



A ground-based spectrometer network to estimate net carbon dioxide and methane emissions from the City of Edinburgh

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Abstract

Progress towards net-zero targets depends on independent verification of urban greenhouse gas emissions. Cities account for
~75% of energy-related CO₂ emissions, yet their quantification remains challenging due to spatially complex sources,
20 heterogeneous meteorology, and limited observation coverage. We deploy seven EM27/SUN ground-based Fourier-transform
spectrometers, within weatherproof enclosures, across the City of Edinburgh to measure column-integrated carbon dioxide,
methane and carbon monoxide (XCO₂, XCH₄, XCO), which are less sensitive than *in situ* sensors to highly localised surface
signals. A comprehensive calibration and quality-control framework leads to inter-instrument precision of 0.26–0.62 ppb for
XCH₄ and 0.05–0.07 ppm for XCO₂. The influence of the weatherproof enclosure dome on XCO₂ and XCH₄ retrievals is
25 negligible but introduces a correctable XCO bias. City-wide observations reveal clear seasonal cycles and detectable intra-city
gradients, with XCH₄ showing consistent upwind-downwind behaviour, while XCO₂ and XCO are strongly influenced by
background variability and atmospheric transport. Comparison with the ICON-ART mesoscale model (6.5 km resolution)
shows that total column variability is dominated by background conditions, with anthropogenic contributions typically < 0.2–
0.3 ppm. These signals exceed model minus observation differences in only ~5% of cases, highlighting the challenge of
30 isolating urban emissions from column measurements. Overall, our results demonstrate that dense column networks provide
robust, scalable constraints on urban greenhouse gas emission estimates and valuable validation for atmospheric transport
models and satellite retrievals, forming a foundation for inverse modelling with the UK-wide network of similar instruments.



1. Introduction

Cities are responsible for a substantial and rapidly growing fraction of global anthropogenic greenhouse gas (GHG) emissions, particularly carbon dioxide (CO₂) and methane (CH₄) (Seto et al., 2014). Accurate quantification of urban GHG fluxes is critical for verifying emissions inventories, supporting national and regional net-zero targets, and evaluating the efficacy of climate mitigation policies. However, uncertainties in current emission inventories remain significant, particularly at the city scale, due to limitations in reporting, spatial resolution, and activity-based accounting (Gurney et al., 2021; Super et al., 2020; Solazzo et al., 2021; Zhao et al., 2017).

In UK urban areas, the principal sources of CO₂ emissions are domestic and commercial energy consumption (including heating), road transport, and industrial activity, with additional contributions from waste processing and peri-urban agriculture (Kurganskiy et al., 2025; Palmer et al., 2018). Urban CH₄ emissions are largely from waste management (e.g. landfill and wastewater), losses from the natural gas distribution network, and in some cases, emissions from livestock and agriculture at the city fringe (IPCC, 2022; Hmiel et al., 2020; Peischl et al., 2018). Carbon monoxide is primarily emitted from incomplete combustion processes, particularly traffic and urban heating systems. City-scale emissions are typically reported in terms of total greenhouse gas emissions (CO₂-equivalent, CO₂-eq), with CO₂ dominating the overall budget. In this study, we focus on the City of Edinburgh, a mid-sized mid-latitude city in the UK (area of ~263 km²; population of ~530,680 in 2024 (Office for National Statistics, 2025)) with a diverse mix of residential, commercial, and transport-related emissions. According to data reported by the Department for Energy Security and Net Zero (DESNZ) (<https://www.edinburgh.gov.uk/climate-2/track-progress-net-zero>), total city greenhouse gas emissions decreased from approximately 2.56 MtCO₂-eq in 2017 to 2.19 MtCO₂-eq in 2022, with a notable reduction during the COVID-19 lockdown period (City of Edinburgh Council, 2025). Stationary energy use in buildings remains the largest contributor (~1.29 MtCO₂-eq in 2022), followed by transport (~0.69 MtCO₂-eq). Waste, wastewater, agriculture, and land-use emissions are smaller but non-negligible components of the city's GHG profile. Despite the overall downward trend, year to year changes, particularly in transport and waste, remain substantial. Edinburgh has set a target of achieving net-zero emissions by 2030, underscoring the need for improved observational constraints on urban greenhouse gases and independent evaluation of reported emission trends.

A range of observational techniques has been used to constrain urban GHG budgets. Ground-based *in-situ* sensors as instrument networks (e.g., Lauvaux et al., 2016; Shusterman et al., 2016; Davis et al., 2017; Sargent et al., 2018; Grange et al., 2025; Creman et al., 2025; Aigner et al., 2026) or eddy covariance towers (e.g., Ward et al., 2013; Stagakis et al., 2023) offer high temporal resolution and sensitivity near the surface and emission sources, but have limited spatial footprints. Remote sensing of trace gases through the total atmospheric column is an alternative observation method with greater spatial coverage. Satellite-based spectrometers retrieve city-wide snapshots of trace gas concentrations by measuring atmospheric absorption of sunlight, enabling the estimation of dry-air mole fractions for CO₂ (XCO₂) and other species. Such Earth Observation (EO)



data have identified urban GHG plumes and supported top-down emission estimates (Nassar et al., 2017; Reuter et al., 2014; Kuhlmann et al., 2019; Wu et al., 2023), though spatial resolution is often coarse relative to urban heterogeneity.

Ground-based total column measurements, using sun-tracking Fourier Transform Infrared (FTIR) spectrometers, extend this
 70 EO methodology at the Earth's surface. Networks such as the Total Carbon Column Observing Network (TCCON) and the
 Collaborative Carbon Column Observing Network (COCCON) employ this approach, providing high-quality, stable reference
 data and supporting the validation of satellites. COCCON's use of portable and relatively inexpensive EM27/SUN
 spectrometers (Bruker GmbH) has enabled multi-instrument studies across finer spatial scales at sub-urban and EO sub-pixel
 resolution. Using multiple EM27/SUN to improve spatial coverage, studies have detected intra-urban XCO₂ gradients (Hase
 75 et al., 2015; Zhao et al., 2019) used to infer urban CO₂ emissions (Chen et al., 2016; Vogel et al., 2019) and evaluate EO
 retrievals (Reuter et al., 2014). These studies are generally based on short-term campaigns and datasets, with the exception of
 the permanent Munich Urban Carbon Column network established in 2019 (MUCCnet, Dietrich et al., 2021).

In the UK, observational constraint of urban GHG emissions has primarily relied on the national-scale tall tower network
 80 (Deriving Emissions linked to Climate Change, DECC (Stanley et al., 2018) which provides robust regional constraints but
 limited intra-city detail. Recently, the Greenhouse gas Emission Monitoring network to Inform Net-zero Initiatives UK
 measurement network (GEMINI-UK) has established a nationwide network of ten EM27/SUNs across the UK (Kurganskiy et
 al., 2025), enabling new opportunities for urban-scale GHG assessment. The ability of the network to constrain urban GHG
 fluxes depends on how well the retrieved total column variability can be interpreted in terms of atmospheric transport, source
 85 contributions, and inter-instrument precision.

Interpreting total-column observations requires careful separation of local enhancements from background variability (Che et
 al., 2024). For long-lived species, background concentrations, driven by large-scale atmospheric transport, constitute a
 substantial fraction of the observed signal and can mask local emissions. CO₂ interpretation is further complicated by
 90 biospheric sinks, while methane and CO exhibit additional variability from atmospheric chemistry. These challenges
 necessitate multi-station observations to disentangle background, biogenic, anthropogenic, and chemical influences.

To address the need for finer-scale, continuous GHG monitoring within cities, we have established a new permanent GEMINI-
 Edinburgh EM27/SUN network for Edinburgh, designed to provide high-quality total-column trace-gas observations at
 95 intra-regional and intra-urban scales. The network operates under challenging conditions for solar absorption spectrometers
 including a relatively high latitude location (~59° North) with large solar-zenith angles, limited winter sun hours, and modest
 emissions magnitudes. A central component of this work is the procurement and evaluation of fully sealed weatherproof
 enclosures (Karn Scientific Ltd) that enable reliable 24/7 operation across seven instruments. We also develop and rigorously
 test calibration and quality control procedures tailored to enclosed EM27/SUN systems, ensuring inter-instrument consistency



100 suitable for urban-scale analysis. Using these calibrated observations, we characterise initial intra-urban and intra-regional differences in total-column CO₂, methane, and CO, and assess the ability of the mesoscale atmospheric model ICON-ART to reproduce observed day-to-day variability in XCO₂. Forward simulations of separately modelled XCO₂ components – background, anthropogenic, and biogenic – are used to interpret inter-station variability and evaluate the relative influence of transport, boundary conditions, and local emissions. Together, these developments provide the foundation for future
105 flux-inversion studies in Edinburgh, support the city’s net-zero ambitions, and contribute to the broader advancement of urban GHG monitoring methodologies in the UK.

In Section 2 we introduce the instruments we use, the network layout and deployment strategy. We describe the data processing workflow, including multi-level data products, automated quality control procedures, and the approach we adopt for
110 inter-instrument calibration and precision assessment. We also describe the ICON-ART modelling framework, including assumed emission inventories, how we sample the model to compare against the EM27/SUN spectrometer data, and the approach we use to identify the anthropogenic contribution to XCO₂. In Section 3, we evaluate our quality control approach, including an assessment of the resulting data availability and of the inter-instrument precision. We also document and assess the performance of the EM27/SUN weatherproof measurement enclosures and report the first results on intra-city variation in
115 total-column trace-gas concentrations. Finally, we conclude the study in Section 4.

2. Data and Methods

Here we outline the *in situ* and remotely sensed instruments used within the GEMINI-Edinburgh network. We describe the associated data-processing pipelines, including the retrieval of CO₂ and CH₄ concentrations, quality control procedures, the weatherproof enclosures used to house the spectrometers, and the automatic weather stations that provide ancillary
120 meteorological information. We also describe the regional atmospheric transport model used to interpret an exemplar subset of the CO₂ observations, including the model–data comparison statistics used to quantify model performance.

2.1. Instrumentation

2.1.1. EM27/SUN spectrometers

At the core of GEMINI-Edinburgh, we have deployed seven Bruker EM27/SUN Fourier Transform Infrared Spectrometers
125 (Appendix A, Table A1 for instrument summary) to collect data from which we retrieve atmospheric column-averaged dry air mole fractions of carbon dioxide (XCO₂), methane (XCH₄), carbon monoxide (XCO) and water vapour (XH₂O). These instruments record interferograms of solar absorption spectra from direct normal spectral irradiance in the range 4,000–12,000 cm⁻¹ (spectral resolution 0.5 cm⁻¹). The direct normal irradiance is tracked to the instrument detector by a 2-axis motorised mirror assembly. A camera evaluates the solar image on the detector to ensure the tracker correctly follows the sun.

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Reliable long-term measurements require robust environmental protection for the EM27/SUN spectrometers. Unlike previous deployments that relied on motorised moving shelters (e.g. Heinle and Chen, 2018) or short-term unprotected observations (e.g. Vogel et al., 2019), each EM27/SUN in this study is housed in a fully sealed, weatherproof enclosure designed and manufactured by Karn Scientific Ltd (Figure 1). The instrument observes the sun through a 5 mm-thick BK7 glass dome integrated with a custom spherical cap shell, which is securely sealed to a 1780 Protector Case (Peli Products UK). BK7 is selected for its high optical transmission from visible to ~2000 nm wavelength range (Figure 1c), low levels of defects and impurities, and strong durability and scratch resistance. While this enclosure has been tested and deployed previously (e.g. SN127, Table A1), this study represents the first systematic quantification of its performance and suitability for long-term operation.

The enclosure houses an industrial personal computer (PC); DDROBOT SEN0438 (or Bosch BME28) temperature, humidity, (and pressure) sensor; two 30W Peltier coolers; a 50 W heater; desiccant bags; an Ardbox 20 PLC Arduino; and all associated AC-DC electronics including an uninterruptable power supply. The enclosures are programmed to maintain internal temperatures between 10 and 50 °C and are externally covered with reflective aluminium tape to reduce solar heating (Figure 1b).

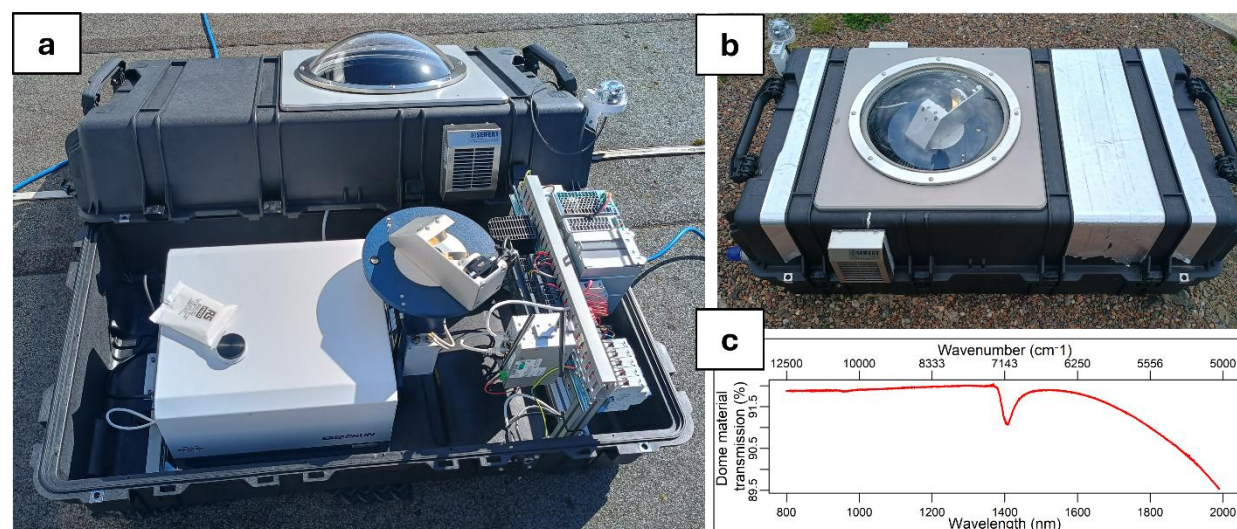


Figure 1. EM27/SUN enclosure from Karn Scientific (a) uncovered to show EM27/SUN and components and (b) covered and with reflective aluminium tape with (c) manufacturer-derived BK7 glass dome transmission spectra.

The Windows PC (Microsoft Windows 10 Long-Term Servicing Channel Operating System) runs measurement capture and control software (Appendix E) including OPUS (Bruker GmbH) to capture interferograms with a 10 kHz scanner velocity and 4 scans per sample (~25 s interferogram sample rate, UTC time) and Pyra (Aigner et al., 2023) to ensure interferograms are

measured when the sun is 2 degrees above the horizon. The Arduino measures data from the enclosure environmental sensor and publishes them to the PC by serial interface.

2.1.2. Automatic weather stations (AWS)

Vaisala WXT536 (Vaisala Oyj) and Davis Vantage Pro 2 (Davis Instruments) AWS measure standard instantaneous meteorological variables every 10 s and 60 s respectively (UTC time), using Raspberry Pi 3 (Raspberry Pi Foundation) data loggers. Data in hourly text files are transferred to remote server by SFTP. Each AWS is installed on the roof of a building on a 2 m mast to best capture representative surface wind direction ($\pm 3.0^\circ$ at 10 m/s) for wind sector analysis of intra-city trace gas concentrations. Atmospheric pressure from the WXT536 (± 0.5 hPa at 0 to $+30^\circ\text{C}$) is used in the EM27/SUN retrieval algorithm.

2.2. Measurement network layout and instrument deployment details

To capture urban GHG signals, the measurement network is designed to capture upwind, city centre, and downwind GHG concentrations by encircling the city with EM27/SUNs. To achieve this, a long-term operational measurement network was configured comprising one city-centre EM27/SUN and four additional EM27/SUNs surrounding the city.

All deployments are within The City of Edinburgh (55.95°N , 3.19°W), bounded by the Firth of Forth to the north and the Pentland Hills to the south, which rise to 579 m above sea level (asl, Figure 2). The surrounding rural areas are predominantly agricultural land and grassland. The five EM27/SUN measurement stations (Table 1), separated by approximately 5–7 km, have been commissioned around the (sub)urban perimeter of Edinburgh (Figure 2). Clockwise from north, the stations are EDLDST on the north coast of the Firth of Forth; EDQMUM to the east of the city; EDJCMB to the south; and EDNARO to the west-southwest, with a final station in the city centre (EDCHRY). EDJCMB additionally serves as the testing and intercomparison station to characterise inter-instrument precision and stability through time.

Deployment and maintenance information are recorded in a dedicated metadata spreadsheet, including instrument latitude, longitude, height asl, start and end dates, and relevant operational comments.

