during this time of year respiration is beginning to offset assimilation in determining the ambient CO2 mole fractions".

It is interesting to notice that according to Kooijmans et al. (2016) and contrasting the referee's comment, MHD and LUT show very close SCA's, with Kooijmans et al. concluding that the seasonal cycle features are probably determined by the monitoring site location.

Concerning the agreement between TM5-4DVAR background obtained by boundary conditions and its comparison with COS SCA presented in Figure S1, the investigation is currently limited by data availability. Only particles dispersion files for January and February 2018 were made available, therefore it is only possible to compare the COS SCA of the first 59 days of the year (see Figure A below). Generally, the available modelled data falls within the range of observations, but of course this cannot *be considered a thorough analysis. It would be interesting to include this interesting insight in future works, possibly within a comprehensive intercomparison of different measuring stations.*

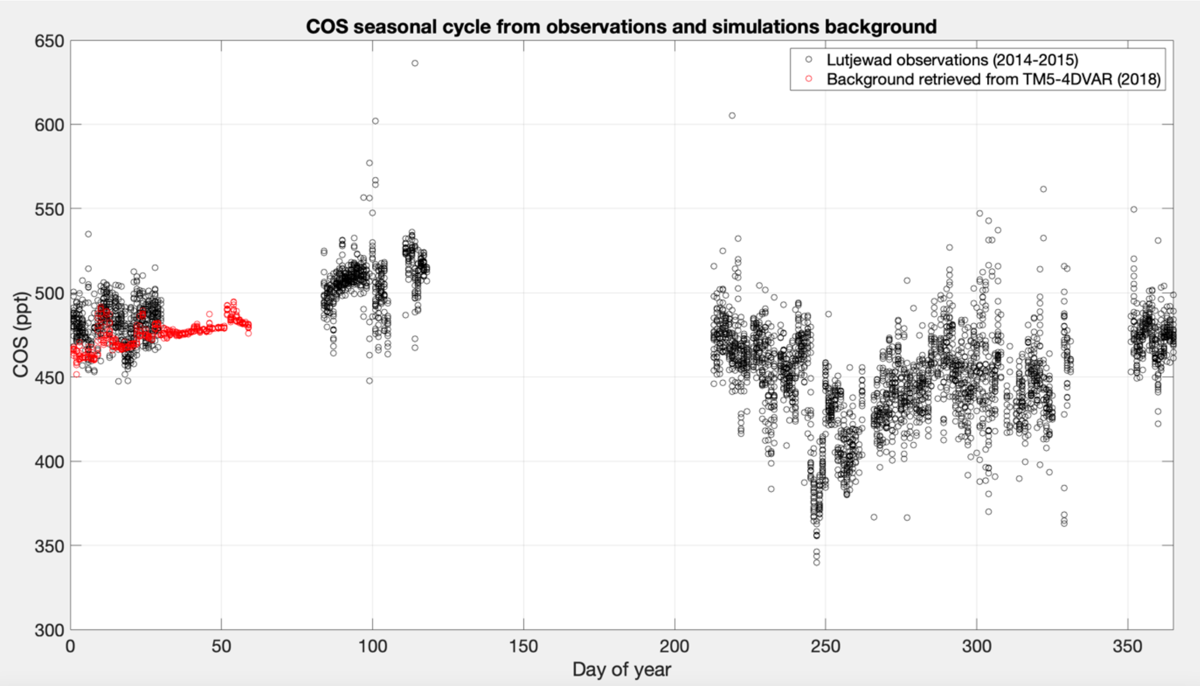


Figure A: comparison of seasonal cycles of COS measured in Lutjewad and retrieved from TM5-4DVAR boundary conditions for January and February 2018.

Evaluation of SiB4 simulations at LUT using observed nighttime fluxes of COS. The authors estimated nighttime fluxes of COS based on the radon-tracer method, but, surprisingly, did not make any further use of those estimates in the manuscript. The authors could take the opportunity to compare SiB4 simulations of nighttime biogenic fluxes at LUT with field observations.

