Supplementary information

Seasonal fit

A non-linear least squares fit was made to the 60 m COS mole fractions from Lutjewad, see Figure S1. The shape of the fit is represented by a harmonics function after Thoning et al. (1989, eq. 1 therein). We used the highest available heights, such that the mole fractions are the least affected by local influences, and we selected only daytime data, such that the measured mole fractions are not influenced by the shallow nocturnal boundary layer. The seasonal fit of CO_2 (not shown) is based on continuous measurements of a co-located cavity ring-down spectrometer in 2014 and 2015 in Lutjewad. For the seasonal fit of CO_2, we selected only data with wind direction from the north (wind direction < 30 ° or > 260 °) to make sure that the data represent background air and are not affected by anthropogenic influences. This data selection was based on the wind direction analysis presented in **Error! Reference source not found.**.

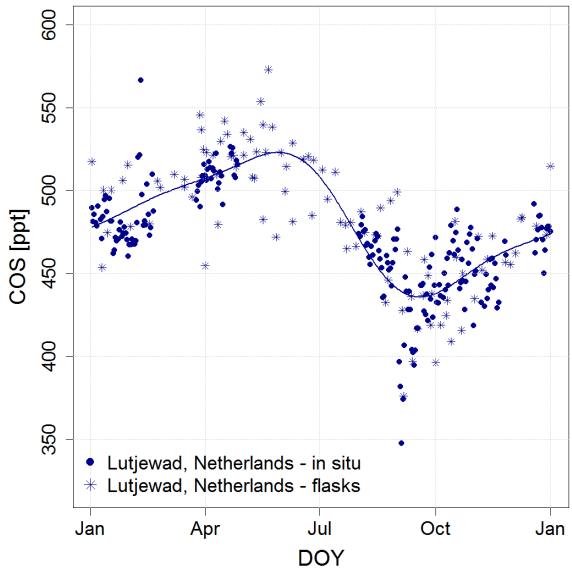

Figure S1: Seasonal cycle of daytime average COS mole fractions at 60 m in Lutjewad. The data consist of in-situ measurements from August 2014 – April 2015 and January – February 2018 (circles) and flask measurements between December 2013 and February 2016 (stars). The in-situ measurements from August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

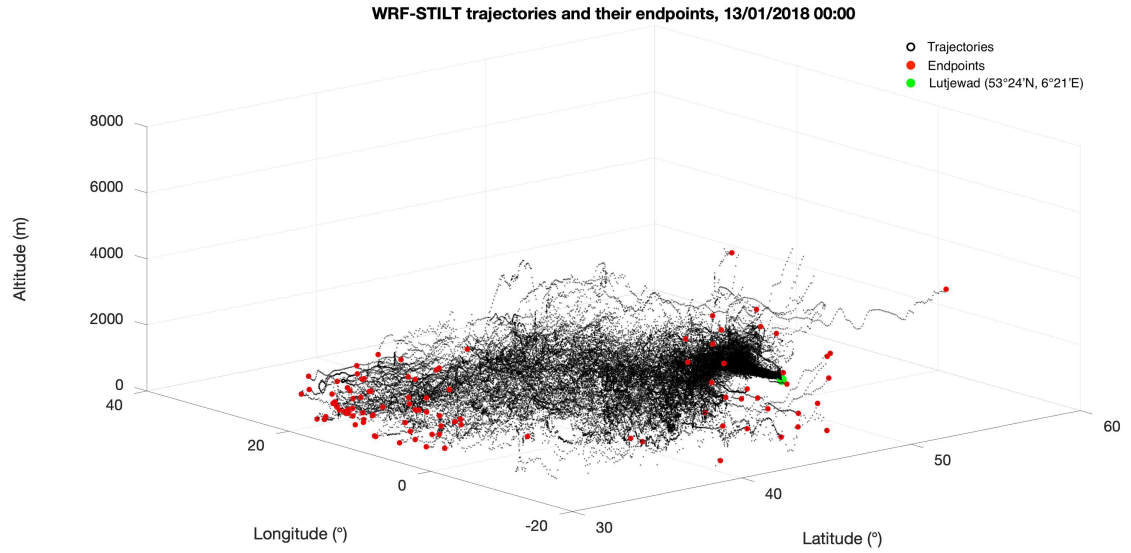

Figure S2: the trajectories of the 100 particles starting on 13/01/2018 00:00 from Lutjewad and their endpoints, according to the STILT model. Each endpoint (coloured in red) was associated to a COS boundary concentration following the TM5-4DVAR model (Ma et al., 2021). The average of these COS concentrations was used as the COS background for the simulation.