Table 1. Measurement stations for EM27/SUN spectrometers (Table A1 for details of each EM27/SUN) and automatic weather station (AWS). Height above sea level based on WGS84 EGM96 Geoid determined using Google Earth Pro v7.3.6.9796. All stations are ongoing as of 4thth July 2026.

Station description	Station code	Lat ($^\circ\text{N}$), Lon ($^\circ\text{E}$)	Height asl (m)	Deployed instrument	Serial number (SN)	Deployment date (DD/MM/20YY)
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James Clerk Maxwell Building, University of Edinburgh (UoE)	EDJCMB	55.921, 3.174	- 95	EM27/SUN	All during 14/10/24 intercomparison period or multiple with retrievals merged as “SNmerged”.
	EDJCMB	55.921, 3.174	- 100.3	WXT536	1540027 03/03/23
Queen Margaret University, Musselburgh	EDQMUM	55.931, 3.072	- 32	EM27/SUN	SN217 06/06/25
Port of Leith distillery	EDLDST	55.983, 3.175	- 40	EM27/SUN	SN216 02/06/25
National Robotarium, Heriot Watt University	EDNARO	55.913, 3.325	- 80	EM27/SUN	SN215 08/04/25
Chrystall Macmillan building, UoE	EDCHRY	55.944, 3.191	- 107	EM27/SUN	SN127 16/05/25
Appleton Tower, UoE	EDAPLT	55.944, 3.187	- 178	WXT536	S0230710 21/05/25
Edinburgh Castle	EDCAST	55.948, 3.199	- 148	Davis Vantage Pro 2	Unknown 07/05/25

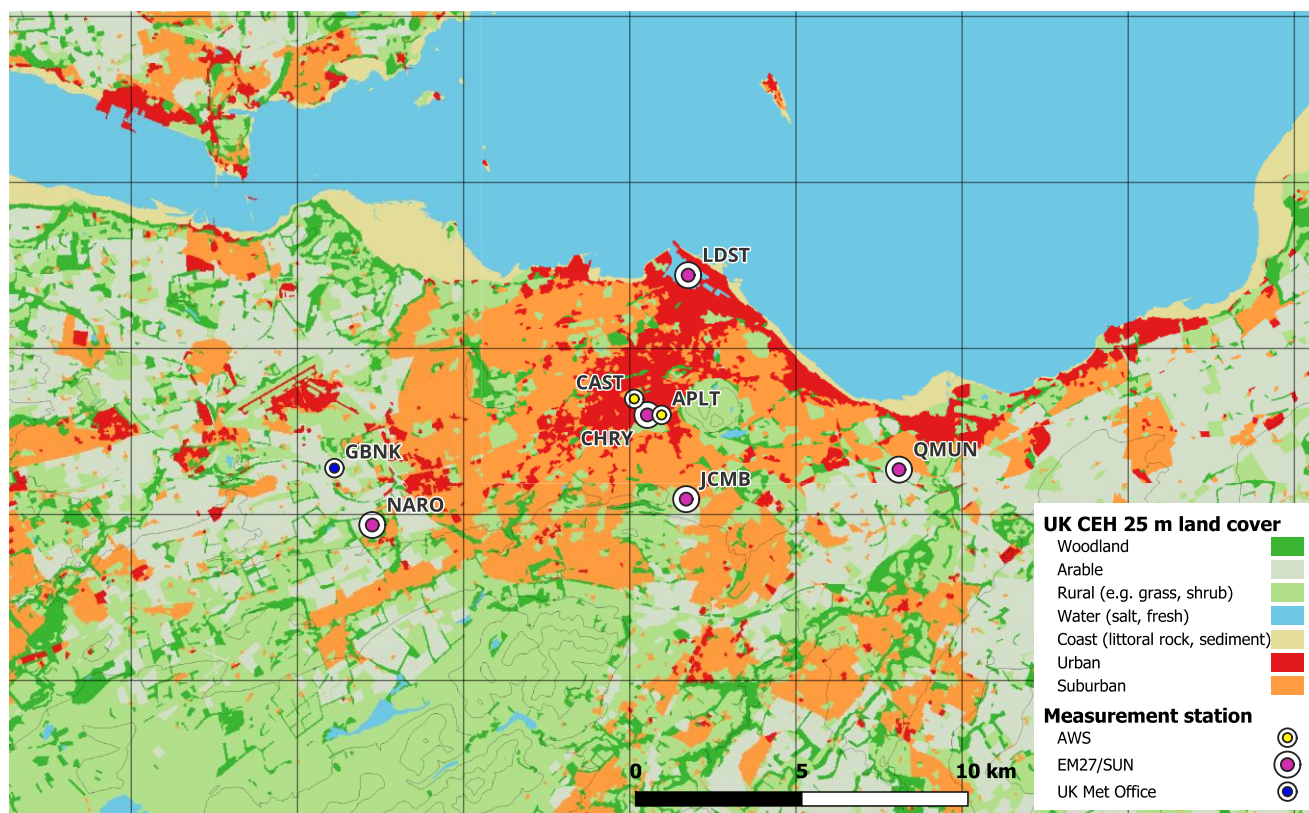


Figure 2. Distribution of measurement stations around The City of Edinburgh with (points and labels) the location and names of measurement stations across (map colours) different land cover types (Morton et al., 2024) with (black lines) 5km grid cf. Section 2.3.4 modelling horizontal resolutions 1) ERA5 model of meteorological initial and boundary conditions: 0.25° (~25 – ~28 km) 2) ICON-ART atmospheric transport model: 6.5 km triangular icosahedral 3) TNO-GHGco 2021 anthropogenic emissions inventory: ~ 5 km. The station name “ED” prefix is excluded from labels.

Operational measurements began on 8th Apr 2025 with EM27/SUN serial number (SN)215 at EDNARO (Table 1). The remaining AWS and EM27/SUN instruments were deployed across the city by 2nd June 2025 (Table 1, Figure 2), with SN218 and SN219 remaining at EDJCMB. SN219 is the long-term EDJCMB static instrument. SN218 is used to continually assess the long-term inter-instrument precision of SN219 and SN218 through time, and available as a spare and for any future mobile intercomparisons. In addition to the EDJCMB AWS, there are AWS stations deployed on tall buildings in the city centre (EDCHRY, EDCAST, Figure 2, Table 1) to capture representative wind flow above the urban canopy.

2.3. Data collection and processing pipeline

The EM27/SUN raw interferograms have processing applied at three levels.

- Level 0 (L0) data are the initial trace gas retrievals without filtering or calibration.
- L1 is derived from L0 data following the application of an automated QC procedure to flag or reject suspect or erroneous measurements.



- L2 is the final, empirically calibrated data product using a reference EM27/SUN and regression-based calibration to minimise inter-EM27/SUN retrieval differences.

2.3.1. Level 0 (L0): retrieved total column trace gases

Total column trace gas retrievals use the Collaborative Carbon Column Observing Network (COCCON) PROFFAST algorithm (Alberti et al., 2022; Frey et al., 2019) version 2.4.1 with the python wrapper PROFFASTpylot version 1.3 (Feld et al., 2024). PROFFAST performs least-squares fitting of the spectra to a priori trace gas and meteorology profiles (Alberti et al., 2022; Frey et al., 2019; Sha et al., 2020). PROFFASTpylot is run for each day of measurements for each EM27/SUN using default settings, published instrument line shape (ILS) parameters (Feld, 2025), atmospheric pressure data at the EM27/SUN measurement altitude, and Total Carbon Column Observing Network (TCCON) GGG2020 meteorology model (.mod) and prior (.map) a priori files (Laughner et al., 2024).

Atmospheric pressure data are scaled from AWS measurements at EDJCMB to the altitude of each EM27/SUN using the hypsometric equation (Stull, 2018):

$$P_1 = P_{\text{JCMB}} \cdot \exp\left(\frac{z_{\text{JCMB}} - z_1}{a \cdot T_v}\right), \text{ (Eqn. 1)}$$

with P_{JCMB} and z_{JCMB} the EDJCMB station pressure (hPa) and height (m), P_1 (z_1) the station pressure (height) at an arbitrary EM27/SUN, a the gas constant for dry air (29.3 m K^{-1}) and T_v the mean virtual temperature (K) between z_1 and z_{JCMB} assuming a standard atmosphere lapse rate (0.0065 K m^{-1}). P_1 are provided to PROFFASTpylot as 10-minute averages. The daily trace gas timeseries .csv files output by PROFFASTpylot are the L0 data.

2.3.2. Level 1 (L1): quality controlled total column retrievals

L0 data at the ~25 s sample rate, Section 2.1.1 are quality controlled to a L1 product which flags a range of suspect and erroneous retrievals. L1 daily data files are generated from the following L1 QC steps (Table 2):

- Load L0 data from PROFFASTpylot output .csv files on a per-EM27/SUN and per-day basis.
- Reject outlier values in retrieved XCO_2 , XCH_4 , XCO , and XH_2O based on physically reasonable thresholds.
- Reject all trace gases when diagnostic parameter X_{AIR} is out of range, as X_{AIR} should be stable and close to 1 (Humpage et al., 2024).
- Flag data when $\text{SZA} > 80^\circ$ where PROFFAST retrievals become less reliable (Herkommer et al., 2024).
- Reject all trace gases based on a moving window X_{AIR} noise threshold. X_{AIR} should be relatively constant through time, with spikes indicating misalignment, transient cloud, or other issue (Herkommer et al., 2024; Humpage et al., 2024).



- Flag trace gases as suspect if the solar tracker becomes misaligned according to the newly implemented PROFFAST (version 2.4.1) doppler shift variables. Any misalignment between the EM27/SUN's line of sight and the centre of the sun will induce an apparent Doppler shift in the measured atmospheric absorption lines (Mostafavi Pak et al., 2023).
- Flag each trace gas as suspect if there are low remaining data within a 10-minute moving time window, as sparse remaining data may indicate unresolved issues.
- Write daily, per-EM27/SUN NetCDF files with all PROFFASTpylot variables and all QC flags.

Table 2. Quality control steps and associated flags assigned to the retrieved trace gases through time

Flag Name	QC Filter/Condition	Data Operation When Flagged
Physically reasonable thresholds		
flag_co2_threshold	XCO ₂ outside 350 ppm to 550 ppm range	Reject XCO ₂
flag_ch4_threshold	XCH ₄ outside 1 to 10 ppm range	Reject XCH ₄
flag_co_threshold	XCO outside 0.01 to 0.5 ppm range	Reject XCO
flag_xh2o_threshold	XH ₂ O outside 0 to 10000 range	Reject XH ₂ O
Generic retrieval diagnostics thresholds		
flag_sza_threshold_warn	Solar zenith angle >80° (Herkommer et al., 2024)	Flag only
flag_xair_noise_threshold	X _{AIR} is an outlier within its moving time window (mean ± 2σ)	Reject XCO ₂ , XCH ₄ , XCO, and XH ₂ O
flag_xair_threshold	X _{AIR} outside 0.99 to 1.0075 range	Reject XCO ₂ , XCH ₄ , XCO, and XH ₂ O
Low data thresholds		
flag_xco2_lowdata	< 2 non-missing XCO ₂ in 10 min window	Flag only
flag_xch4_lowdata	< 2 non-missing XCH ₄ in 10 min window	Flag only
flag_xco_lowdata	< 2 non-missing XCO in 10 min window	Flag only
flag_xh2o_lowdata	< 2 non-missing XH ₂ O in 10 min window	Flag only
flag_any_lowdata	Any of the per-gas low data flags are true	Flag only
Misalignment quality control		
flag_suntracker_sol_threshold_warn	Calculate linear regression between SZA and each PROFFAST doppler shift “sol” variable (job02_sol, job03_sol, job04_sol)	Flag only



over a 7-day window. For each time point, the residual (difference between observed and predicted value) is computed. Flag when the residual $> 2.5\sigma$ for any sol variable.

As the QC only uses L0 data, it is highly transferrable to other systems. Supplementary datasets from e.g. AWS or CamTracker systems are not used as their availability and reliability are inconsistent and may not transfer to other measurement networks. Moreover, independent CamTracker alignment diagnostics were found to often produce false positives, particularly under high camera gain settings or sporadic cloud cover. Here CamTracker images were used to manually identify cases of misalignment during validation of the automated approach.

2.3.3. Level 2 (L2): calibrated and aggregated retrievals

L1 retrievals (Section 2.3.2) are aggregated to 5-minute mean values and a multi-linear regression model determines empirical correction factors (β) against a reference EM27/SUN (SN219):

$$XGAS_{SN219} = \beta_1 XGAS_{SNXXX} + \beta_2 \theta + \varepsilon, \text{ (Eqn. 2)}$$

with $XGAS_{SN219}$ the trace gas retrieval (e.g. XCO_2) from SN219, X_{SNXXX} the corresponding retrieval from an arbitrary comparison EM27/SUN, β_1 the slope and ε the residual error term. In agreement with previous work, some instruments have a clear SZA dependence for XCO retrievals (Herkommer et al., 2024) that may be attributed to ILS uncertainty (Alberti et al., 2022). In these cases, the correction factor β_2 is applied, with θ the SZA at the given measurement time and is based on manual comparison of both correction approaches across a multi-week period of data.

The derived coefficients are then applied to all L1 retrievals for the given EM27/SUN, and the corrected retrievals are aggregated to 5-minute means to give the final L2 product. All L1 QC flags (Section 2.3.2) are carried over to L2. Any 5-minute average is flagged if it contains at least one flagged retrieval.

The main intercomparison period for determination of correction factors across all enclosed EM27/SUNs was 26th March 2025 to 15th May 2025. SN219 was selected as the reference instrument as initial L0 retrievals were qualitatively found to be most representative of the average across all EM27/SUN retrievals. Additional to this intercomparison period at EDJCMB was the deployment of an enclosed EM27/SUN (SN217) on 14th Oct 2024, marking the start of the long-term data record. Between 31st Jan and 29th May, enclosed SN216 was also deployed alongside SN217 as a long-term intercomparison. The WXT536 AWS at EDJCMB captured standard meteorological variables throughout. This calibration approach can easily be extended to include an absolute calibration to a reference EM27/SUN using e.g. a COCCON travelling standard (Alberti et al., 2022). The performance of each calibration was assessed using mean absolute error (MAE) and mean bias error (MBE).



265 To determine the impact of the enclosure's glass dome on the observations, SN219 and SN216 were deployed both with and without the glass dome for a selection of clear-sky days at EDJCMB between 14th November 2024 and 24th March 2025 and compared to the co-located and enclosed SN217.

2.3.4. ICON-ART atmospheric transport model

In this study, we use the mesoscale numerical weather prediction and atmospheric transport model ICON-ART (Zängl et al., 2015; Rieger et al., 2015; Schröter et al., 2018; Hoshyaripour et al., 2026) to provide an exemplar simulation for interpreting the observed XCO₂ signals across the GEMINI-Edinburgh network. The model provides the relationship between surface fluxes and atmospheric concentrations. ICON-ART has previously been applied to regional and urban-scale greenhouse gas inversion studies (Steiner et al., 2024a, b; Thanwerdas et al., 2025; Ponomarev et al., 2026). In this study, we use a limited-area configuration over a European domain at 6.5 km horizontal resolution (R3B8 grid), closely following the one described by Ponomarev et al. (2026). The domain covers central Europe, including Scotland. The simulations provide hourly output of meteorological variables and tracer mole fractions information on approximately 83,000 horizontal grid cells at 60 vertical levels. The vertical grid is non-uniform and based on the terrain-following Smooth LEvel Vertical (SLEVE) vertical coordinates. The lowest layer has a thickness of 20 m, and the layer spacing increases with height, extending up to 22 km.

280 Meteorological initial and boundary conditions were taken from the ERA5 reanalysis (Hersbach et al., 2020). To maintain large-scale atmospheric consistency throughout the simulation period, the model state variables were weakly nudged towards ERA5 fields. The simulations span 1 January to 17 June 2025, corresponding to the period of available input data. Initial and boundary conditions for CO₂ were prescribed from the CAMS global inversion-optimized product (Chevallier et al., 2010).

285 Anthropogenic CO₂ emissions were prescribed using the TNO-GHGco 2021 inventory at around 5 km spatial resolution (Super et al., 2020). Biospheric CO₂ fluxes were computed online during the simulation using the Vegetation Photosynthesis and Respiration Model (VPRM) (Mahadevan et al., 2008), driven by ICON-simulated shortwave radiation and near-surface temperature, in combination with MODIS-derived vegetation indices (EVI and LSWI) (Vermote, 2015). Land-cover information specifying vegetation types and their spatial distribution was based on the CORINE 2018 dataset (European Environment Agency, 2020).