Because the STILT simulations are of central importance to the study, the way the STILT methodology is illustrated in the manuscript (cf. Fig. 2) is very disappointing. Figure 2c identifies the sources influencing Lutjewad in North-Eastern Germany 10 days before the start of the atmospheric COS survey at LUT. The associated COS enhancement is in the range 0-2 ppt/grid cell. One would conclude that the impact of the sources inventoried by Zumkehr et al. (2018) in North-Eastern Germany is estimated to be negligible to a COS enhancement that has not been quantified in the field... In fact, Fig 2c does not really identify the sources influencing Lutjewad in NE Germany because the anthropogenic and biogenic contributions are not separated from each other. Moreover, the color scale adopted in Fig. 2b does not allow at all localizing the direct COS sources inventoried by Zumkehr et al. (2018). I would use a log scale in the range 1 to 1000 pmol $m^{-2} s^{-1}$. Figures 2b and 2c are misleading and should be redrawn. A date belonging to period 4 could be chosen to better illustrate the enhancements attributed to industry in the Ruhr area. Larger panels are required. It would be also interesting to document the largest excess of COS recorded at the LUT station between February 5-10 (period 3) the one that could not be ascribed to air transport from anthropogenic sources inventoried by Zumkehr et al. (2018).

Answer: *the authors agree with the referee's comment. Unfortunately, SiB4 data for Lutjewad were requested only for January and February 2018. The average COS nighttime flux was estimated at Lutjewad coordinates over these two months and resulted to be -2.1 ± 0.2 pmol/(m^2*s). On page 10, Lines 22-24, the following sentence was added:*

The average SiB4 COS nighttime (9PM – 6AM) flux was retrieved for Lutjewad (53.4°N, 6.3°E) for January and February 2018 and was estimated to be -2.1 ± 0.2 pmol m^{-2} s^{-1}.

On the other hand, the problem for Figure 2c was not the missed separation between biospheric and anthropogenic fluxes, but a missing "log" in the reported unit of measure. We have chosen 15 February 2018 of period 4, as an example, to illustrate the COS enhancements attributed to industries in the Ruhr area. The figures have been redrawn with larger panels and a log scale as suggested, adopting restricted color scales to help the visualization and the caption has been modified as reported below:

