CO_2 EMISSION INVENTORY VERIFICATION THROUGH ASSIMILATION OF NETWORK DATA

Introduction

Atmospheric dispersion models are typically used in 'forward mode', meaning that source emission rates are specified and then the dispersion model is used to determine the concentration of pollutants in the air depending on the prevailing meteorology. An alternative 'inverse mode' assimilates measured concentration data enabling optimisation of emission rates and subsequent improved estimate of pollution concentrations. While emissions inventories take a long time to compile, using sensor data with this inverse approach can improve emissions estimates and hence modelled concentrations in the short term, since our knowledge can be continuously updated and the data can capture events that are not captured by inventories, such as the onset of the Covid-19 lockdown, or fugitive emissions of methane from landfill sites. The method can also be used in the longer term, for instance, to optimise or verify annual reporting of emissions of both toxic pollutants and greenhouse gases.

CERC have developed an inverse model, specifically a Bayesian-based method which combines hourly modelled pollutant concentrations from the very high resolution ADMS-Urban model (e.g. Hood et al, 2018) with hourly sensor measurements (Carruthers et al, 2019). Critical to the approach is that it can allow for the uncertainty associated with the initial (a priori) estimates of the rate of pollutant emissions from each of the emissions sources and the uncertainty of each of the sensors. It can also allow for correlations in the uncertainty between the emissions rates for different sources (for example, arising for road sources using the same emissions factors) and also between different sensors, if any. Estimating the uncertainties and correlations of these input parameters is a key part of the model set-up as the inverse model output can be highly sensitive to their values. Model output comprises a revised set of hourly concentrations at each sensor location and hourly emission rates for each source.