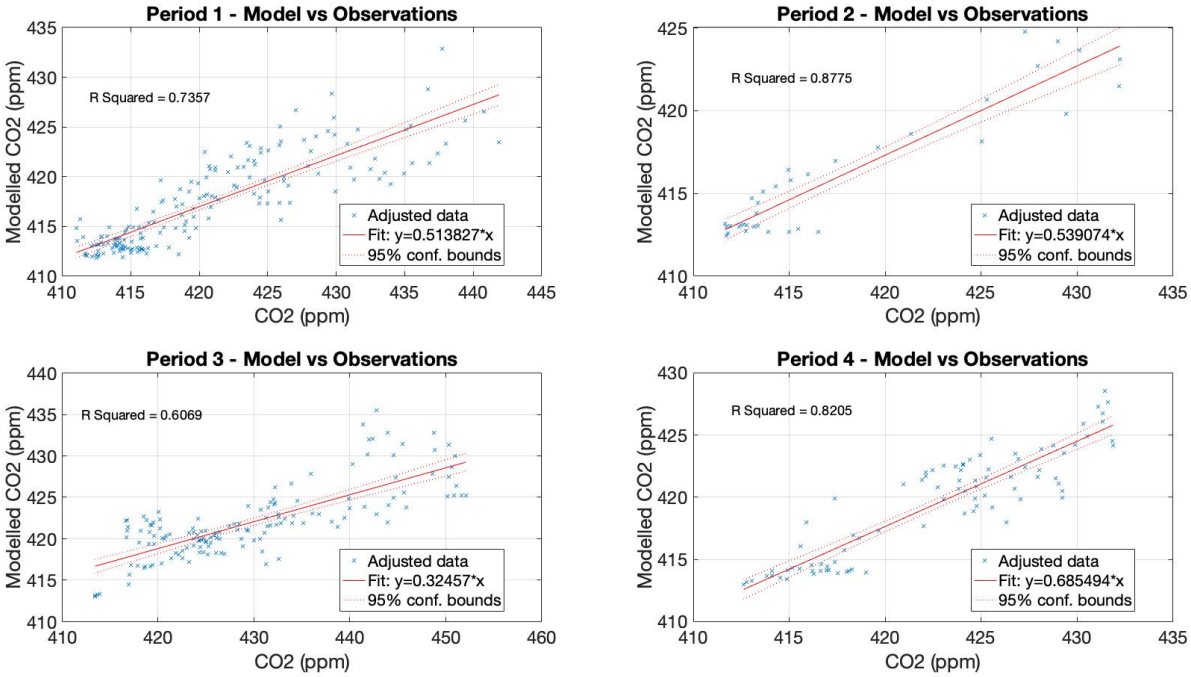

Figure S3: relationships between modelled CO_2 results and observations for the selected time periods.