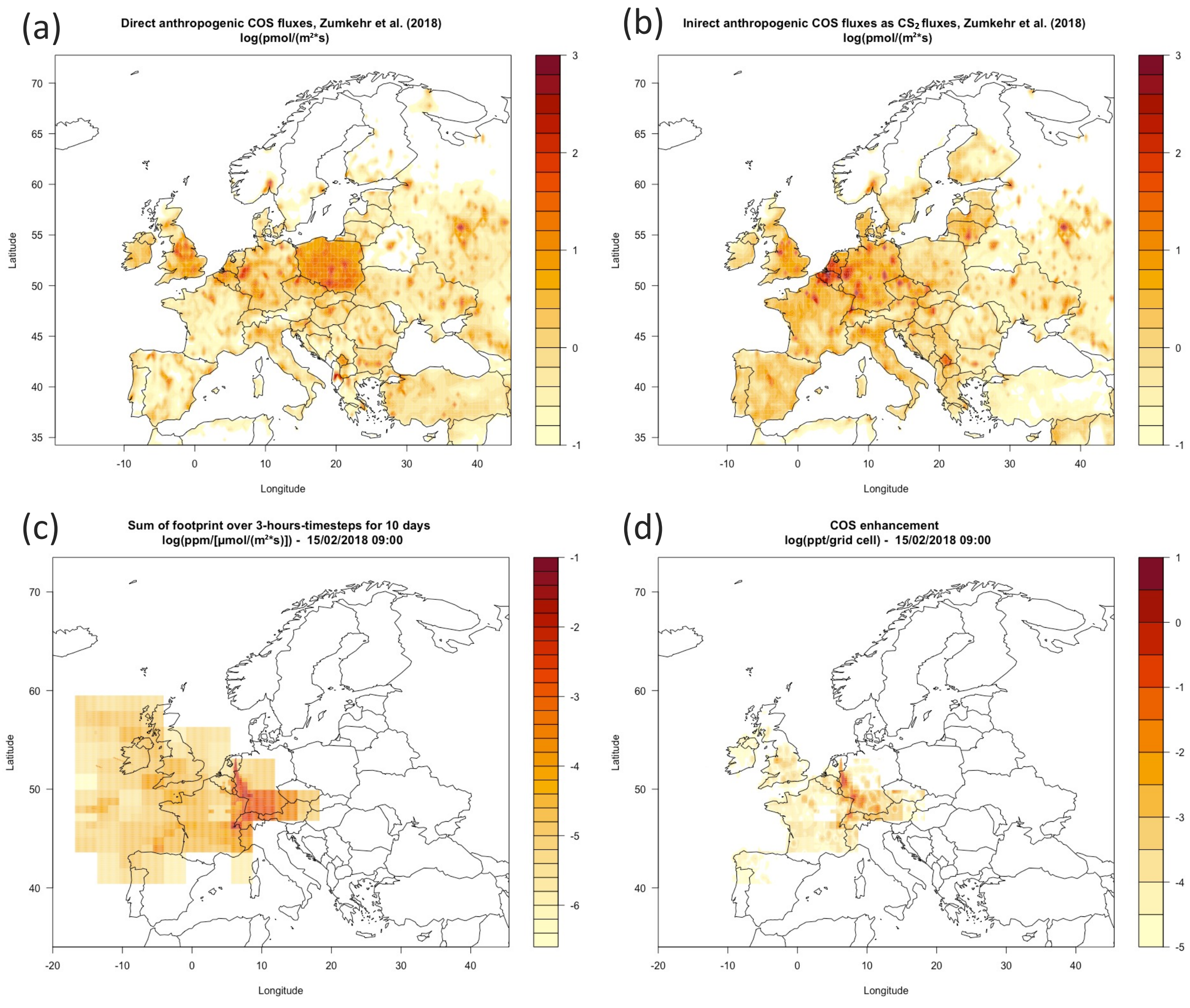


Figure 2: reported in logarithmic scales: panels (a) and (b) show the localized COS and CS_2 sources according to Zumkehr et al. (2018), (c) shows an example of localized footprint values resulting from the STILT model simulations, summed over 10 days before the starting timestep (15/02/2018, 09:00), (d) the modelled enhancement resulting from the product of footprint and fluxes (see Section 2.4), identifying the sources influencing Lutjewad in the Ruhr area (the ranges of these scales were adjusted for clarity purposes).

Other methodological aspects to be clarified are the following:
-100 particles released for 10 days back in time: isn't it a too small number of particles?

***Answer**: the authors believe the number of released particles for these simulations is sufficient, since it is in line with other studies related to STILT applications (see, for example, Galkowski, 2015; Gerbig et al., 2003; Maier et al., 2022; Thilakan et al., 2022; Van Der Woude et al., 2023). Maier et al. (2022), in particular, report a case-sensitivity analysis between 100 particles over a 3-days dispersion against*

500 particles over a 10-days dispersion, which resulted only in minor differences. For this study, the 10-days-long dispersion was necessary given the estimated lifetime of CS_2.

-Is the horizontal resolution of the ECMWF-IFS database of 0.1°x0.1° or coarser?

***Answer**: the ECMWF-IFS resolution is 0.25°x0.25°. Page 8, line 5 was modified to include this information as follows:*

…"driven by ECMWF-IFS operational analysis <u>at a 0.25°x0.25° resolution</u>"…

-At what time are the particles released to the atmosphere?

