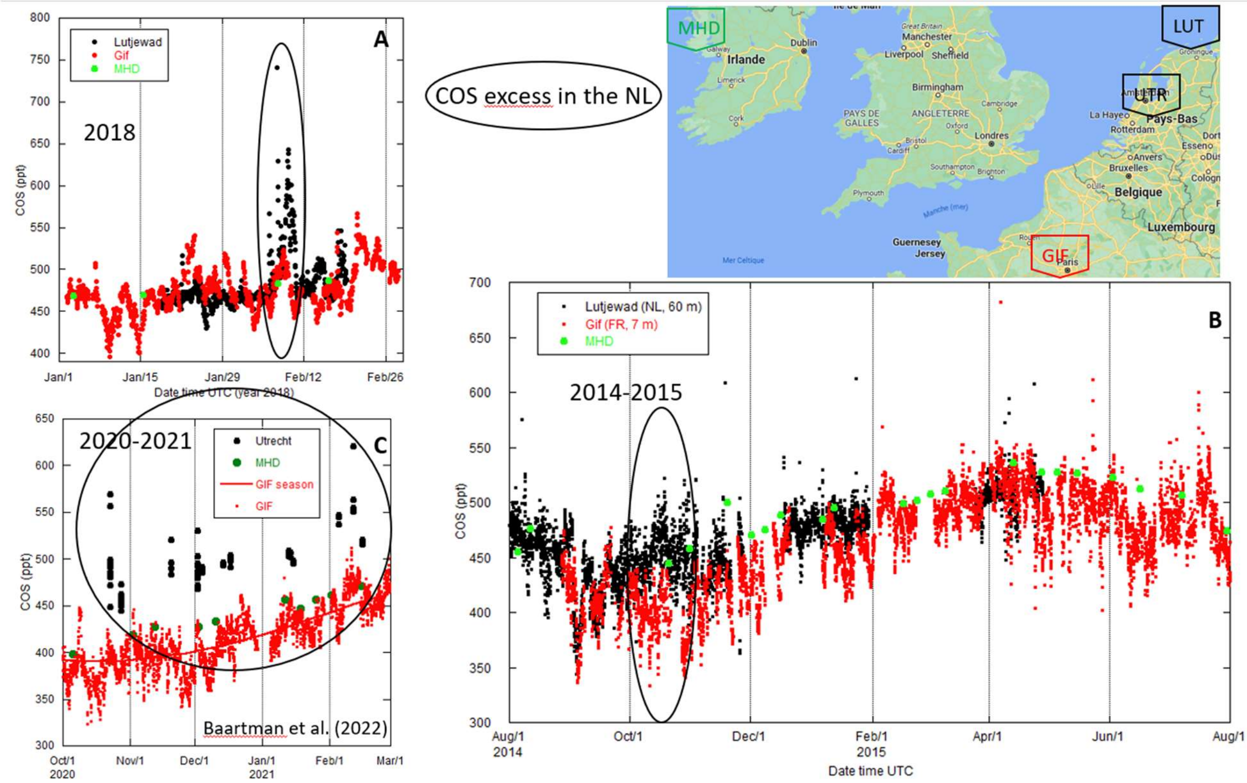
Central to this study is the Stochastic Time-Inverted Lagrangian Transport (STILT) model used in combination with anthropogenic emissions inventories (Zumkehr et al., 2018) and biospheric fluxes (from the SiB4 model) to simulate the COS mole fractions at the Lutjewad (LUT) tall tower for the months of 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).



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.

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).

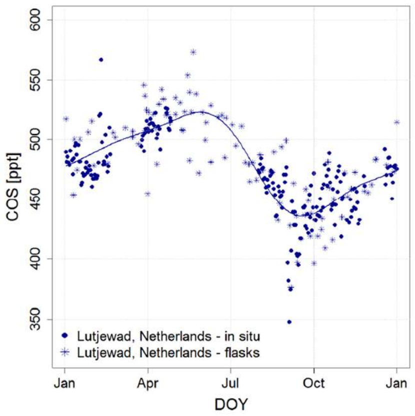
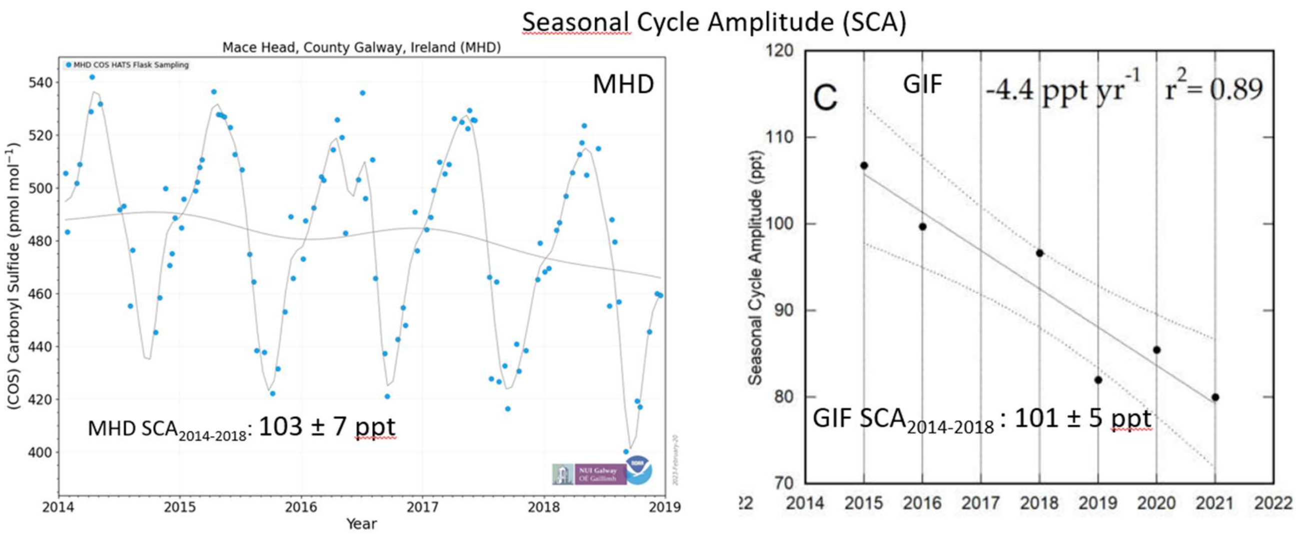


Figure S1: Seasonal cycle of daytime average COS mole fractions at 60 m in Lutjewad. The data consist of in-situ measurements from August 2014 – April 2015 and January – February 2018 (circles) and flask measurements between December 2013 and February 2016 (stars). The in-situ measurements from August 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)?

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).

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?

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

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

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.

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.

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

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

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

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

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

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

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

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.

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?

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?

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?

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.

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