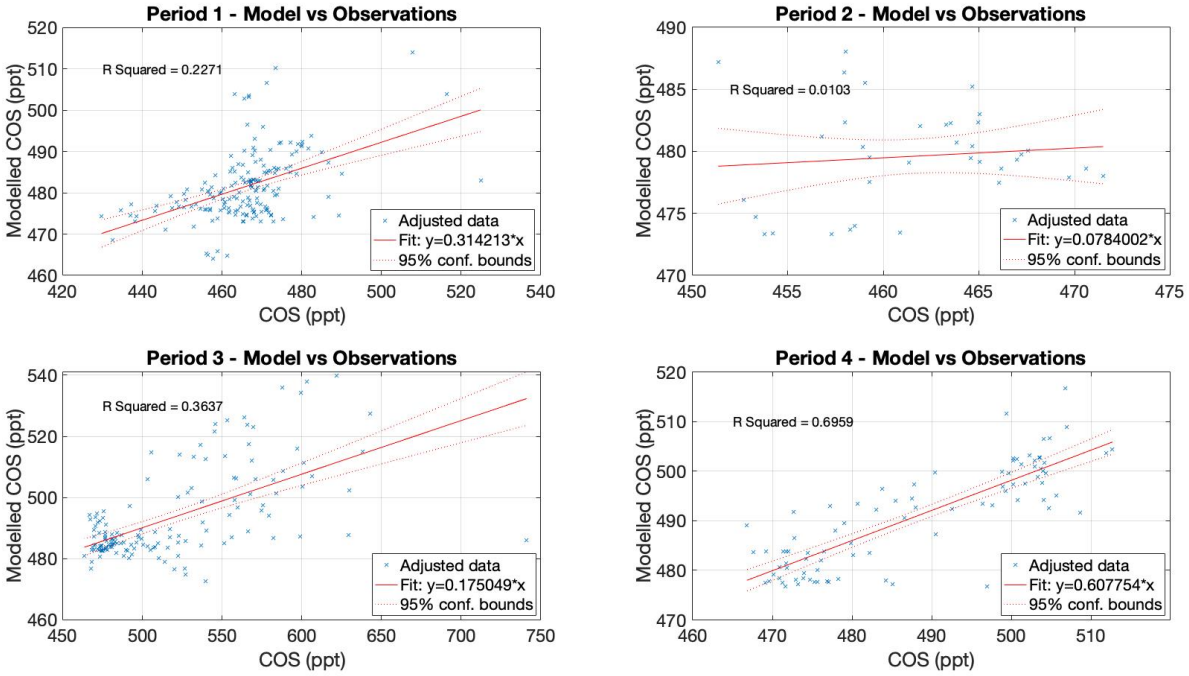

Figure S4: relationships between modelled COS results and observations for the selected time periods, where the only significant relationship is found for Period 4, identifying the source of these enhancements in the Ruhr region (Germany).

Table S1: means ± standard deviations of gas species concentration for the samples collected in the Eemshaven area.

Sample origin	**COS (ppt)**	**CH_4 (ppb)**	**CO_2 (ppm)**	**CO (ppb)**	**N_2O (ppb)**
Sludge ponds	461 ± 21	2013.27± 0.79	411.74 ± 0.25	129.57 ± 0.47	333.94 ± 0.14
Sludge ponds	448 ± 13	2018.85± 0.35	411.58 ± 0.10	130.52 ± 0.59	333.91 ± 0.16
Coal storage	448 ± 17	2138.20± 0.49	409.53 ± 0.17	136.09 ± 0.51	333.93 ± 0.06
Coal storage	437 ± 13	2236.51± 0.37	472.96 ± 0.24	136.57 ± 0.88	333.95 ± 0.08
Wastewater	439 ± 9	2009.62± 0.71	406.34 ± 0.09	133.35 ± 0.46	333.71 ± 0.14
Wastewater	419 ± 6	2003.21± 0.19	405.14 ± 0.07	135.01 ± 0.33	333.61 ± 0.06
Background	424 ± 4	2008.63± 0.21	406.84 ± 0.08	136.62 ± 0.52	333.72 ± 0.07
Background	426 ± 8	2006.95± 0.33	407.38 ± 0.07	132.34 ± 0.52	333.55 ± 0.11

Table S2: means ± standard deviations of gas species concentration for the samples collected at the ATTERO facilities (Groningen).

Sample origin	**COS (ppt)**	**CH_4 (ppb)**	**CO_2 (ppm)**	**CO (ppb)**	**N_2O (ppb)**
Waste loading	534 ± 2	5396.52± 0.70	440.89 ± 0.09	136.38 ± 0.14	344.59 ± 0.05
Waste loading	473 ± 6	5659.73± 0.66	420.94 ± 0.17	139.01 ± 0.36	337.49 ± 0.08
Biodigesters	427 ± 9	3420.27± 1.34	433.47 ± 0.10	276.26 ± 0.14	334.12 ± 0.10
Biodigesters	429 ± 6	2494.50± 0.47	403.66 ± 0.54	163.59 ± 0.21	333.97 ± 0.12
Gas processing	425 ± 11	2636.08± 0.67	446.20 ± 0.14	141.62 ± 0.48	334.97 ± 0.16
Gas processing	435 ± 10	2321.13± 0.33	424.87 ± 0.16	135.54 ± 0.38	335.17 ± 0.13
Background	407 ± 21	2004.57± 0.57	403.27 ± 0.19	146.38 ± 0.49	334.57 ± 0.11
Background	413 ± 4	2008.74± 0.85	403.52 ± 0.18	150.21 ± 0.41	334.71 ± 0.12