***Answer**: the particles are released to the atmosphere at the time when the observations were made, and the particles are transported backward in time based on 3-hourly wind fields covering January and February 2018.*

Page 8, lines 11-17 were modified as follows:

<u>"The STILT model establishes the link between the emissions in the upwind influencing area and the measurements at a defined location and time. This is realized by releasing particles to the atmosphere that are driven by meteorological winds and transported backward in time to determine the origin of air parcels influencing the measurements. Each simulation run releases 100 particles from the Lutjewad station, at a height of 60 m. The transport of these particles is reconstructed within the selected domain (latitude 34.0°N-73.5°N, longitude 20.0°W-45.5°E, to cover Europe), in 3-hours timesteps over 10 days back in time."</u>

Figure 7. Again, I don't understand the reason why the authors provided modelled COS concentrations when observations are not available (e.g., Fig. 7, right column, red curves, dates before 01-18-2018 17:00 and after 02-19-2018 8:00:00). The consequences are that the difference between measurements (black curve) and modelled values (red curves) are poorly visible. Please redraw Figure 7 accordingly. As an alternative, the contributions of background, background + biogenic fluxes, background + biogenic fluxes + direct anthropogenic emissions, background + biogenic fluxes + direct & indirect anthropogenic emissions could be displayed on the same plot. Data displayed in Fig. 7 and Fig. 9 could be combined by plotting background + biogenic fluxes + direct & indirect anthropogenic emissions + local sources identified from mobile flask and in-situ measurements.

***Answer**: the authors agree with the referee and would prefer to keep these figures separated given the different purpose of the two (e.g. showing contributions of different parameters on one and of the newly introduced sources on the other). Therefore, Figure 7 has been redrawn with a shorter x-axis, comprising only the period when measurements are available.*

I also question the interest of Figure 3, where the deviation of mole fractions of COS from their seasonal cycle in Lutjewad is compared, because the COS background at 60 m set from data gathered in Fig. S1 is not well constrained for the months of January and February. I would rather suggest the use of cluster analysis applied to HYSPLIT back trajectories calculated every 3 h at the LUT site during the months of January and February 2018.

***Answer**: Figure 3 reports the usual relationship between observed trends in COS, CO_2 and CO molar fraction enhancements or depletions and wind directions, compared to the average measured seasonal* *cycle. The intention of both Section 3.1 and Section 3.2 is to show the results which set the context for the measurements described in the following paragraphs. However, the authors recognize Section 3.1 was rather self-standing and not well contextualized within this study. Therefore, it has been moved together with Figure 3 and Section 4.2 to the supplementary materials. The title of the new Section S1 was modified as "Observed deviations from seasonal cycles by wind directions during stationary measurements". Figure 3 has now become Figure S1.*

Other comments of less importance are listed below:

-Title: Sources and sinks of carbonyl sulfide inferred from tower and mobile atmospheric observations in the Netherlands

***Answer**: the authors agree with the referee's suggestion. The title has been modified accordingly.*

-page 2, line 36: remove "on average"

***Answer**: done.*

-page 3, line 5: NOAA data can be visualized on-line at https://gml.noaa.gov/dv/iadv/

***Answer**: on page 3, line 8, it was added "This dataset is still being updated and can be visualized online (NOAA, 2023)."*

-page 3, line 9: ...were analyzed by gas chromatography and mass spectrometry.

***Answer**: corrected.*

-page 3, line 18: Moreover, this instrument enabled the collection of...

***Answer**: corrected.*

-page 6, line 8 and Table 2: no overview of the average precision is given in Table 2. Remove Table 2.

***Answer**: page 6, line 8 was modified as "the average precision for the 2014-2015 period was 5.3 ppt". However, the authors prefer to keep Table 2 since it reports also the different measurement heights and times in the measurement periods.*

-page 12, line 12: CO molar fractions are not displayed in Fig. 5.