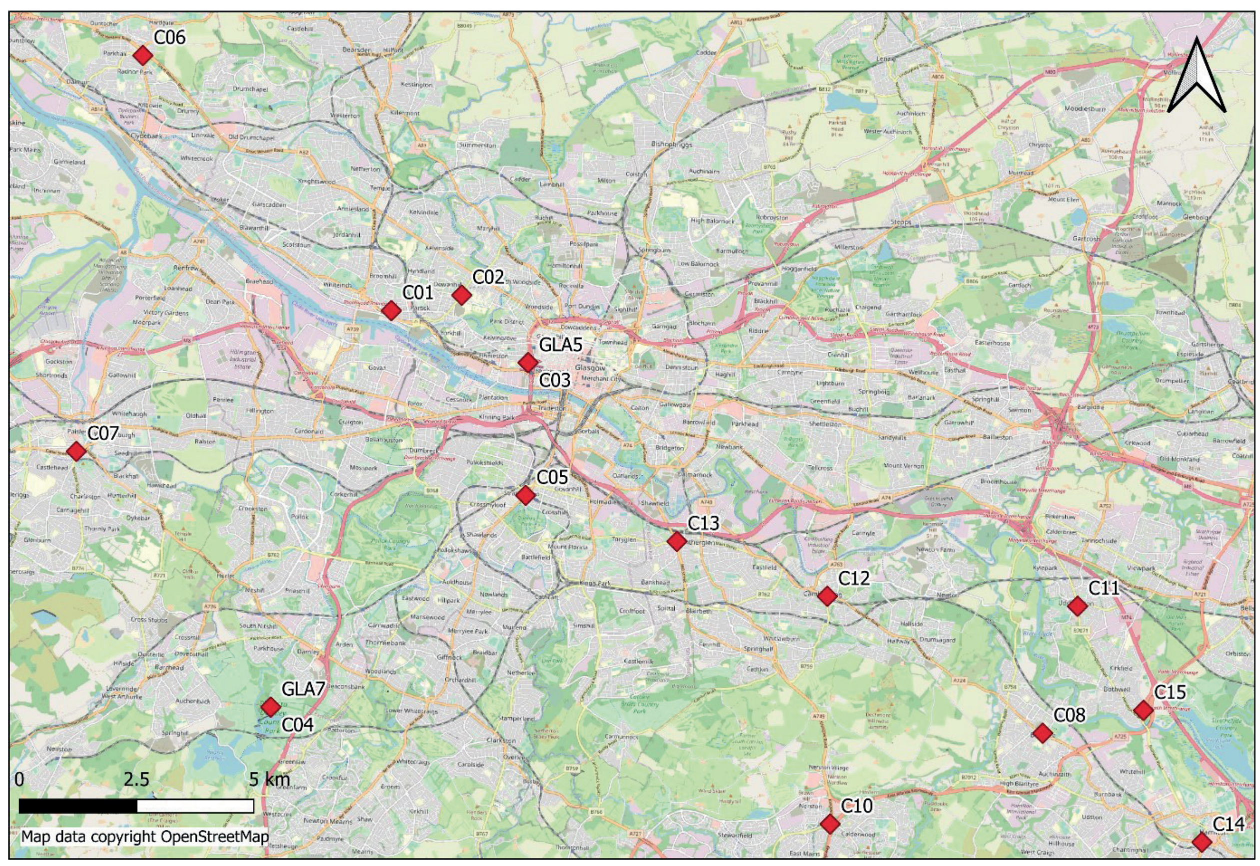

Figure 1 Map of Glasgow area showing the location of the devices measuring CO_2 concentrations: AQMesh pods (C01-C15) and the LI-COR reference grade instruments (GLA5 and GLA7). The AQMesh pods were co-located with the Scottish Air Quality Network (SAQN) reference monitors for NO_x, NO_2, PM_{10}, $PM_{2.5}$.

The inverse model has previously been applied to NO_x and NO_2 concentrations in London during the Covid-19 lockdown (Stidworthy et al, 2021). In this note we describe the application of the system to emissions of CO_2 in Greater Glasgow. The measurement period coincided with the COP26 conference in Glasgow, highlighting the critical importance of such methods on the path to Net Zero.

Method

CO_2 data was collected from 15 AQMesh sensors located at a height of 2 metres across the Glasgow area, at roadside, urban background and rural sites (Figure 1), and collocated with existing regulatory network monitors for air quality. The sensors recorded CO_2 concentrations at 1-minute intervals, which were used to calculate hourly averages. LI-COR reference monitors measuring CO_2 were also collocated at two of the sites: GLA5 (urban background) and GLA7 (rural).

CO_2 traffic emissions were estimated for main roads from DfT vehicle flow rates and vehicle splits, and COPERT 5 emissions factors using road geometry obtained from Open Roads. Road elevations were all assumed to be zero. Road widths were estimated from the road classification and refined near monitoring sites. The impacts of street canyons on dispersion were modelled using the ADMS-Urban advanced street canyon tool; street canyon geometry was determined from GeoFabrix building outlines. Emissions of other sources were represented by 1 km x 1 km gridded emissions from the National Atmospheric Emissions Inventory (Tsagatakis et al, 2020).

Hourly background CO_2 concentrations were estimated from baselines extracted from the 1-minute AQMesh data from the 'C03' gold pod sensor collocated with the LI-COR instrument on the site of the GLA5 regulatory monitor (Figure 1). Meteorological data was obtained from Bishopton weather station located approximately 20 km north west of the centre of Glasgow.

For the inversion scheme the measurement uncertainties were

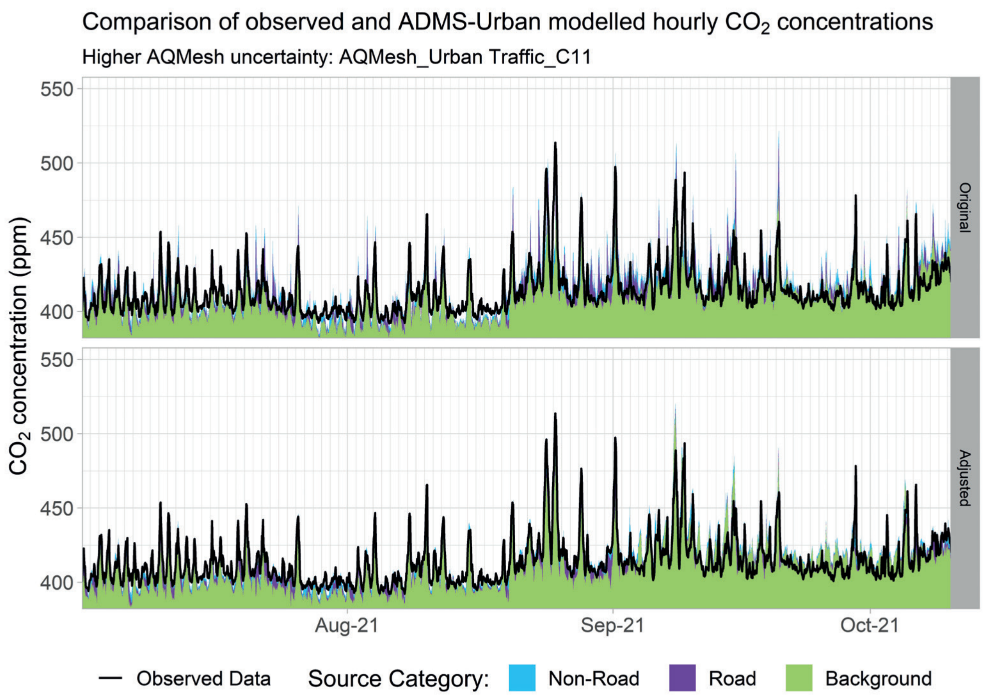

Figure 2. Comparison of observed and ADMS-Urban modelled hourly CO_2 concentrations, at urban traffic site C11.