295 We evaluate the performance of the ICON-ART simulations by sampling model output at each EM27/SUN station location (Table 1) and comparing against observed values. As JCMB hosts multiple EM27/SUNs, L2 JCMB observations were merged into a single time series (JCMB SNmerged), using SN219 as the reference instrument and gap-filling missing data with the mean of any other available co-located instruments. The analysis period spans 1st January to 17th June 2025, corresponding to the availability of reanalysis input data. For each hourly model output time, X, 5-minute L2 data were averaged over a 1-hour time window centred on X (± 30 min). Hourly model output was horizontally interpolated to each station location using inverse-



distance weighting of the five nearest grid cells, to account for the mismatch between the model grid resolution and the exact instrument locations. Model vertical profiles of CO₂ mole fraction (up to 20 km) were interpolated to the EM27/SUN retrieval grid to account for differences between the model vertical discretisation and the retrieval sensitivity described by the instrument averaging kernels. Daily mean prior profiles were used to calculate model-equivalent XCO₂. Details of XCO₂ model calculation, including the equation used to integrate interpolated mole fraction profiles, are provided in Appendix C.

As the model CO₂ vertical profile extent does not match the prior profile, values above the model's vertical domain were taken directly from the prior profile. The model carbon dioxide components forced by Anthropogenic emissions (Anthr.), vegetation Respiration fluxes (RA) and Gross Primary Production fluxes (GPP) did not have separate prior profiles. For these, values above the model top (20-70 km) were extrapolated linearly, reflecting that most of the fluxes occur within the first few kilometres above the surface and do not significantly contribute at higher altitudes.

We assess model performance using standard statistical metrics, including the mean bias, centred root-mean-square error (cRMSE), and the Pearson correlation coefficient (r). All statistics are computed using hourly averaged data to ensure the consistency between model output and observations. The cRMSE is computed after removal of the mean bias isolating differences in temporal variability between model and observations. The Pearson correlation coefficient is used to quantify the temporal co-variability between the two datasets. Additional quality control flags described in the observational data (Table 2) are used to remove potential outliers and assess the robustness of the comparison results. Metrics are calculated excluding flagged observations, hereafter referred to as `bias_flagged`, `cRMSE_flagged` and `Corr_flagged`.

We quantify the contribution of anthropogenic emissions to total columns of XCO₂ by using separately simulated model components. These contributions are represented in ICON-ART using tagged tracer components that isolate different sources while being transported by the same meteorological fields. Anthropogenic emissions are driven by surface emissions as represented in the inventory. The two biogenic tracers describe net ecosystem exchange, separated into RA and GPP. A background tracer (BG) is defined using only initial and temporally and spatially varying lateral boundary conditions, with no surface fluxes. It represents the advected large-scale CO₂ signal.

To provide a consistent metric across all stations and the analysis period, we compute the ratio of the modelled anthropogenic enhancement to the absolute model-observation difference. This ratio provides an indication of the extent to which the differences can be attributed to anthropogenic sources, as opposed to other factors such as background variability, biogenic activity or model transport errors.



3. Results

330 Here we evaluate the quality, consistency, and availability of the EM27/SUN observations across the network and examine their interpretation using the ICON-ART model. We assess the impact of quality control and calibration, analyse network-wide and intra-city variability, and investigate the contribution of anthropogenic emissions to observed XCO₂ signals.

3.1. L1 quality control

We evaluate the L1 quality control performance by comparing retrievals from collocated EM27/SUN instruments SN219 and
 335 SN204 at station EDJCMB between 7th April 2025 – 18th August 2025. Retrievals are compared under three QC scenarios: no quality control (L0, Figure 3, red), “standard” L1 QC excluding the newly introduced misalignment checks (Figure 3, green), and full L1 QC misalignment screening (Figure 3, blue). The intercomparison includes a range of observing periods, including clear, cloud, rain, to cover a range of representative and challenging measurement conditions. For XCO₂, the standard deviation (1σ) of SN219 minus SN204 differences decreases from 0.204 ppm at L0 to 0.168 ppm after applying the standard
 340 L1 QC, with approximately 5% of the retrievals rejected (Table 3). Including the misalignment QC further reduces 1σ to 0.104 ppm, rejecting an additional ~15% of retrievals. We find similar improvements for XCH₄ and XCO. This demonstrates that the misalignment QC step substantially improves inter-instrument precision beyond the standard L1 filtering. A timeseries example of QC is in Appendix B.

345 **Table 3.** Intercomparison statistics for EM27/SUN SN204 and SN219 between 17th April 2025 and 18th August 2025 using 5-minute median of trace gas retrievals at level 0 (L0) and L1 quality control stages, with 1σ the standard deviation of SN219 - SN204 differences, n the number of SN219 - SN204 sample points, and L1 (no misalignment QC) the L1 QC without the new misalignment quality control steps (Table 2).

	1σ (L0)	1σ (L1 no misalignment QC)	1σ (L1)	n (L0)	n (L1 no misalignment QC)	n (L1)
XCO ₂ (ppm)	0.204	0.168	0.104	3717	3538	3085
XCH ₄ (ppb)	0.748	0.602	0.506	3717	3538	3085
XCO (ppb)	1.448	1.291	0.570	3621	3475	3049

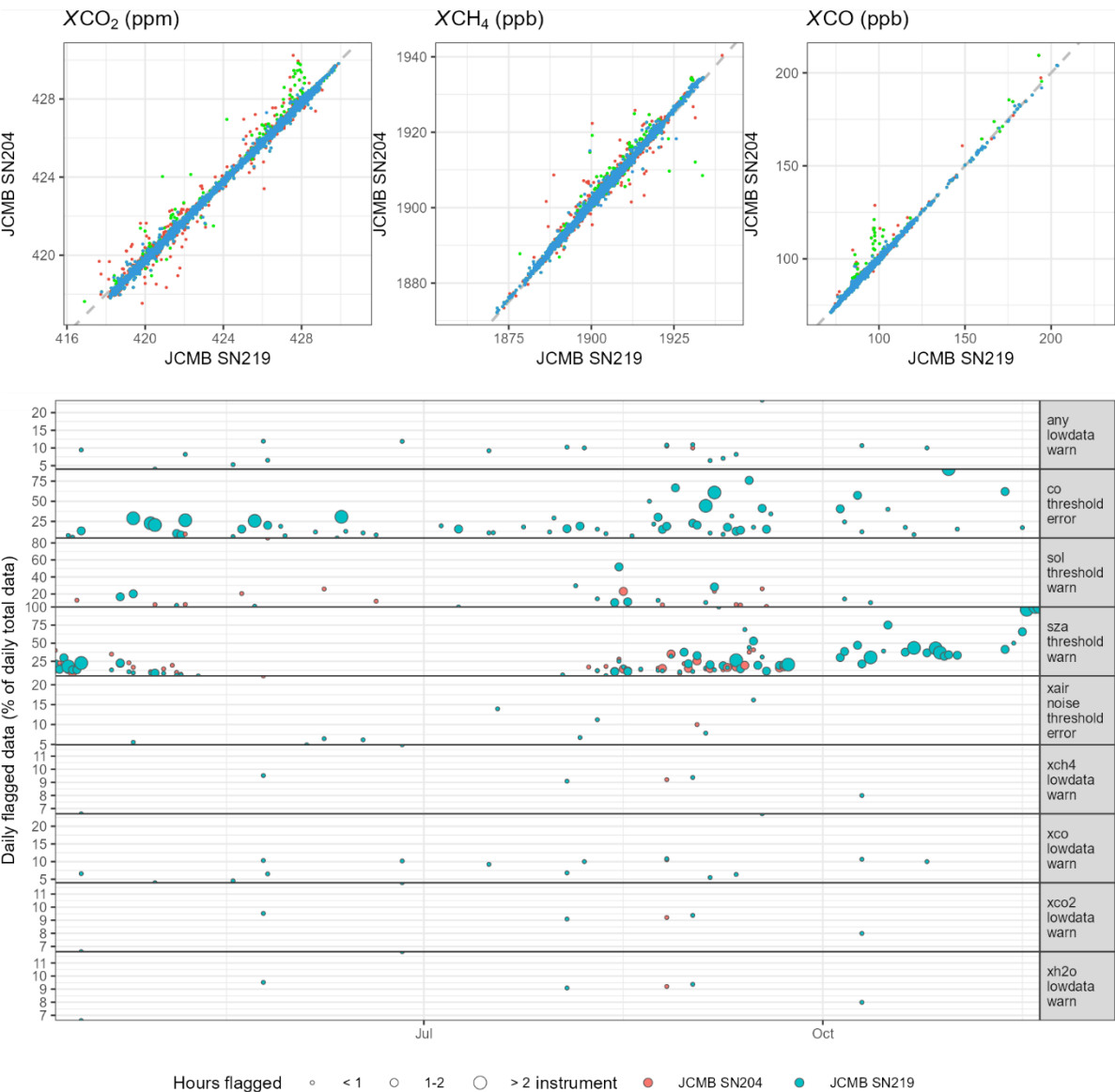


Figure 3. Evaluation of quality control (QC) steps for EM27/SUN retrievals using co-located SN204 and SN219 at station EDJCMB between 7th April 2025 and 18th August using 5-minute medians of (a) XCO₂ (b) XCH₄ and (c) XCO retrievals with (red) level 0 (L0) and no QC, (green) L1 QC but without the new misalignment QC steps, and (blue) all L1 QC.

3.2. Data availability

The availability of EM27/SUN L2 data during the ICON-ART analysis period (1 Jan – 17 June 2025) is shown in Figure 4. Daily data coverage varied across instruments and time, generally following the instrument deployment schedules (Table 1). Continuous measurements are available at JCMB. Data availability is strongly influenced by seasonal changes in SZA at Edinburgh’s northern latitude. During winter, retrievals are limited to typically fewer than < 5 h per day, increasing to ~10–15



h per day in summer. On 1st Jan 2025, SZA <80° for only 2.5 h (~11:00 – 13:30 UTC), otherwise retrievals are flagged as
 360 suspect (high SZA, flag_sza_threshold_warn, Section 2.3.2). Only after 10th Feb is SZA < 70°. Long summer days result in
 SZA < 80° conditions for over 14 h (~06:00 – ~20:20 UTC, 21st June) with 15.2 hours of retrievals at NARO on 11th June.
 QMUM has the fewest retrievals due to latest deployment (6th June, Table 1) and unexpected data collection issues.

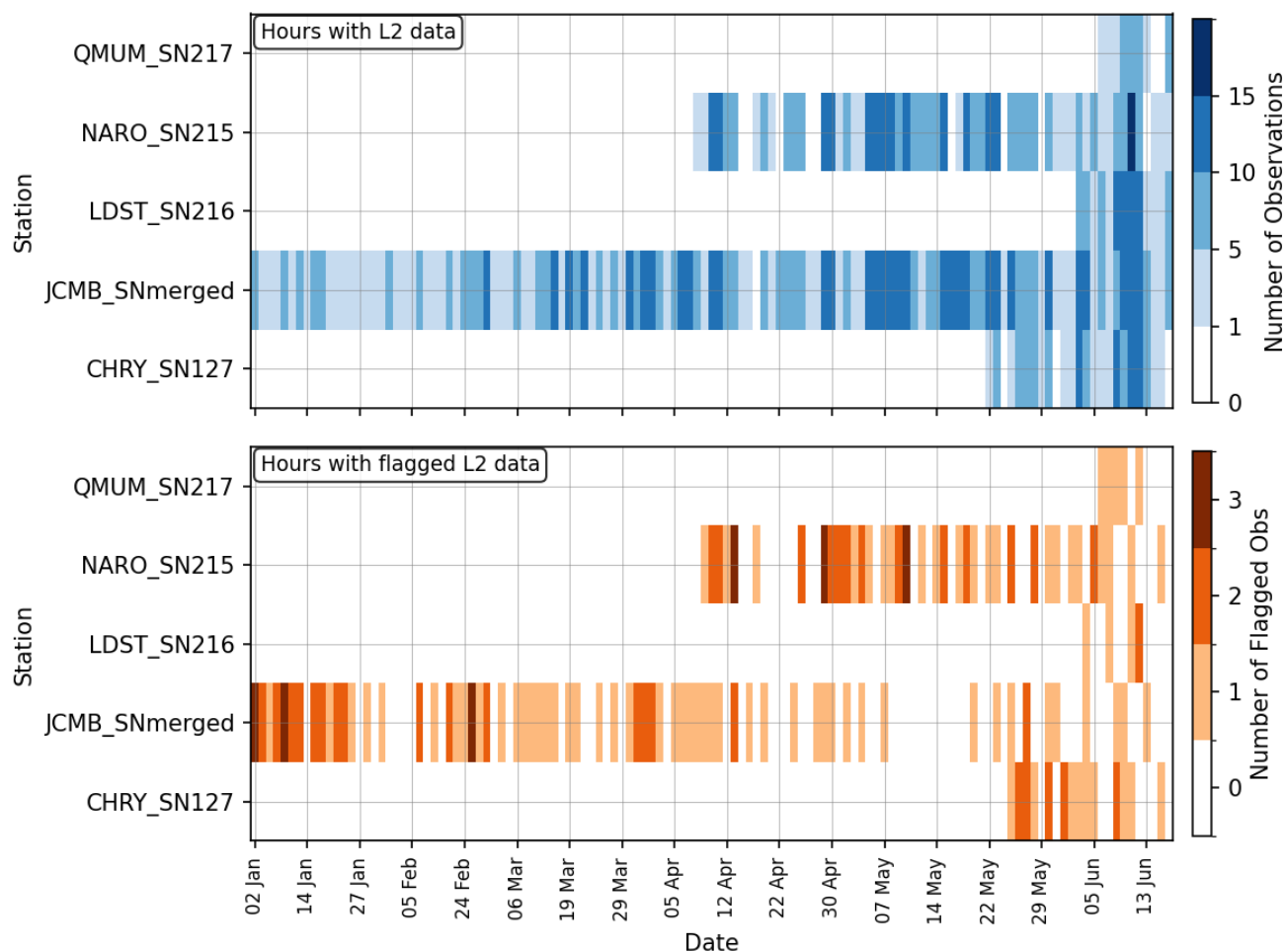


Figure 4. Daily coverage of EM27/SUN L2 retrievals aggregated as hourly averages from 1 January to 17 June 2025 for comparisons with
 365 ICON-ART model. Each bar represents one day of data. The top panel shows the total number of observations per day at each station, while
 the bottom panel shows the number of flagged observations.

3.3. L2 JCMB intercomparison and calibration coefficient generation

We use an all-instrument intercomparison period between 26 March and 15 May 2025 to evaluate calibration performance.
 Compared to L1 data (Figure 5, grey), L2-calibrated retrievals (Figure 5, black) show systematic improvements in MAE, MBE,
 370 and inter-instrument standard deviation for all trace gases (Table 4). Despite incomplete temporal overlap among instruments

due to deployment timing and tracker misalignment, each instrument contributes at least 647 five-minute means of co-located retrievals during a record high for spring sunshine and air temperature in Scotland (Kendon et al., 2026).

We find that L1 retrievals show an approximately linear relationship with the reference instrument SN219 (Figure 5, grey) for all species, except XCO from SN218 (MAE 13.69 ppb, Table 4), which exhibits a pronounced SZA dependence. SN215 exhibits a nuanced SZA dependence. This behaviour is corrected at L2 using an additional SZA-dependent calibration term. The older SN127 instrument performs comparably to newer units, despite earlier ILS characterisation and various prior deployments (Humpage et al., 2024), supporting the documented long-term stability of EM27/SUN spectrometers (Alberti et al., 2022). L2 XCO₂ precision ($1\sigma \approx 0.07\text{--}0.09$ ppm) agrees with previous EM27/SUN studies, as do XCH₄ and XCO results (Table 3, e.g. Chen et al., 2016; Frey et al., 2015; Vogel et al., 2019), except for SN218 XCO prior to calibration. Empirical calibration coefficients used to derive L2 products are provided in Table 5.