***Answer**: the sentence has been modified: "...CO_2 and CO (this latter not shown in Figure 4) molar fractions…"*

Page 13: this very descriptive paragraph should be rewritten in order to better identify the data in Fig. 5 and Fig. 6 to which the authors refer to. A letter should be attributed to each panel to guide the reader.

***Answer**: the authors included letters in the panels and references to Figure 5 and Figure 6 throughout the paragraph. However, the authors would prefer to keep the paragraph in its current form since it includes some of the key findings of this study – namely, the mismatch between modelled results and measurements in Period 3 – which could be better described with a thorough description and comparison against the other selected periods of interest.*

Page 16: Please provide an illustration of how the COS fluxes were calculated with in-situ measurements collected at ground level. Is it realistic to use a Gaussian dispersion model when the vertical distribution of COS remains unknown? Was a 3D sonic anemometer coupled with the QCLS?

***Answer**: an example of measurements is provided in Figure B below (location: SuikerUnie, 53°12'N, 6°30'E). The authors acknowledge that major simplifications were made to apply the Gaussian dispersion model for these estimations. The peaks measured during the mobile sampling campaign closely followed a Gaussian shape. Unfortunately, there were no 3D-sonic measurements coupled with this sampling campaign. Furthermore, in most cases the exact location of the emission sources remained unknown (e.g., it was not possible to see any industrial chimney, or there was no visible plume). Therefore, it was necessary to assume that emissions occurred at the sampling height. The emissions were reconstructed using the parametrization of σ_y and σ_z defined after Pasquill-Gifford stability classes (Csanady, 1973), estimated after a Monte Carlo simulation based on distance from the source and wind speed (obtained from approximated estimates from Google Maps and weather data). This, in fact, results in fluxes uncertainties that range between 44% and 92% of the estimated flux means. The authors would like to stress that the Gaussian plume modelling was applied to obtain some rough estimate; the used approach is not considered a reliable representation of reality, but rather a tool to get the best estimate possible given the available data.*