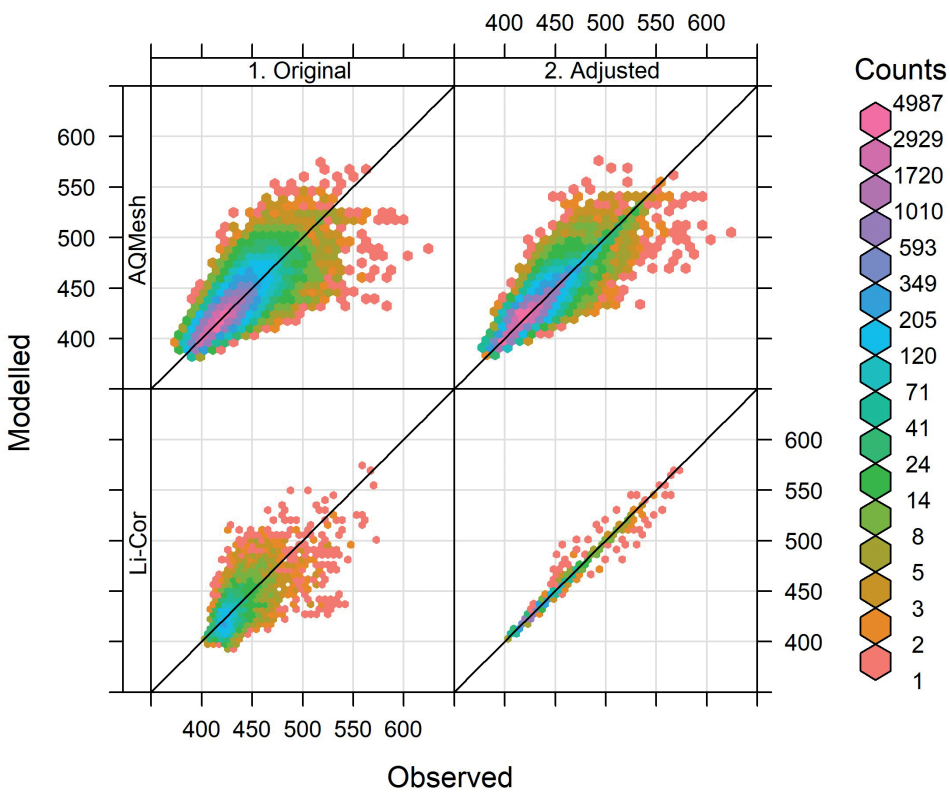

Figure 3. Scatter plots comparing monitored and modelled hourly average CO_2 concentrations (ppm) across all AQMesh and LI-COR sites before and after data assimilation.

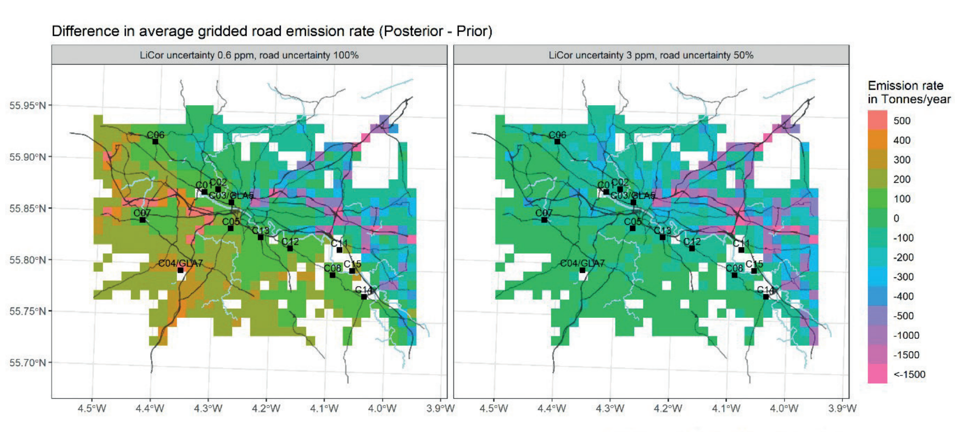

Figure 4 Difference in average gridded road emission rate (posterior – prior). Grid cells are shown where they are adjusted by the model assimilation for more than 300 hours in the period 1 July - 31 December 2021.