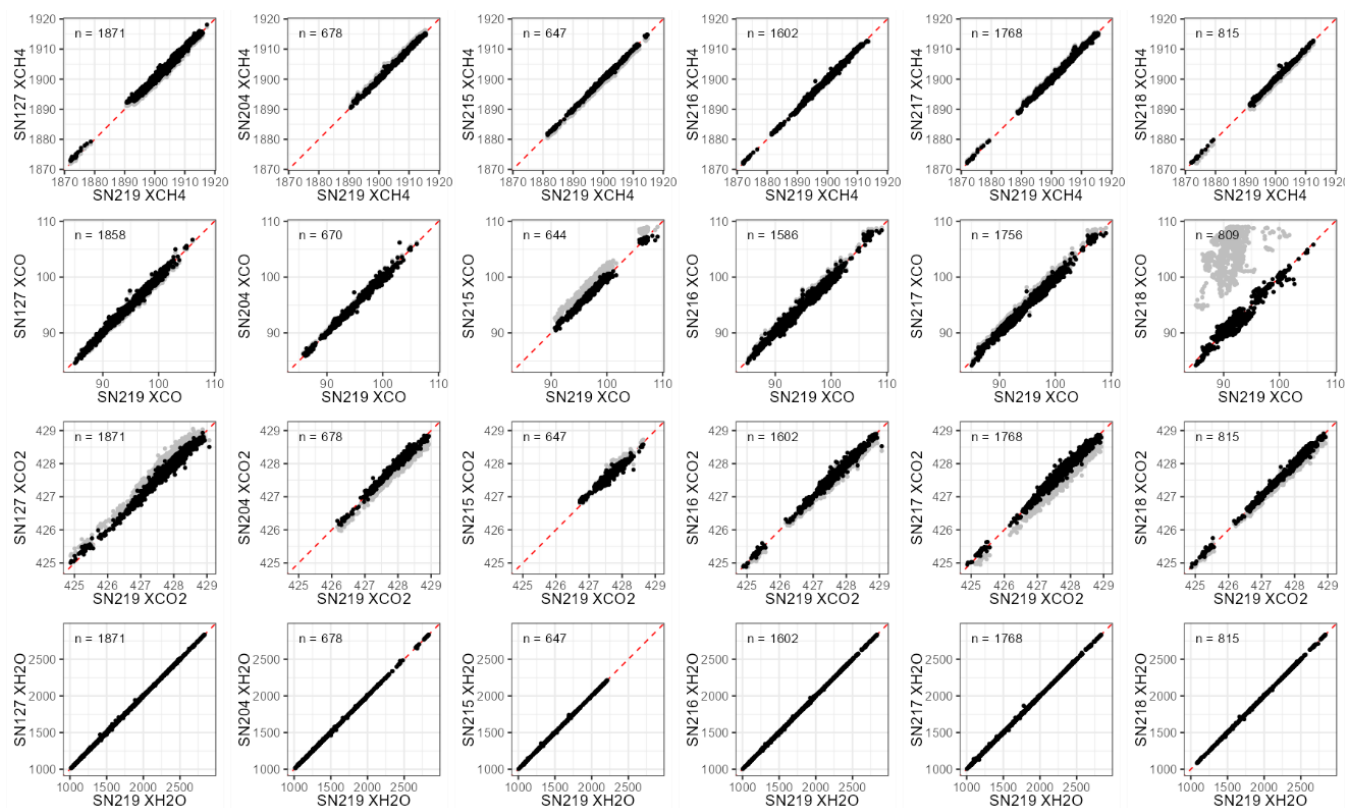


Figure 5. Intercomparison of retrievals from co-located EM27/SUN at station EDJCMB for 26th March 2025 to 15th May 2025 with (grey) level 1 (L1, uncalibrated) and (black) level 2 (L2, calibrated) data products, with (columns) the different EM27/SUNs, (rows) the different trace gases and (red dashed line) the 1:1 line. EM27/SUN with serial number 219 (SN219, x axis) is the reference EM27/SUN.



Table 4. Intercomparison statistics associated with data in Figure 5 with mean absolute error (MAE), mean bias error (MBE) as SN219 – SNXXX), the SN219 – SNXXX standard deviation of bias (1σ) and the number of samples (n).

EM27/SUN		XCH ₄ (L1)	XCH ₄ (L2)	XCO (L1)	XCO (L2)	XCO ₂ (L1)	XCO ₂ (L2)	XH ₂ O (L1)	XH ₂ O (L2)
SN127	MAE	0.991	0.619	0.410	0.325	0.209	0.074	2.976	2.683
	MBE	0.902	0.006	0.254	0.000	-0.206	-0.001	1.096	-0.018
	1σ	0.762	0.750	0.454	0.418	0.121	0.093	4.983	4.950
	n	1871	1871	1858	1858	1871	1871	1871	1871
SN204	MAE	0.908	0.328	0.362	0.328	0.212	0.060	3.316	3.293
	MBE	-0.902	0.022	-0.137	0.008	0.212	0.003	-0.456	-0.287
	1σ	0.458	0.427	0.454	0.442	0.078	0.075	5.730	5.726
	n	678	678	670	670	678	678	678	678
SN215	MAE	1.174	0.267	1.675	0.292	0.076	0.060	2.259	1.991
	MBE	1.174	0.010	-1.674	-0.001	-0.056	0.003	0.845	0.112
	1σ	0.345	0.343	0.521	0.370	0.079	0.079	4.290	4.268
	n	647	647	644	644	647	647	647	647
SN216	MAE	0.367	0.307	0.541	0.464	0.141	0.050	2.409	2.377
	MBE	0.223	0.021	-0.291	0.021	0.138	0.004	0.352	0.160
	1σ	0.407	0.406	0.610	0.596	0.070	0.068	4.365	4.364
	n	1602	1602	1586	1586	1602	1602	1602	1602
SN217	MAE	0.511	0.372	0.681	0.396	0.294	0.060	2.416	2.385
	MBE	-0.424	0.005	-0.622	0.019	0.294	0.000	-0.348	-0.041
	1σ	0.496	0.481	0.504	0.495	0.081	0.079	4.859	4.861
	n	1768	1768	1756	1756	1768	1768	1768	1768
SN218	MAE	1.149	0.257	13.690	0.735	0.145	0.053	2.652	2.456
	MBE	1.139	0.001	-13.69	-0.017	0.143	-0.003	0.949	-0.049
	1σ	0.375	0.357	3.368	0.921	0.075	0.068	4.822	4.816
	n	815	815	809	809	815	815	815	815

390

Table 5. Empirical calibration coefficients for slope (β_1) and solar zenith angle dependence (β_2) (Eqn. 2) used to calibrate each EM27/SUN to the reference SN219 during a co-located intercomparison.

	XCO ₂		XCH ₄		XCO		XH ₂ O	
	β_1	β_2	β_1	β_2	β_1	β_2	β_1	β_2
SN127	0.9995	0	1.0005	0	1.0025	0	1.0006	0
SN204	1.0005	0	0.99952	0	0.99824	0	0.99987	0
SN215	0.99986	0	1.0006	0	0.95826	4.13E-05	1.0006	0
SN216	1.0003	0	1.0001	0	0.99651	0	1.0001	0
SN217	1.0007	0	0.99976	0	0.99303	0	0.99981	0
SN218	1.0003	0	1.0006	0	0.75814	0.00023	1.0005	0



395 We analyse diurnal variability of the intercomparison across a four-day subset of the intercomparison (Figure 6) to complement
the previous statistical analysis. For all trace gases, the inter-EM27/SUN spread is largest during morning and evening,
associated with high SZA and greater retrieval uncertainty (Alberti et al., 2022). SN215 L1 XCO with distinct SZA dependence
(Figure 5) vary through time but are generally greater in the morning than evening (Figure 6a,b, green). With L2 SZA-
dependent correction (Eqn. 3), SN215 XCO differences reduce to within ~ 0.5 ppm (~ 0.5 %) (Figure 6c,d, green). L2 XCH₄
has a morning positive bias over the reference SN219 of 1.0 - 2.0 ppm (Figure 6c, top row) (0.05 - 0.10 %, Figure 6d) for
400 SN127 and SN217 and up to approximately 1.0 ppm negative bias in the afternoon. For XCO on 11th Apr around 14:00 UTC
there are some outliers likely associated with thin cloud that are not rejected by L1 QC.

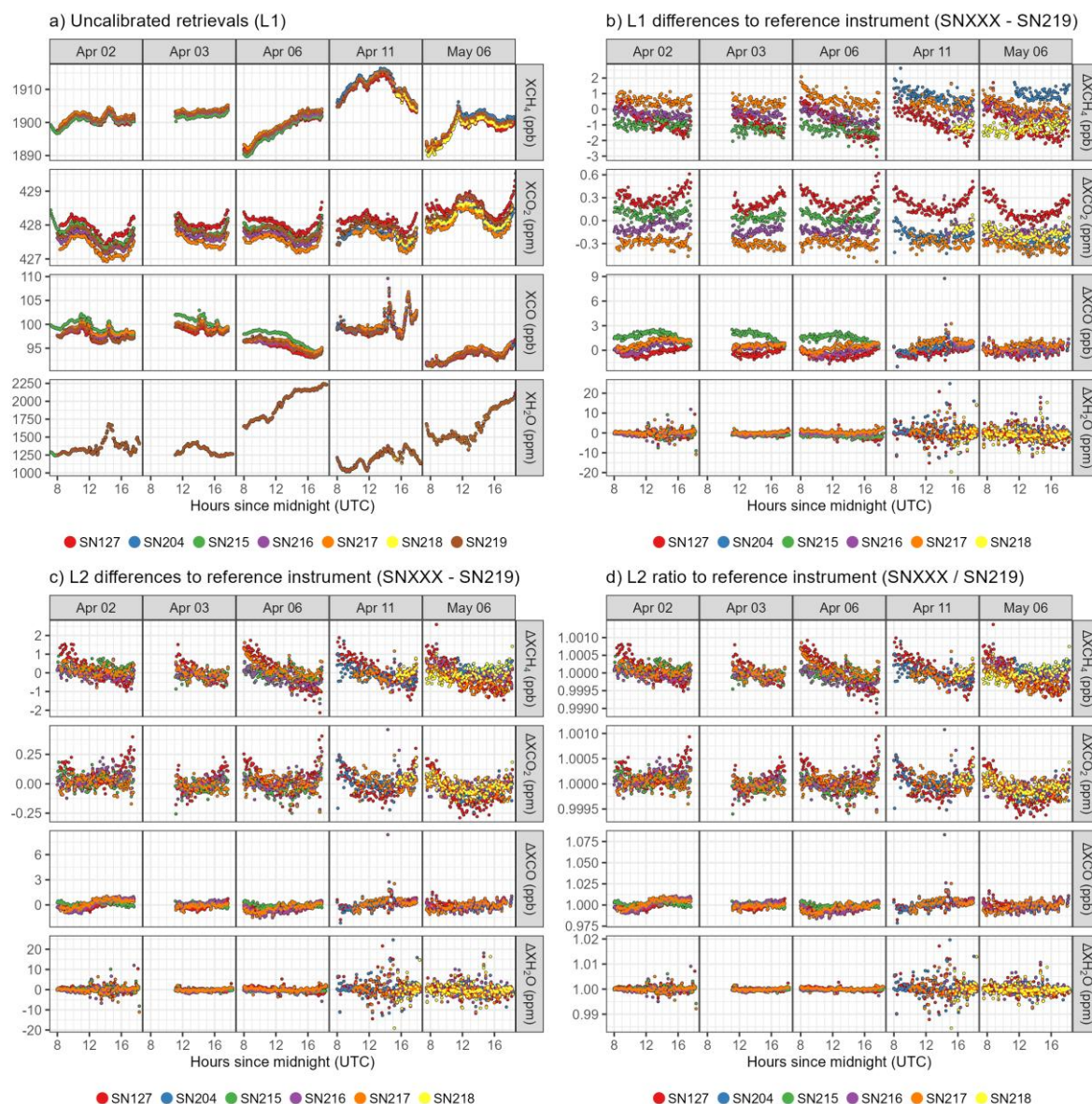


Figure 6. Five days of trace gas retrievals from (colours) seven co-located EM27/SUNs at station EDJCMB with a) quality-controlled retrievals (i.e. L1, Section 2.3.2) b) inter-EM27/SUN L1 differences of each EM27/SUN (SNXXX) to the reference EM27/SUN (SN219) c) inter-EM27/SUN L2 differences to SN219 also as (d) ratio. Solar zenith angles in the displayed data points range from 39.2° (2025-04-02_2025-05-06_intercomparisonV015_26052121434612:10 UTC May 6th) to 84.8° (18:15 UTC Apr 2nd). To constrain plot axes, suspect XCO SN218 is excluded.

3.4. Enclosed vs unenclosed L2 EM27/SUN intercomparisons

We assess the impact of the enclosure glass dome (Section 2.1) on retrieval performance using L2 data from co-located enclosed and unenclosed EM27/SUNs at EDJCMB (Figure 7). For XCH₄ and XCO₂ enclosure effects are small: MAE



increases by ≤ 0.2 ppb for XCH_4 and ≤ 0.003 ppm for XCO_2 when instruments operate unenclosed (Table 6). These effects are one to two orders of magnitude smaller than typical inter-instrument differences at L1, which have MAE ranging from 0.26 – 0.62 ppb for XCH_4 and 0.05 – 0.07 ppm for XCO_2 (Section 3.3, Table 4). The limited impact of the dome on retrieved XCO_2 and XCH_4 can be explained by the spectral channels used by PROFFAST, approximately 1.5 – 1.7 μm , (Frey et al., 2019; 415 Humpage et al., 2024), which lie within the spectrally flat spectral regions of the dome’s manufacturer-supplied transmission curve (Figure 1c).

For XCO, however, enclosure effects are more pronounced. Reduced dome transmission above $\sim 1.6 \mu\text{m}$ (Figure 1) coincides with the 2.315 - 2.375 μm XCO retrieval window (Humpage et al., 2024) and introduces a significant XCO bias. Enclosed 420 SN217 XCO has a small negative bias to enclosed SN216 (MBE = -0.176 ppb, Table 6). However, when SN216 is deployed unenclosed, SN217 reports substantially more XCO than SN216 (MBE 0.73 ppb). A similar bias is also found with the SN219 comparison (Table 6, MBE 1.10 ppb).

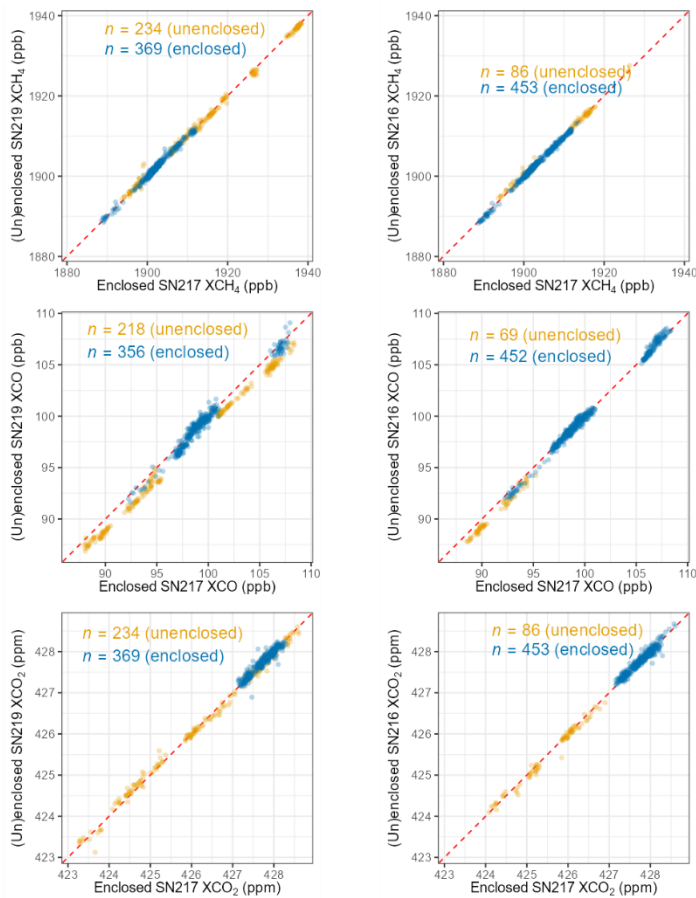


Figure 7. L2 retrievals from enclosed SN217 compared to co-located retrievals from (left) SN219 and (right) SN216 in (blue) enclosed and (red) unenclosed configurations at station EDJCMB. Solar zenith angles in the displayed data points range from 52.7° (12:20 UTC Mar 28) to 80.12° (09:35 UTC Feb 02).

Table 6. Intercomparison statistics for Figure 7 L2 data comparing SN217 that is always enclosed with L2 SN219 and SN216 that are enclosed and unenclosed through time.

		XCH4 (un- enclosed)	XCH4 (enclosed)	XCO (un- enclosed)	XCO (enclosed)	XCO2 (un- enclosed)	XCO2 (enclosed)
SN219	MAE	0.391	0.358	1.099	0.509	0.067	0.063
	MBE	0.207	0.241	1.099	0.330	-0.011	-0.029
	1σ	0.431	0.382	0.350	0.521	0.083	0.071
	n	126	105	121	100	126	105
SN216	MAE	0.507	0.321	0.730	0.355	0.055	0.058
	MBE	-0.038	0.142	0.730	-0.176	0.019	-0.042
	1σ	0.608	0.369	0.230	0.375	0.067	0.067
	n	58	78	50	77	58	78



3.5. Network-wide seasonal cycle of XCO₂, XCH₄, and XCO

The combined L2 dataset from all instruments provides a network-wide view of the seasonal variability in total column GHGs. The deployment phase, during which the number of operational instruments increased, began in November 2024 and concluded in June 2025. Figure 8 shows the annual cycle of XCH₄, XCO₂, XCO and XH₂O. As expected, the number of valid retrievals
435 increases over the deployment period, consistent with the gradual rollout of the network, with the highest data availability during summer 2025.