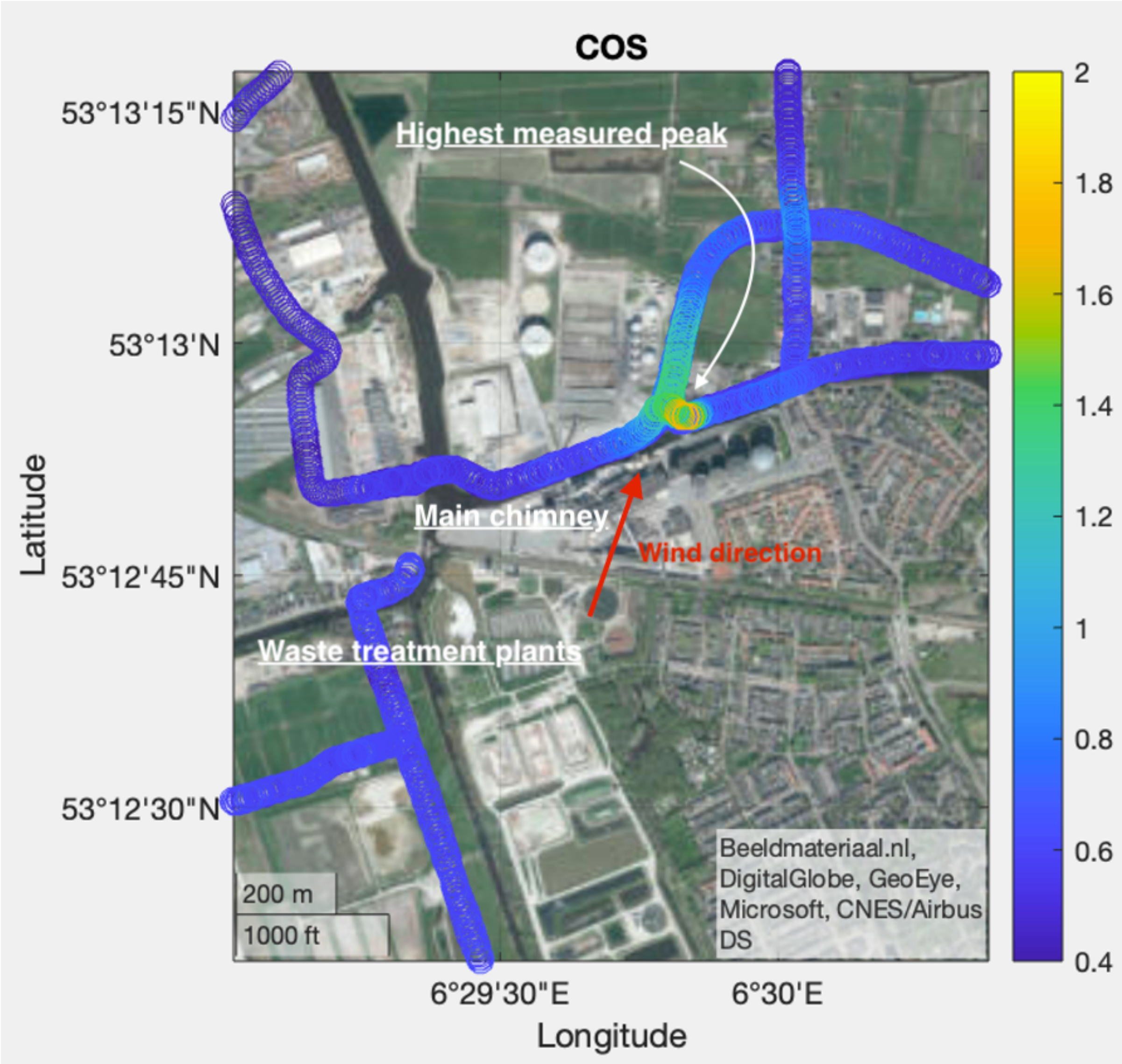


Figure B: COS in-situ measurements at SuikerUnie, now CosunBeet Company, 53°12'N, 6°30'E (Hoogkerk, Groningen, NL).

Page 19, line 6: Do you mean that rapeseed is grown in the Groningen province in spring and that soils are fertilized in winter with rapeseed byproducts?

Answer*: indeed, rapeseed was grown in some fields of the Groningen province in spring and the byproducts were occasionally used as fertilizers. The sentence on page 17, line 25 was modified as follows: …"and, <u>knowing that rapeseed was grown in some fields in the province of Groningen, it is still possible that a fertilizer based on rapeseed byproducts</u> (Belviso et al., 2022) or the soil act as a COS source occasionally"…*

Page 19, line 31: Are you aware of any explosions at ESD-Sic in October 2014 when atmospheric COS levels at LUT were over 500 ppt?

Answer*: unfortunately, it was not possible to obtain detailed records of all the explosions occurred at ESD-SiC preceding 2018. An interrogation presented at the Second Chamber of the Netherlands (Lacin, 2019) (Dutch only) reports 148 explosions between 2014 and 2017. A TNO investigation on the composition of soot in ESD-SiC explosions (Tromp & Duyzer, 2019) (Dutch only), reports the explosions per year between 2014-2018 to be 50, 35, 29, 34 and 30, respectively. Moreover, a local newspaper* *reported the villages of Meedhuizen and Tjuchem, situated West of ESD-SiC, to be covered in soot following an explosion at ESD-SiC on Friday, January 23rd 2015 (RTVNoord, 2015). As shown in Figure C, on such date COS enhancements were measured in Lutjewad for about 3 hours. Unfortunately, particle dispersion files were not (made) available for 2015 and therefore it was not possible to perform a thorough analysis of air transport for this period. However, the gathered information suggests that, at least for January 2015, ESD-SiC may have already been the cause of the measured enhancements.*