10ppm (AQMesh) and 0.6 ppm (LI-COR) and the background uncertainty was 10ppm, since this was derived from an AQMesh measurement. The emissions uncertainties were set as 100% and 150% of a priori emission rates for roads and other emission sources respectively. Uncertainty covariances as a percentage of uncertainty were 5% between the same measurement devices (otherwise zero), 40% between road source emission rates and 20% between other emission sources.

Results

ADMS-Urban was used in 'forward mode' to model hourly CO_2 concentrations at the sensor locations for the period 1 July until 31 December 2021. Then the inversion scheme was applied to assimilate the measurements with the modelled data, resulting in adjusted hourly CO_2 emission rates for sources across the city. As an example of the model output, Figure 2 shows a comparison between the model predictions and measurements of CO_2 concentrations at site C11 before and after the emissions were adjusted. C11 is a roadside site south east of Glasgow and west of the M74 motorway. The plots show the contribution of the background, road emissions and other emissions to total concentration. The gap between the coloured region and the black line represents the difference between the modelled and observed levels of CO_2. Using the adjusted emissions, it is seen that the gap is reduced by changes in both the road (reductions) and non-road (increases) emissions.

Figure 3 shows scatter plots comparing the monitored and modelled hourly average CO_2 concentrations across all the AQMesh and LI-COR sites before and after data assimilation. These plots show that the inversion scheme improved the model predictions, more especially at the locations of the LI-COR instruments, as these instruments have lower measurement uncertainty.

Figure 4 shows an example of the changes in emission rates arising from the application of the inversion scheme, in this case for road emissions only. It is seen (left-hand plot) that the model estimates that, overall, the a priori road emissions are somewhat overestimated (2.1%), with a larger overestimate to the east of Glasgow and a smaller underestimate to the west. Increasing the specified measurement uncertainty in the LI-COR (right plot) decreases the asymmetry in the plot between east and west, but retains a similar overall overestimate. This overestimate is to be expected as no allowance was made for the impact of COVID on traffic flows in the a priori emissions; in 2021 these were still somewhat depressed. The model suggested emissions of other sources (not shown in the figure) were overestimated by 2.9%.

Conclusions

Previous studies have applied the assimilation scheme to the adjustment of emissions of toxic air pollutants. This initial study for CO_2 emissions, demonstrates the potential of this data assimilation technique as a powerful tool for verifying the accuracy of greenhouse gas emissions inventories using ambient measurements. Overall the study suggests that in this case the CO_2 emissions inventory needed little adjustment during this period, though some features of the emissions results need further investigation (e.g. sensitivity to specified emission and measurement uncertainties, and effects of biogenic emissions and sinks).

Currently the assimilation technique treats each hour independently. Future developments will allow for correlations of emissions over different hours for example successive hours or the same hour each day. They will also refine the a priori estimates of uncertainties of emissions and measurements and their correlation, and, in the case of CO_2, take account of the large biogenic sources and sinks which are highly variable depending on both the land surface and time.

References

Carruthers D, Stidworthy A, Clarke D, Dicks J, Jones R, Leslie I, Popoola OAM and Seaton M, 2019: Urban emission inventory optimisation using sensor data, an urban air quality model and inversion techniques. International Journal of Environment and Pollution, vol. 66, issue 4, pp. 252-266, DOI: 10.1504/IJEP.2019.104878.

Hood C, MacKenzie I, Stocker J, Johnson K, Carruthers D, Vieno M and Doherty R, 2018: Air quality simulations for London using a coupled regional-to-local modelling system. Atmospheric Chemistry and Physics, vol. 18, pp. 11221-11245, DOI: 10.5194/acp-18-11221-2018.

Tsagatakis, I., Richardson, J., Evangelides, C., Pizzolato, M., Pearson, B., Passant, N. & Otto, A. (2020) UK Spatial Emissions Methodology: A report of the National Atmospheric Emission Inventory 2018. Retrieved from: https://naei.beis.gov.uk/reports/reports?report_id=958
Stidworthy A, Carruthers D, Oades M, McCosh G, Jones R, Popoola O, Mills J, Sharp F, 2022: Quantifying the Impact of COVID-19 Restrictions on Emissions using Inverse Modelling and Measurements. 21st International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, Aveiro, Portugal, September 2022. Article online

Author Contact Details

David Carruthers[1], Amy Stidworthy[1], Molly Oades[1], George McCosh[1], Rod Jones[2], Olalekan Popoola[2] and Jim Mills[3]

- **[1]CERC, 3 King's Parade, Cambridge, CB2 1SJ, UK, [2]Yusuf Hamied Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge, CB2 1EW, UK, [3]Scotswolds, UK**

Email: david.carruthers@cerc.co.uk • **Web: www.cerc.co.uk**