XCH₄ exhibits a distinct seasonal cycle, with lower values in summer (minimum in June 2025, around 1875 ppb) and higher values in winter (maximum around February 2025, about 1940 ppb). This seasonality is likely driven by increased heating-
440 related emissions during winter and enhanced atmospheric mixing and dispersion during warmer months. XCO₂ also shows a pronounced seasonal cycle, but with a different phase. The higher values were recorded in late spring (maximum around May 2025, around 428 ppm), following by a decrease to late summer minima (minimum in August 2025, about 416 ppm). This pattern is consistent with biospheric uptake during the growing season and carbon release outside the growing season, superimposed on background variability and atmospheric transport effects. XCO displays more complex variability.
445 Concentrations increased from approximately 90 ppb in winter 2024-2025 to 110-115 ppb in early spring, followed by a decline towards summer minima of about 75 ppb in June and July 2025. Superimposed on this seasonal cycle, two pronounced summer peaks were observed in June and August, reaching values of approximately 140 ppb. The early June enhancement may be partly associated with long-range transport of biomass burning emissions from Canadian wildfires, for which Copernicus Atmosphere Monitoring Service reported trans-Atlantic smoke transport reaching northwestern Europe (Copernicus Atmosphere Monitoring Service, 2026). The early August peak likely reflects short-term meteorological variability and
450 potentially residual influence from elevated wildfire activity across southern Europe in June and July.

Among all species, XH₂O concentration demonstrated the largest variability. During the early deployment phase, concentrations ranged from approximately 1000-3000 ppm, increasing to values exceeding 5000 ppm in summer 2025. A
455 notable fluctuation occurred in late July and early August 2025, characterised by sharp decrease followed by a rapid increase. This coincides with short-term variability in the other trace gases, suggesting a common meteorological driver, most likely related to changes in the boundary layer height and prevailing weather conditions. According to the Met Office, this period corresponded to a transition from persistent warm, dry anticyclonic conditions in July to a more cyclonic, Atlantic-driven regime in early August, culminating in Storm Floris on 4–6 August 2025 (Met Office, 2025).

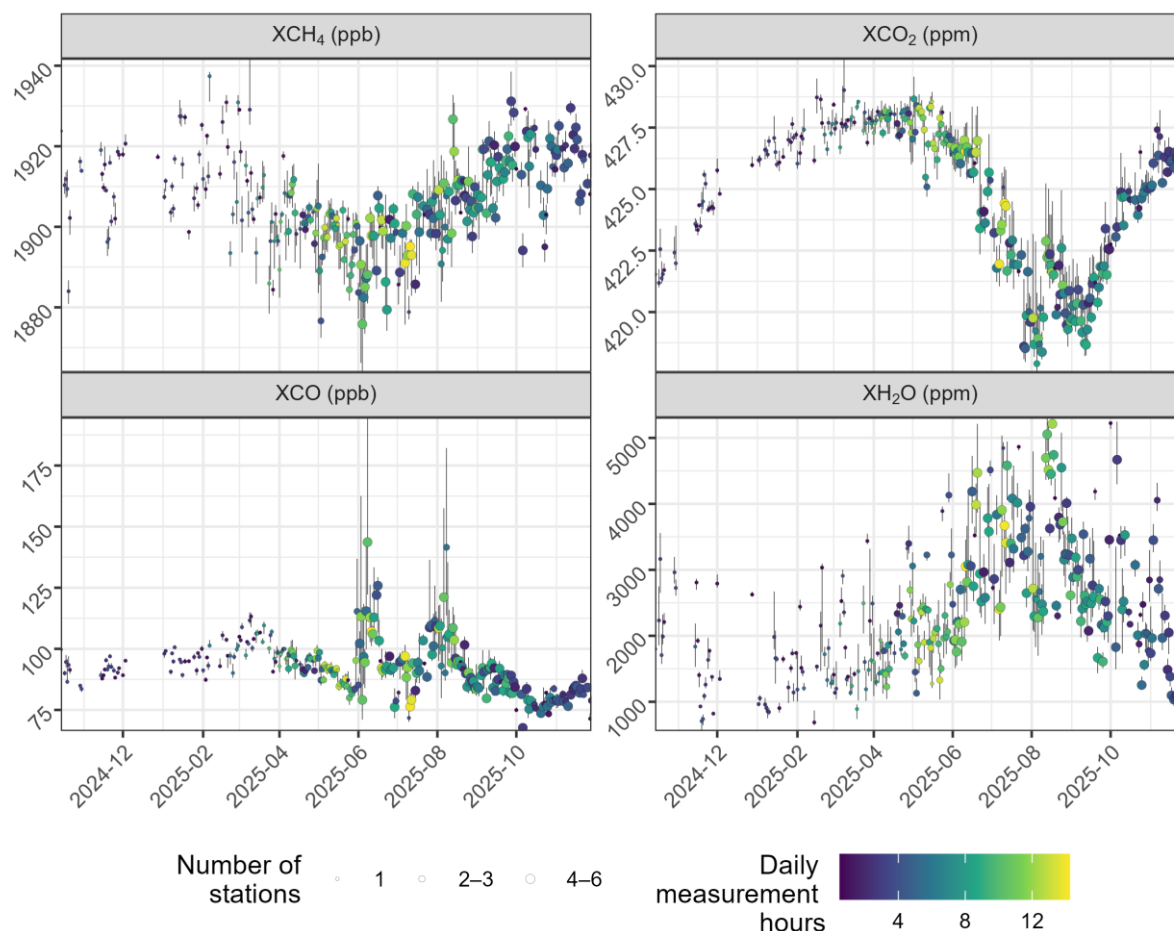


Figure 8. Network-wide annual cycle of XCH_4 , XCO_2 , XCO and XH_2O with (circles) daily median across all available EM27/SUNs, (colours) the number of hours of measurements in a day across (circle size) the number of EM27/SUNs on the given day.

3.6. Intra-city differences in L2 retrievals

We analyse upwind-downwind differences in observed trace gases using ratios between each station and the city-centre reference station (CHRY), expressed as station/CHRY. Under the assumption that urban emissions of XCO_2 , XCO and XCH_4 emissions enhance concentrations downwind of the city, ratios are expected to be < 1 when a station is located upwind of CHRY and > 1 when the station is downwind. We group available observations by wind sector (Figure 8), with the analysis focusing on north-easterly marine flow and the prevailing south-westerly flow (Figure 2). These wind regimes typically traverse the NARO → CHRY → LDST transect (Figure 2), allowing a consistent assessment of upwind and downwind conditions relative to the city centre.

Among the analysed species, XCH_4 shows the most consistent behaviour with respect to the expected upwind-downwind contrast (Figure 9). The LDST/CHRY ratios shows values < 1 under north-easterly flow when LDST is upwind of the city centre and > 1 under south-westerly flow when LDST lies downwind. This is consistent with enhanced methane concentrations originating from urban emissions and subsequent downwind transport.

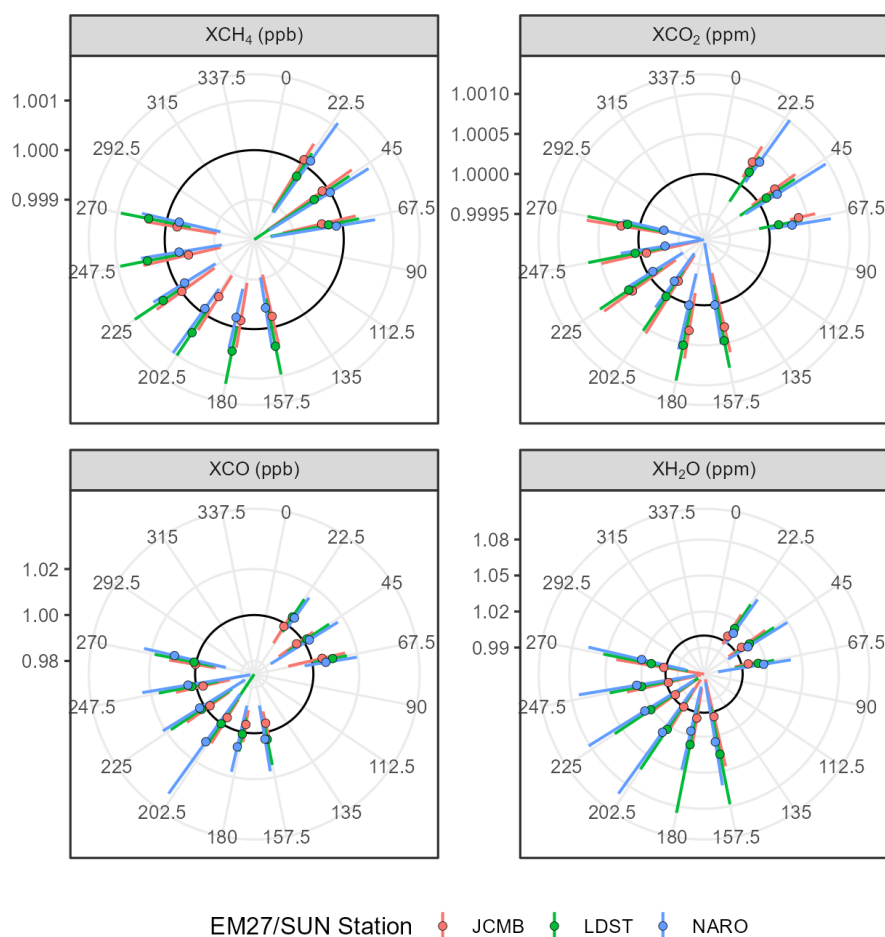


Figure 9. Wind-sector dependence of station to city centre ratios (station/CHRY) for XCO_2 , XCO , XCH_4 , and XH_2O . Retrievals are grouped by wind direction sectors derived from the JCMB automatic weather station. Only sectors meeting minimum data coverage criteria are included, requiring at least 12 hours of total data per sector and a minimum of 30 minutes of observations per day. Ratios below (above) one indicate lower (higher) column-averaged concentrations relative to the city centre.

In contrast to XCH_4 , the behaviour of XCO_2 and XCO is more complex and does not conform to a simple upwind-downwind interpretation. For XCO_2 , all stations tend to show station/CHRY ratios > 1 under north-easterly flow, indicating higher column values outside the city centre regardless of their relative position to CHRY. Under south-westerly flow, LDST and JCMB consistently shown higher XCO_2 than CHRY, while NARO more frequently exhibit ratios < 1 . This spatial pattern suggests



485 that XCO_2 gradients are not governed solely by local urban emissions but are strongly influenced by variations in background concentrations, air mass origin, representativity of surface wind against vertical profiles and wind shear, and potentially biospheric uptake associated with urban and peri-urban vegetation.

A similarly ambiguous pattern is observed for XCO. Although XCO is primarily associated with fossil fuel combustion and
490 might therefore be expected to show clearer urban enhancements, the station/CHRY ratios do not show a consistent directional dependence. For both north-easterly and south-westerly flow, NARO and LDST frequently have higher XCO than the city centre, while JCMB tends to remain lower than the city centre. The absence of a clear upwind-downwind signal suggests that sources outside the city centre, as well as atmospheric transport, contribute significantly to the observed total column variability.

495 The behaviour of the NARO station, located south-east outside of the city, is particularly noteworthy. Although initially considered representative of urban background conditions due to its location, NARO frequently shows higher XCO_2 and XCO column concentrations than the city centre. This is most evident under south-westerly flow when NARO is nominally upwind of CHRY. Such behaviour indicates that NARO is influenced by additional sources or transported enhancements, potentially
500 including regional anthropogenic emissions. These findings underscore the difficulty of defining a true background station in a heterogeneous urban and peri-urban environment, especially when using total column measurements that integrate signals over large spatial scales.

For XH_2O , all stations consistently show higher values than the city centre across all wind sectors, indicating systematically
505 enhanced humidity outside the urban core. This pattern is consistent with reduced evapotranspiration and increased impervious surface coverage within the city centre relative to surrounding areas.

The analysis of statistically significant differences relative to CHRY (Figure 10) further highlights the complexity of the observed spatial patterns. For XH_2O , a clear and consistent positive bias is again observed at all non-central stations,
510 reinforcing the interpretation of higher humidity outside the urban core. For XCO, NARO stands out, showing the largest fraction of positive differences relative to CHRY, particularly during daytime. For XCO_2 and XCH_4 , a pronounced diurnal structure emerges in the station-CHRY differences, characterised by more negative values during the early morning and late evening periods and more positive values during the day. This behaviour is not readily explained by a simple upwind-downwind urban enhancement framework. For XCO_2 in particular, nighttime respiration combined with reduced boundary
515 layer mixing may enhance concentrations at specific locations, while daytime boundary layer growth and photosynthetic uptake modify the spatial gradients.

Month-to-month variability is clearly present in the station-CHRY differences for all tracers except XH_2O , where the positive differences remain relatively constant. For XCO, LDST and JCMB show significant variations, with their values decreasing



520 towards September. For XCO_2 , the spatial pattern is also changing substantially between months. The NARO site transitions from predominantly positive differences in June-July to mostly negative differences later, while LDST shows increased positive differences in August-September. For XCH_4 , JCMB exhibits pronounced negative difference in August, while LDST shows a positive difference in September.

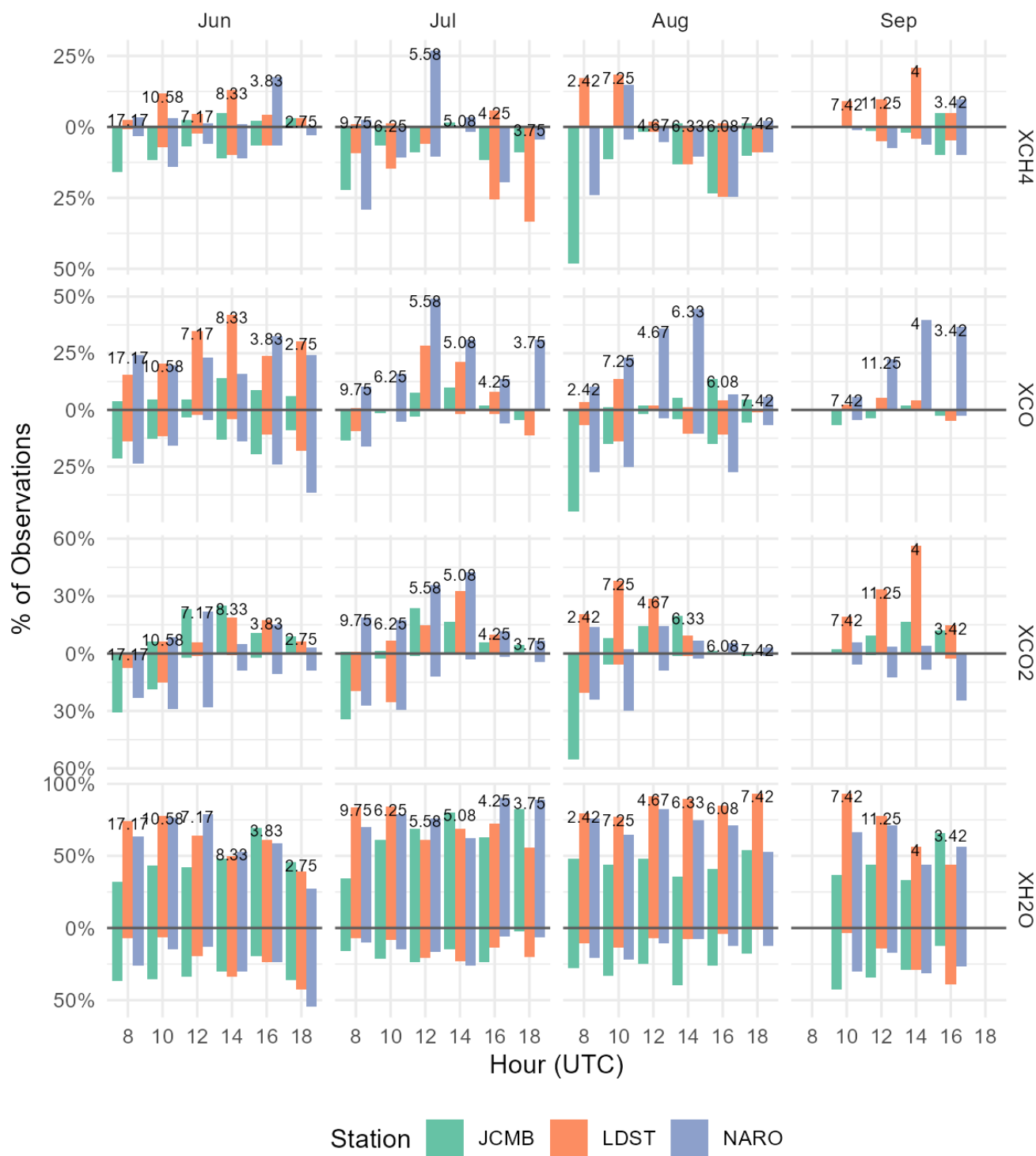




Figure 10. Percentage of hourly observations that exceed a significant difference relative to the city centre station (CHRY), with significant difference defined as $(\text{station} - \text{CHRY}) > 3\sigma$, where σ represents the inter-instrument precision derived from calibration (Table 4; maximum value across L2 products used for each species) split by (rows) XCO, XCO₂, XCH₄, and XH₂O trace gases and (column) months over the period 2 June to 29 September 2025. Bars above (below) the zero line represent the % of L2 observations that are 3σ greater (less) than the city centre with (bar labels) the number of hours of observations within the analysis e.g. 17.17 h is 206 5-minute L2 observations across all EM27/SUN for mutual timesteps.