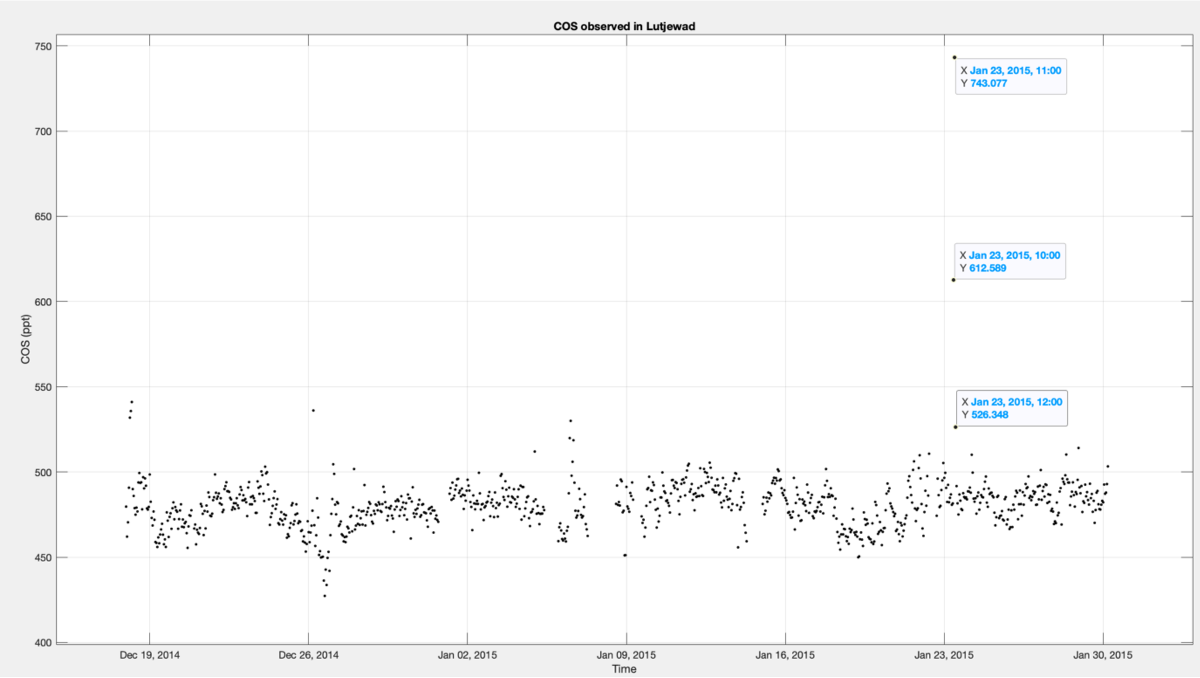


Figure C: measured COS in Lutjewad between December 2014 and January 2015.

Last remark.

I would like to inform you of the existence of a manuscript entitled "The Z-2018 emissions inventory of COS in Europe: a semiquantitative multi-data-streams evaluation", authored by I. Pison, J.-E. Petit, A. Berchet, M. Remaud, L. Simon, M. Ramonet, M. Delmotte, V. Kazan, C. Yver-Kwok, M. Lopez and myself (S. Belviso), in press in Atmospheric Environment. I will be keen to share a preprint with you upon request. Chapter 3.3 provides examples of cluster analysis of winter COS measurements and back trajectories. One event is dated February 2018.

The authors would like to thank the referee for the useful comments, which helped to get a more comprehensive overview for this study and to contextualize it within the existing observational network. *A last sentence was added at the end of the manuscript (page 20, lines 17-22): "Our study demonstrates that the influence of local to regional anthropogenic sources should be considered when using COS measurements as a tracer for GPP, especially for atmospheric measurements that are close to urban areas. This approach, combining COS stationary measurements, mobile measurements and models, could be applied in other existing measurement locations. It could allow a broader assessment of local anthropogenic influences, to prevent biases in COS budget and seasonality estimates.".*

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