To better understand the origin of the observed intra-city differences, we complement the observational analysis with ICON-ART model values and focus on a small number of selected days. We analyse two days, 11 May and 11 June 2025, which we selected based on the presence of pronounced flux signals in the model. By examining periods during which the model indicates clear contributions from anthropogenic and biogenic fluxes, we assess the extent to which the observed intra-city differences can be attributed to emissions, atmospheric transport, or background variability.

On 11 May 2025 (Figure 11, top row), only two stations were operational: JCMB and NARO. According to the model, XCO₂ at the reference location station JCMB shows a clear diurnal cycle, with a minimum around midday coinciding with a peak in GPP. However, this midday decrease is not evident in the observations. A similar discrepancy is present in the modelled JCMB minus NARO differences (Figure 11). The ICON-ART simulations show higher XCO₂ at JCMB than at NARO by approximately 0.2 ppm, whereas the observations indicate higher concentrations at NARO by about 0.1 ppm. This case represents a particularly challenging scenario for data assimilation. Despite the strong flux signal in the model, the disagreement with observations appears to result from a combination of errors in model atmospheric transport, background concentrations, and surface fluxes. Separating these contributions would require simultaneous retrievals from multiple stations and a clear characterisation of the error correlations associated with each component.

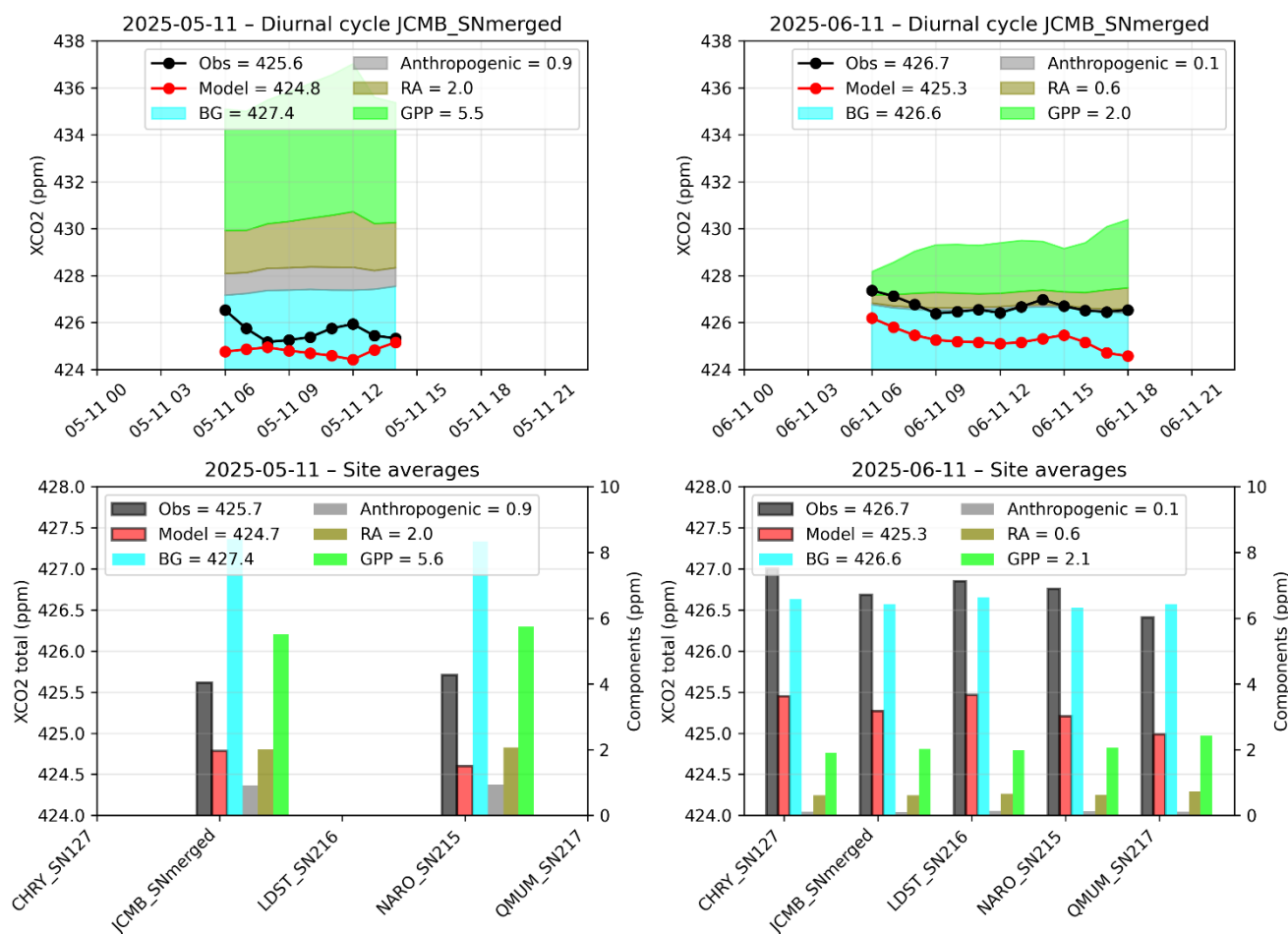


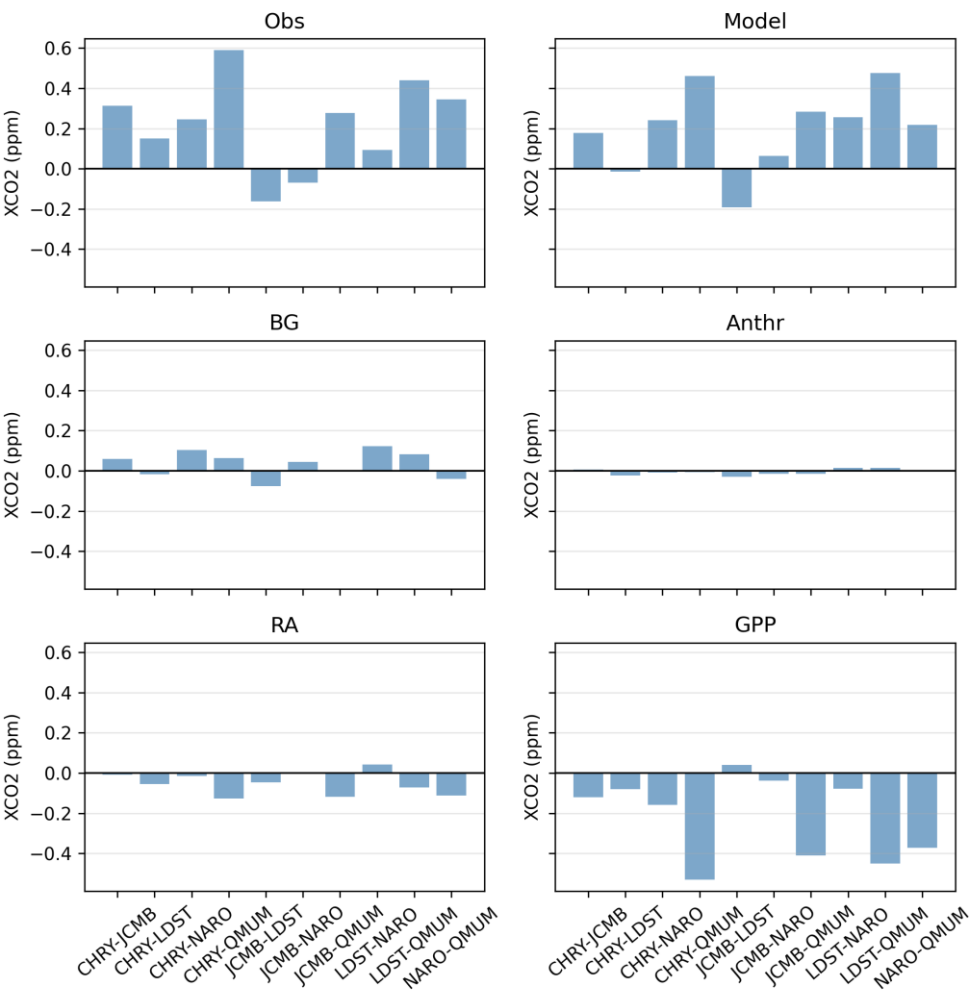
Figure 11. Diurnal variation of XCO₂ at the reference station (JCMB), top row, and daily mean distribution across the network for two case study days (11 May and 11 June 2025), bottom row. Top panels show the total column observed and modelled XCO₂ hourly time series, together with the model components shown as shaded areas for background (BG), anthropogenic (Anthr), vegetation respiration (RA), and the gross primary productivity (GPP). Bottom panels present daily mean XCO₂ for different stations, comparing observed and modelled values, the model values include total and the individual components.

In contrast, the 11 June 2025 case shows substantially better agreement between the model and observations in terms of diurnal variability, although a nearly constant negative offset is present throughout the day (Figure 11, top left). With all five stations operational on this date, the spatial gradients are generally well captured by the model. Pairwise differences show consistent patterns between model and observations (Figure 12). The largest positive gradients occur between the city central station CHRY and QMUM (approximately 0.60 ppm in observations and 0.45 ppm in the model), followed by LDST-QMUM (approximately 0.40 in the observations and 0.45 ppm in the model). The smallest and negative gradients are found for JCMB-LDST (approximately 0.20 ppm for both model and observations) and JCMB-NARO (slightly negative in the observations and slightly positive in the model). Compared to the May case, the magnitude of the flux-driven components is smaller on 11 June, with average contributions of approximately 0.60 ppm from ecosystem respiration (RA) and 2.10 ppm from GPP across



stations. The reduced biogenic signal is consistent with the weaker diurnal variability and likely contributes to the improved agreement in spatial gradients.

565 Overall, these case studies highlight the importance of combining multi-station observations with atmospheric modelling to robustly interpret urban greenhouse gas measurements. They further demonstrate that reliable attribution of observed gradients requires both dense observational coverage and accurate representation of background concentrations, atmospheric transport processes, and surface fluxes, which are all key prerequisites for future inverse modelling and data assimilation efforts.



570 **Figure 12. Daily mean XCO₂ differences between all available station pairs on 11 June 2025. Panels show the difference between stations in daily mean observed XCO₂, model total XCO₂, and the differences in individual model components (background, anthropogenic, respiration, and GPP contributions). Negative GPP differences correspond to smaller vegetation sink contribution.**



3.7. Model interpretation of GEMINI-Edinburgh XCO₂ data

We assess the ability of ICON-ART to reproduce the day-to-day variability of retrieved XCO₂ across May 2025 by comparing it against EM27/SUN measurements (L2 JCMB SNmerged; Figure 13). The model XCO₂ is decomposed into contributions from background conditions (driven by initial and boundary conditions), anthropogenic emissions, and biogenic fluxes (RA and GPP). Over most of that study period, variability in total column XCO₂ is dominated by the background contribution. Anthropogenic and biogenic components are comparatively small and become clearly distinguishable during limited episodes, ranging from individual days to about a week, when local influences are enhanced.

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These episodic periods are particularly informative for analysing differences between upwind and downwind stations, as they reflect sensitivity to locally driven signals rather than large scale advection. Provided that atmospheric transport and the prior flux variations are realistic represented in the model, such conditions are favourable for interpreting observations and for use in atmospheric inversions. Outside these periods, variability is largely governed by large-scale background fluctuations. Under such conditions, differences between upwind and downwind stations are less indicative of local emissions and instead reflect background air mass properties. Consequently, for a substantial fraction of time, the dominant constraint provided by the observations is on the background XCO₂ rather than on local flux contributions.

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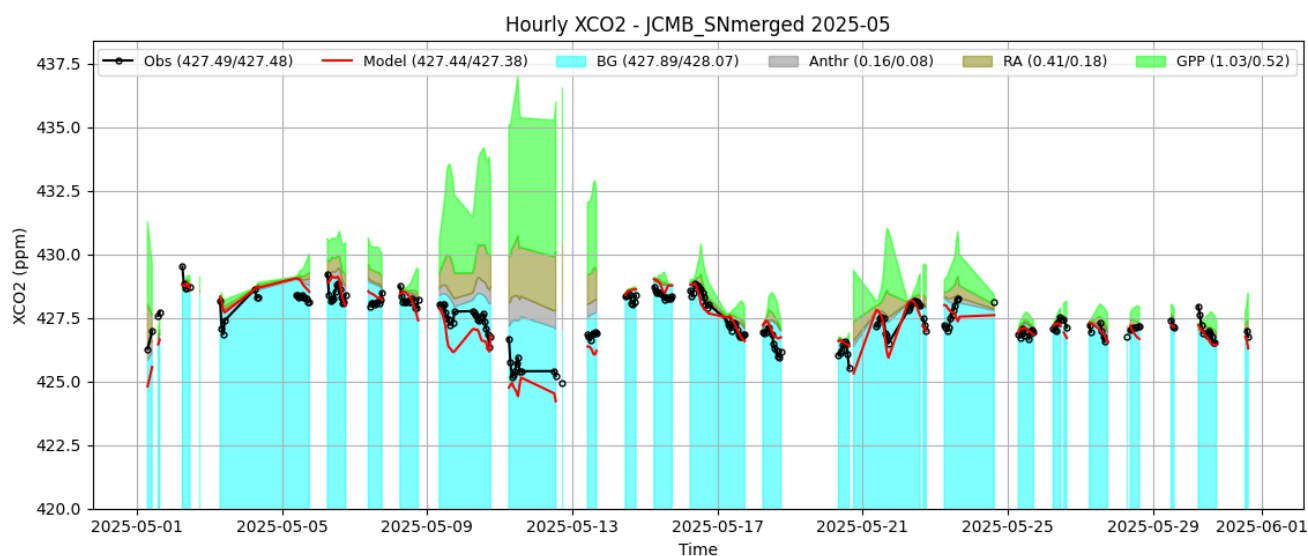


Figure 13. Example time series of EM27/SUN JCMB SNmerged XCO₂ for May 2025: L2 flagged data (Obs) compared with ICON-ART simulations (total column Model and individual components: background BG, anthropogenic Anthr, vegetation respiration RA, gross primary production GPP). The mean and median values over the shown period are indicated in the legend, separated by a slash.

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Across all EM27/SUN stations, aggregated monthly statistics (Figure 14) show that the overall mean model-observation bias across the study period is close to zero. However, month-to-month variability ranges from approximately -0.50 ppm in June to +0.40 ppm in March 2025. The cRMSE averages around 0.56 ppm, with a gradual increase towards late spring, reaching approximately 0.60 ppm in June. A similar seasonal evolution is observed in the correlation coefficient, which averages 0.60 and decreases slightly in June. Both model and observations show increasing monthly mean XCO₂ values from winter into spring (Figure 14, bottom panel), consistent with changes in the background CO₂ that typically peaks in late spring due to wintertime accumulation and the delayed onset of biospheric uptake.

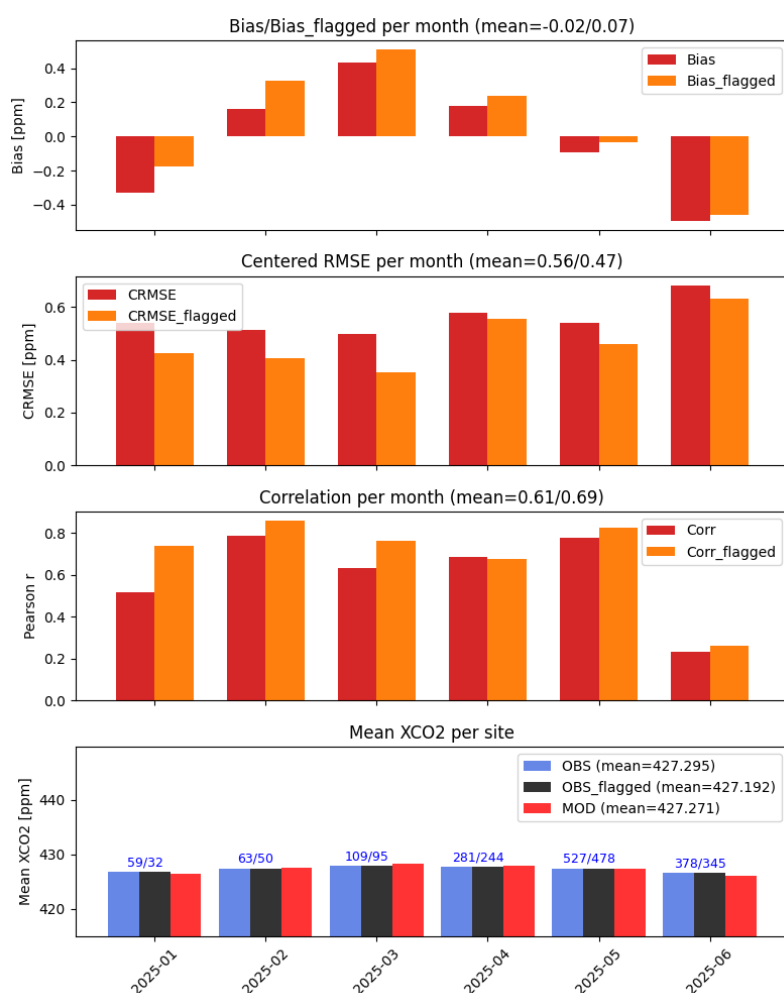


Figure 14. Monthly statistics of forward ICON-ART XCO₂ compared with L2 data from each EM27/SUN station with: bias, cRMSE, correlation, mean concentrations with “_flagged” indicating application of all optional L2 quality control flags (Table 2). The number of hourly intervals used for comparison is indicated above bars on the bottom panel in blue (total number / excluding flagged observations).

Figure 15 shows station-specific ICON-ART XCO₂ statistics. The average bias across all stations is -0.30 ppm, with the largest negative bias at the eastern QMUM station (around -0.50 ppm). The south-eastern JCMB station has a bias close to zero.



Stations with a larger number of observations (Figure 4) tend to show smaller biases and lower cRMSE than stations with fewer than 200 (or even fewer than 100) hourly averaged measurements. The mean correlation coefficient across all stations is 0.44, indicating moderate agreement at the individual station level.

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These error metrics generally improved when we apply the optional L2 QC flags (Section 2.3.2). However, the mean bias remains essentially unchanged. This is consistent with the bias being dominated by slowly varying background CO₂ differences rather than unflagged L2 outliers. Although the model-observation bias is relatively small, differences between stations are significant when compared to the expected precision of the L2 EM27/SUN L2 data products (on the order of 0.05-0.08 ppm, Table 4). The absolute accuracy of the instruments, as tied to the COCCON reference and ultimately to TCCON/WMO scales, is approximately ± 0.50 ppm. This indicates that the remaining discrepancies are more likely related to uncertainties in input fluxes or atmospheric transport rather than to measurement error. Overall, the model provides a reliable representation of observed XCO₂ variability while capturing differences that are large enough to be informative for flux attribution.

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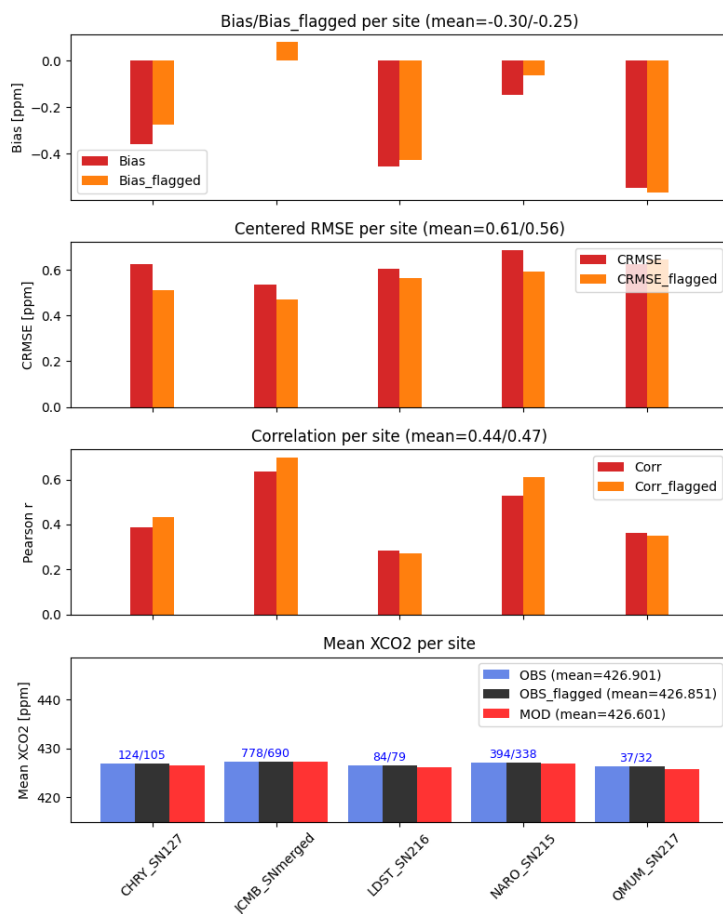


Figure 15. Inter-station statistics of forward ICON-ART XCO₂ compared with L2 EM27/SUN observations: bias, cRMSE, correlation, and mean concentrations. The number of observations used for comparison is indicated above bars in blue (total number / excluding flagged observations).

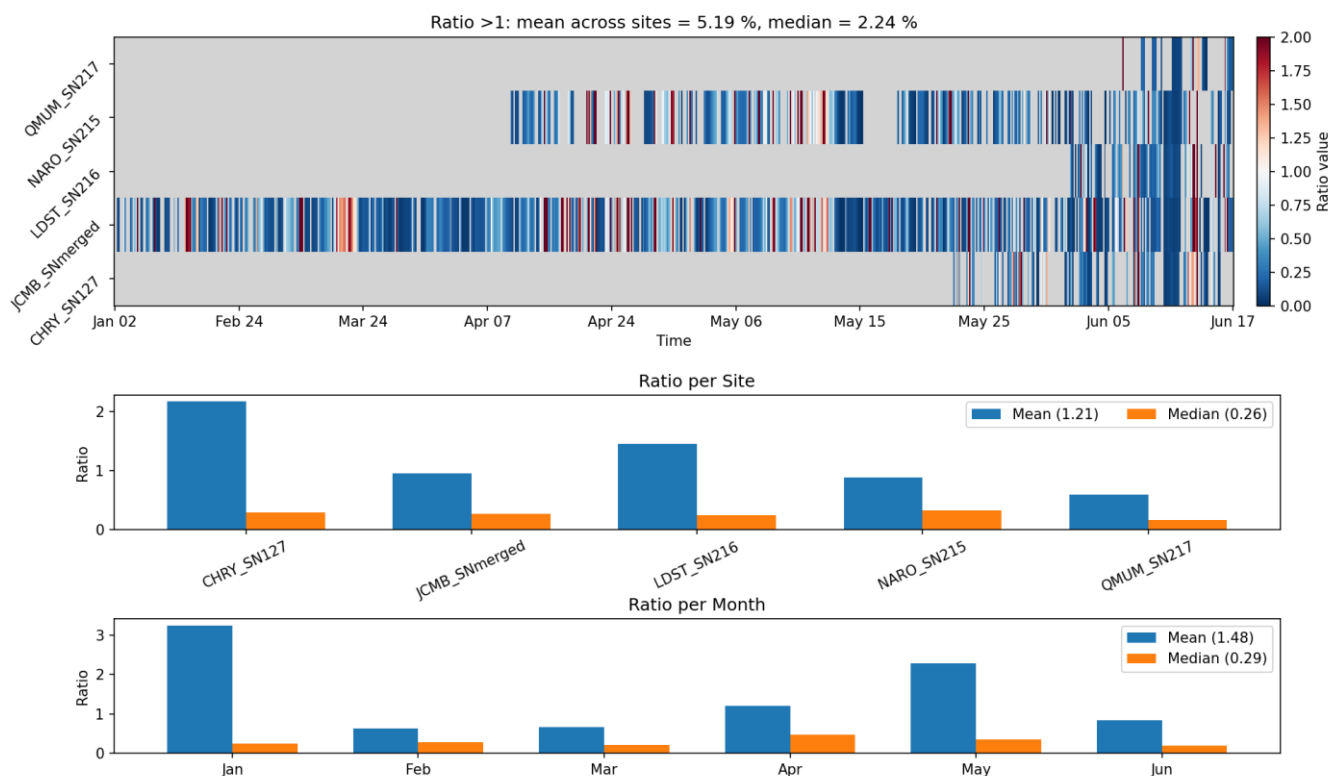


Figure 16. Anthropogenic XCO₂ contribution compared to model–observation differences. (Top) Heatmap of the ratio of the absolute anthropogenic XCO₂ signal to the absolute model–observation difference, $|XCO_2_{mod_A}| / |XCO_2_{obs_flagged} - XCO_2_{mod}|$, for all station-sensor pairs over the analysis period; with ratios greater than 1 showing where the anthropogenic signal exceeds the absolute model–observation difference. (Middle) Mean and median ratio per station. (Bottom) Mean and median ratio per month. Mean and median fractions of times where the ratio exceeds 1 are indicated in the heatmap title.

Time series of station-sensor pairs (e.g., Figure 13) further demonstrate that the anthropogenic contribution (Anthr) is generally quite small (typically < 0.20 ppm), with the signal often smaller than the random background variability. The analysis shows that in only about 5% of cases does the anthropogenic signal exceed the absolute model-observation bias. While the mean ratio between anthropogenic signal magnitude and absolute bias across all EM27/SUN stations and months is close to unity, the median is much lower (approximately 0.25), indicating a strongly skewed distribution. The elevated mean is driven by rare events during which the anthropogenic signal clearly exceeds background variability. During such events, the model and observed values are typically in good agreement.

The limited contribution of anthropogenic fluxes to total column XCO₂ is further illustrated by vertical profile analysis. Appendix D shows domain-averaged vertical profiles of total CO₂, along with its anthropogenic and biospheric components, for a representative timestep, including layer mass and the corresponding mass-weighted contributions to the total column. Most of the atmospheric mass resides between 2.5–10 km, while anthropogenic and biospheric sources are concentrated near



the surface. This vertical separation explains why the anthropogenic contribution to total column XCO_2 remains small and why even higher near-surface sensitivity (e.g. via larger values of the averaging kernel) would not substantially increase the contribution to the column-integrated signal.

645 4. Concluding Remarks

We have successfully established an EM27/SUN measurement network across the City of Edinburgh to provide continuous total column observations of CO_2 , CH_4 , and CO at intra-urban scales. The network design captures upwind, urban core, and downwind conditions, within a compact spatial footprint ($\sim 5\text{--}7$ km station spacing), enabling the first coordinated assessment of spatial gradients in total column greenhouse gases over a mid-sized UK city. These observations provide critical information
650 for evaluating urban GHG fluxes where long term ground-based atmospheric constraints are currently sparse.

Inter-instrument comparisons demonstrate that the inter-EM27/SUN precision using quality controlled and calibrated “level 2” (L2) data is robust and consistent with previously reported EM27/SUN studies (Chen et al., 2016; Humpage et al., 2024). The intercomparison dataset spans approximately six weeks (26 March to 15 May 2025) and includes 600–1850 observations
655 per instrument pair under clear-sky conditions. While the number of co-located observations are sufficient to characterise inter-instrument behaviour under these conditions, the analysis is restricted to a single season and does not capture potential variability under different weather and annual trace gas cycles. Inter-instrument differences show a clear diurnal dependence, with increased spread at higher solar zenith angles. The EM27/SUN enclosure design has a negligible impact on XCO_2 and XCH_4 retrievals but introduces a systematic XCO bias requiring correction.

660 Intra-city L2 retrievals cannot be consistently interpreted using a simple upwind–downwind enhancement framework. While XCH_4 exhibit coherent directional behaviour, XCO_2 and XCO are strongly influenced by background variability, air-mass history, and regional transport. As a result, urban-scale gradients in total column measurements are typically subtle and episodic, emphasising the need for multi-station observations combined with atmospheric transport modelling.

665 ICON-ART mesoscale model simulations support this interpretation, reproducing the observed day-to-day variability with low mean bias and moderate correlation. Model minus observation differences are primarily driven by background concentrations and transport, rather than local emissions. The anthropogenic contribution to column XCO_2 over the City of Edinburgh is typically small ($\sim 0.2\text{--}0.3$ ppm compared to a background of ~ 420 ppm) and exceed model minus observation differences is
670 only $\sim 5\%$ of cases, highlighting the intrinsic difficulty of isolating urban emission signals from total column measurements. This contribution is comparable to a study for the Paris metropolitan network (Doc et al., 2026).



Our results demonstrate that, for mid-sized UK cities such as Edinburgh, observed spatial gradients are frequently dominated by background variability rather than local emissions alone. Robust attribution of urban fluxes therefore requires dense observational coverage and accurate representation of atmospheric transport and background conditions.

Future work will extend the observational record to capture year-to-year variability and incorporate an inverse modelling framework to estimate urban emissions from the atmospheric data, enabling tracking of progress toward emission reduction targets in accordance with the UK Net Zero Strategy and the Global Methane Pledge. Such a data-modelling framework will also enable a systematic assessment of the relative benefits of different observing strategies, including comparisons between the EM27/SUN network with urban-focused satellite observations, such as the city-scans capability of the MicroCarb mission (Wu et al., 2023). Together, these approaches provide a pathway toward robust, observation-based verification of urban greenhouse gas emissions, supporting evidence-based evaluation of environmental policy.



Appendix A. Further details of GEMINI-Edinburgh EM27/SUN instruments

Table A1 gives further details on the EM27/SUN instruments used in this study, including instrument serial numbers, instrument line shape (ILS) calibration dates, manufacturer repairs, and deployment notes.

690 **Table A1** Summary of EM27/SUNs used in this study, using instrument line shape (ILS) published by the Collaborative Carbon Column Observing Network (COCCON) in PROFFASTpylot software (Feld et al., 2024).

EM27/SUN serial number (SN)	ILS date	Repairs by manufacturer (Bruker, Germany)	Further comments
SN127	2019-12-11		Deployed in field campaigns around the United Kingdom 2020 – 2024.
SN204	2023-04-19		Used for testing and intercomparison at EDJCMB only. Prior to this study used for ~days of tests and measurements.
SN215	2023-12-14	Preamp board reconfiguration (mid-2024); general electronics failure (Jan 2025); suspect XCO ₂ bias, faulty flip mirror (Nov 2025, ongoing as of April 2026).	New in this study.
SN216	2023-12-14		New in this study.
SN217	2023-12-06	Preamp board reconfiguration (mid-2024).	New in this study.
SN218	2023-12-06	He–Ne laser realigned (April 2025). Further repairs ongoing as of April 2026.	New in this study.
SN219	2023-12-06		New in this study and used as the reference for inter-EM27/SUN precision.

Appendix B. QC subset

This appendix provides additional examples and diagnostics illustrating the performance and impact of the level 1 (L1) quality control. Using three days of varied clear-sky, cloudy, and sun tracker-misaligned conditions, the QC framework identifies and removes EM27/SUN SN204 retrievals at station EDJCMC (Table A1). 8th August 2025 is mostly cloudy with periods of direct sun. With manual reference to CamTracker images, around 17:00 the sun tracker is misaligned, and these values are successfully removed by automated QC (Figure B1, red). Similarly, but more clearly on 16th August, the sun tracker is misaligned around 12:00 -> 15:00 (CamTracker images, Figure B2). The effect in the retrieval is nuanced for XCO₂ and XCH₄ and the values are again successfully removed by automated QC. On a clear-sky day 17th August, there is no misalignment but some noise in XAIR diagnostic triggers removal of a small number of retrievals according to flag_xair_noise_threshold.

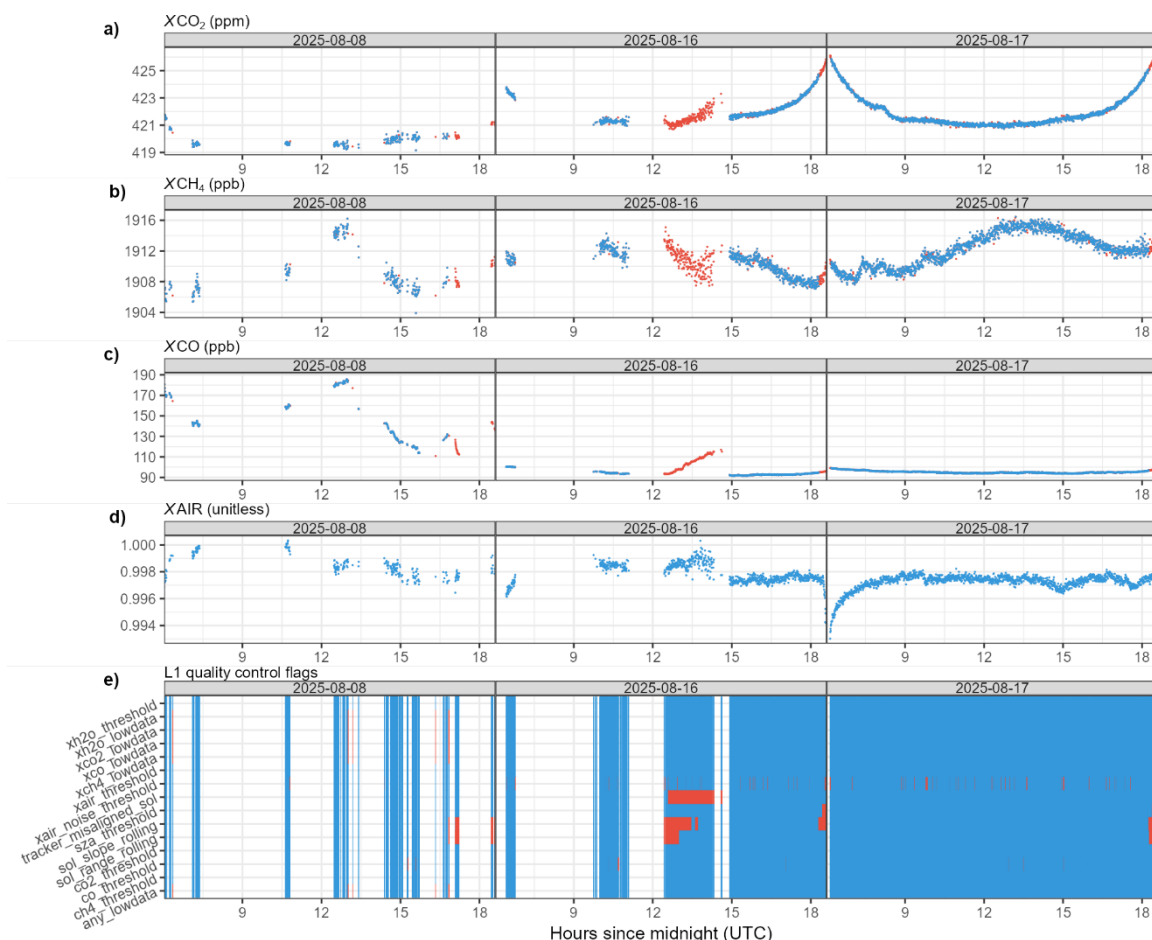


Figure B1. Total column concentration retrievals of (a) XCO₂, (b) XCO and (c) XCH₄ from EM27/SUN SN204 for three days in August 2025 with (red) values rejected from the level 0 (L0) data and (blue) the L1 quality controlled (QC) data using (d) PROFFAST diagnostic variable XAIR and (e) a series of QC flags (details and thresholds in Section 2.3.2 and Table 2).

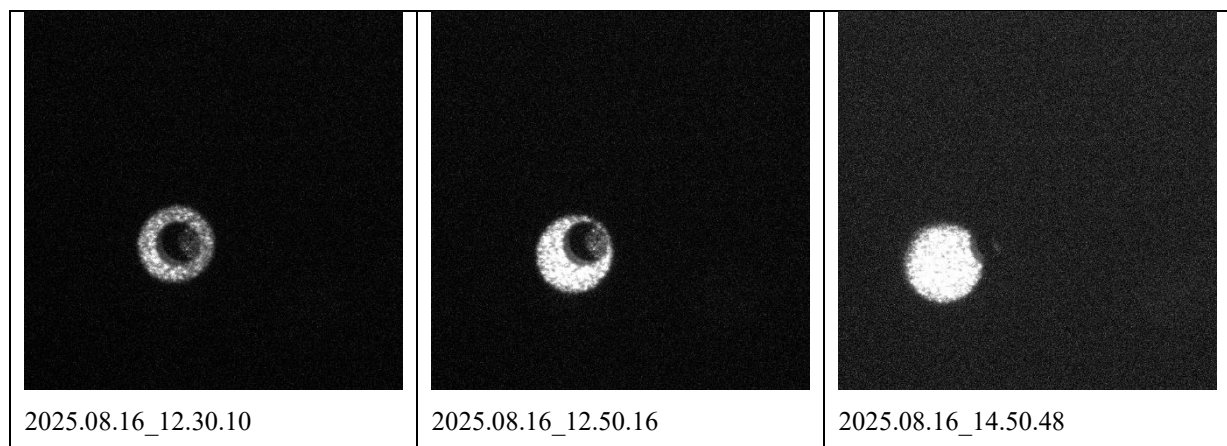


Figure B2. CamTracker images for EM27/SUN SN204 for 16th August 2025 with (white circle) the picture of the solar disk on/near the field stop and (black dot) the field stop opening with (right) aligned sun tracker with solar disk aligned over the field stop (middle) gradual misalignment of solar disk with field stop and (right) near-complete misalignment.



710 Appendix C. XCO₂ model data extraction algorithm

This appendix provides a schematic overview of the XCO₂ extraction procedure from ICON-ART simulations (Figure C1). After horizontal interpolation to the station location, model CO₂ profiles and dry air density are vertically matched to the EM27/SUN retrieval grid. Total CO₂, and its individual components (background (BG), anthropogenic (Anthr.), vegetation respiration (RA), and gross primary production (GPP)) are treated separately to account for differences in their prior information. The final total column XCO₂ values are computed by vertically integrating the CO₂ profiles weighted by the averaging kernels, dry air density, and layer thickness.

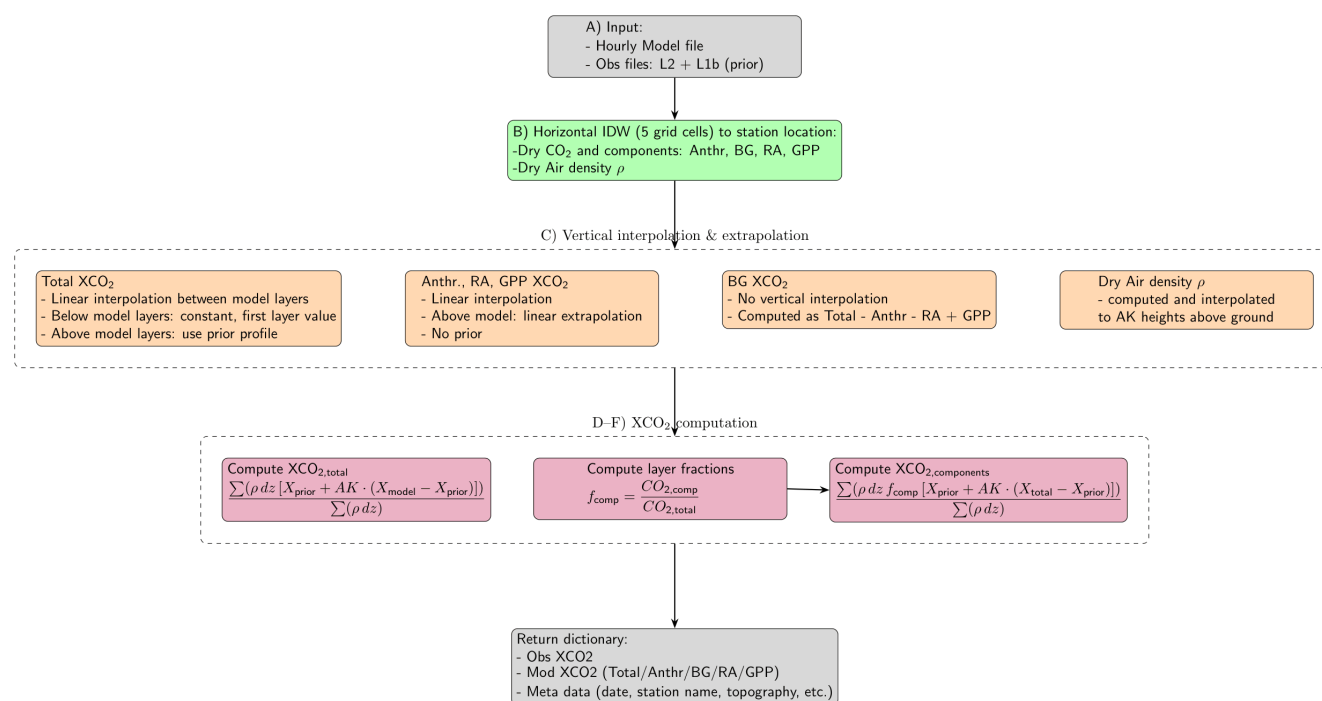


Figure C1 Schematic flowchart for extracting modelled XCO₂ from ICON-ART simulations at EM27/SUN stations. Panels show (A) input data including hourly model output and level 2/1b observation files, (B) horizontal interpolation to station locations, (C1–C4) vertical interpolation and extrapolation for total CO₂ and its components (Anthr., RA, GPP), background CO₂, and dry air density, (D–F) computation of total and component XCO₂ including layer fractions and vertical integration, and (G) final output dictionary containing modelled and observed XCO₂ and metadata.

Appendix D. XCO₂ anthropogenic signal

This appendix provides an illustration of the limited anthropogenic contribution to total column XCO₂ (Figure D1). The left panel shows a representative example of the vertical CO₂ profiles for different modelled components averaged across the model domain. The middle panel presents the mass-weighted contribution of each component to the total column, while the right panel shows the vertical distribution of atmospheric layer mass ($\rho \times dz$). Although anthropogenic enhancements are strongest near the surface, most of the atmospheric mass is located higher in the column. As a result, the anthropogenic contribution to total XCO₂ remains small compared to the dominant background signal.

Vertical CO₂ Profiles — 11 Jan 2025, 10:00 UTC

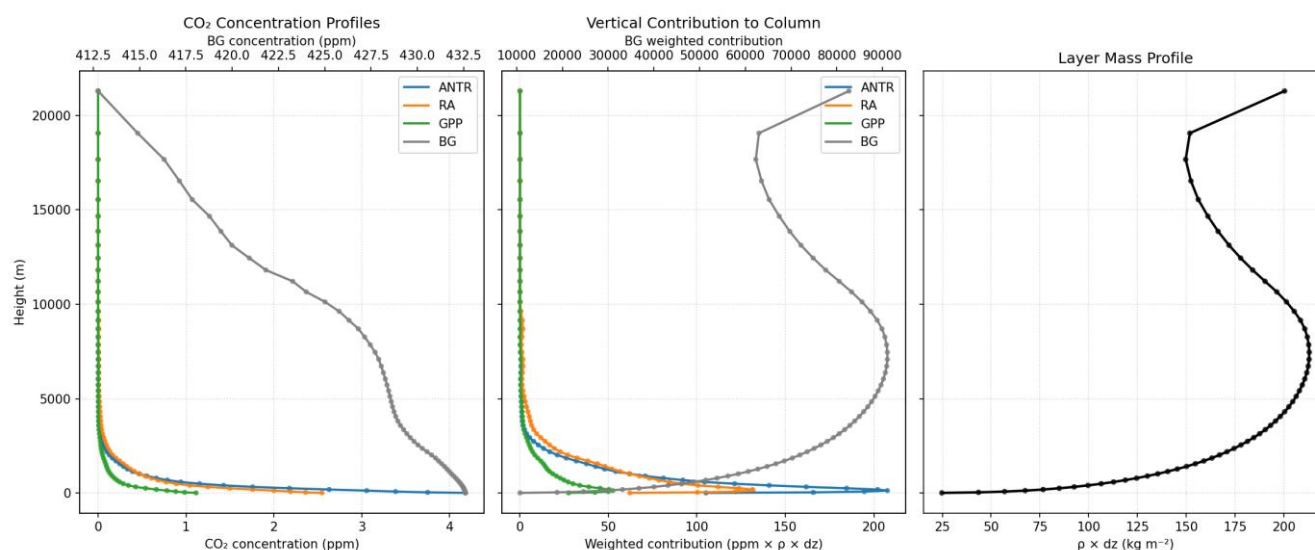


Figure D1. European domain-average vertical profiles of CO₂ for 11 January 2025, 10:00 UTC: (Left) Component concentrations (Anthropogenic ANTR, Respiration RA, Gross Primary Production GPP) with background (BG); (Middle) Vertical contributions to column (ppm × $\rho \times dz$) including BG; (Right) Layer mass profile ($\rho \times dz$).



Appendix E. EM27/SUN measurement software and scripts

The measurement software and automation framework described in this appendix underpin the reliable, continuous operation of the Edinburgh EM27/SUN network. A combination of manufacturer-supplied software and custom Python scripts enables fully automated solar tracking, interferogram acquisition, environmental monitoring, data screening, and secure data transfer with minimal human intervention.

Table E1. Measurement software installed on each computer locally linked to each EM27/SUN.

Enclosure PC Software	Summary of software and task(s)
OPUS version 8.8.4 (64 bit)	EM27/SUN data capture software. An OPUS macro facilitates the continuous capture of interferograms, automated by Pyra software.
Pyra version 4.2.0	Measurement automation software (Aigner et al., 2023; Dietrich et al., 2021).
NetTime version 3.14	Reliable and verbose time syncing tool (all computers set to UTC time).
CamTracker version 3.9 or 4.4.0.2 (Bruker GmbH)	Official EM27/SUN software to manage the solar tracker. Older EM27/SUN with USB connection (SN127, Appendix A) use CamTracker version 3.9.
Python 3.9 (Python Software Foundation)	Python environment for various python scripts housekeeping tasks that facilitate reliable, automated measurements.
MobaXterm version 25.1 (Mobatek)	Unix command line and SSH (Secure Shell) environment to facilitate Secure File Transfer Protocol (SFTP) on Microsoft Windows.

Python scripts on the enclosure PC perform the following tasks:

- Every 10 s, append to daily files the enclosure diagnostic variables (internal temperature, humidity, pressure, warning and error flags).
- To reduce the amount of stored data, every 60 minutes delete low signal OPUS interferograms where the signal range is less than $1e-4$ (i.e. no signal due to e.g. overcast, misalignment). Retain every 50th low signal file for diagnostics purposes.
- After deleting low signal interferograms, upload the enclosure diagnostics and remaining OPUS interferograms by Secure File Transfer Protocol (SFTP) to remote SFTP server.

Every night, delete the oldest local data files if local storage capacity is above 75 %.

Code and data availability

Processing code and all EM27/SUN level 2 (L2) retrievals are available on Zenodo (DOI: 10.5281/zenodo.23674295).



755 Author contributions

DF, PP and WM planned and designed the measurement network, and WM implemented it with support from JW (EM27/SUN enclosure design and build). NP and WM developed the data analysis and visualisation. NP, PP and WM prepared the manuscript with contributions from all co-authors.

Acknowledgements

760 We thank Prof. Frank Hase and team at Karlsruhe Institute of Technology (KIT) for continued support of the Collaborative Carbon Column Observing Network (COCCON) and the PROFFAST software used in this study, and in particular Lena Feld for valuable PROFFAST discussion and support. We thank Jia Chen and Moritz Makowski at TU Munich for valuable discussion and for Pyra software development and support.

We are grateful to our station hosts and institutional partners for their essential support: John Walker and Queen Margaret
 765 University (QMUM station hosting); Fraser Walker and Port of Leith Distillery (LDST station hosting); University of Edinburgh Estates; Steve Maclaren and National Robotarium (NARO station hosting); Arthur Troy and Historic Environment Scotland. At The University of Edinburgh, we thank GeoSciences IT for compute and data transfer support, and members of Paul Palmer's group for fieldwork support.

Financial support

770 This work was funded by The University of Edinburgh School of GeoSciences and College of Science and Engineering (Prof. John Womersley), the Data-Driven Innovation (DDI) Edinburgh and South East Scotland City Region Deal (Simon Chapple, University of Edinburgh Data Technology), and the National Centre for Earth Observation (NCEO).

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