Continuous CH4 and δ^{13}CH4 Measurements in London Demonstrate Under-Reported Natural Gas Leakage

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Abstract

Assessment of bottom-up greenhouse gas emissions estimates through independent methods is needed to demonstrate whether reported values are accurate or if bottom-up methodologies need to be refined. Previous studies of measurements of atmospheric methane (CH4) in London revealed that inventories substantially underestimated the amount of natural gas CH4 1,2. We report atmospheric CH4 concentrations and δ13CH4 measurements from Imperial College London since early 2018 using a Picarro G2201-i analyser. Measurements from May 2019-Feb. 2020 were compared to the values simulated using the dispersion model NAME coupled with the UK national atmospheric emissions inventory, NAEI, and the global inventory, EDGAR, for emissions outside the UK. Simulations of CH4 concentration and δ13CH4 values were generated using nested NAME back-trajectories with horizontal spatial resolutions of 2 km, 10 km and 30 km. Observed concentrations were underestimated in the simulations by 12 %, and there was no correlation between the measured and simulated δ13CH4 values. CH4 from waste sources and natural gas comprised of 32.1 % and 27.5 % of the CH4 added by regional emissions. To estimate the isotopic source signatures for individual pollution events, an algorithm was created for automatically analysing measurement data by using the Keeling plot approach. Over 70 % of source signatures had values higher than -50 based on model-data comparison of δ13CH4 and on Keeling plot source signature emission both indicate that emissions due to natural gas leaks in London are being under-reported in the NAEI. These results suggest that estimates of CH4 emissions in urban areas need to be revised in the CH4 emissions inventories. 1 Helfter, C. et al. (2016), Atmospheric Chemistry and Physics, 16(16), pp. 10543-10557 2 Zazzeri, G. et al. (2017), Scientific Reports, 7(1), pp. 1-13

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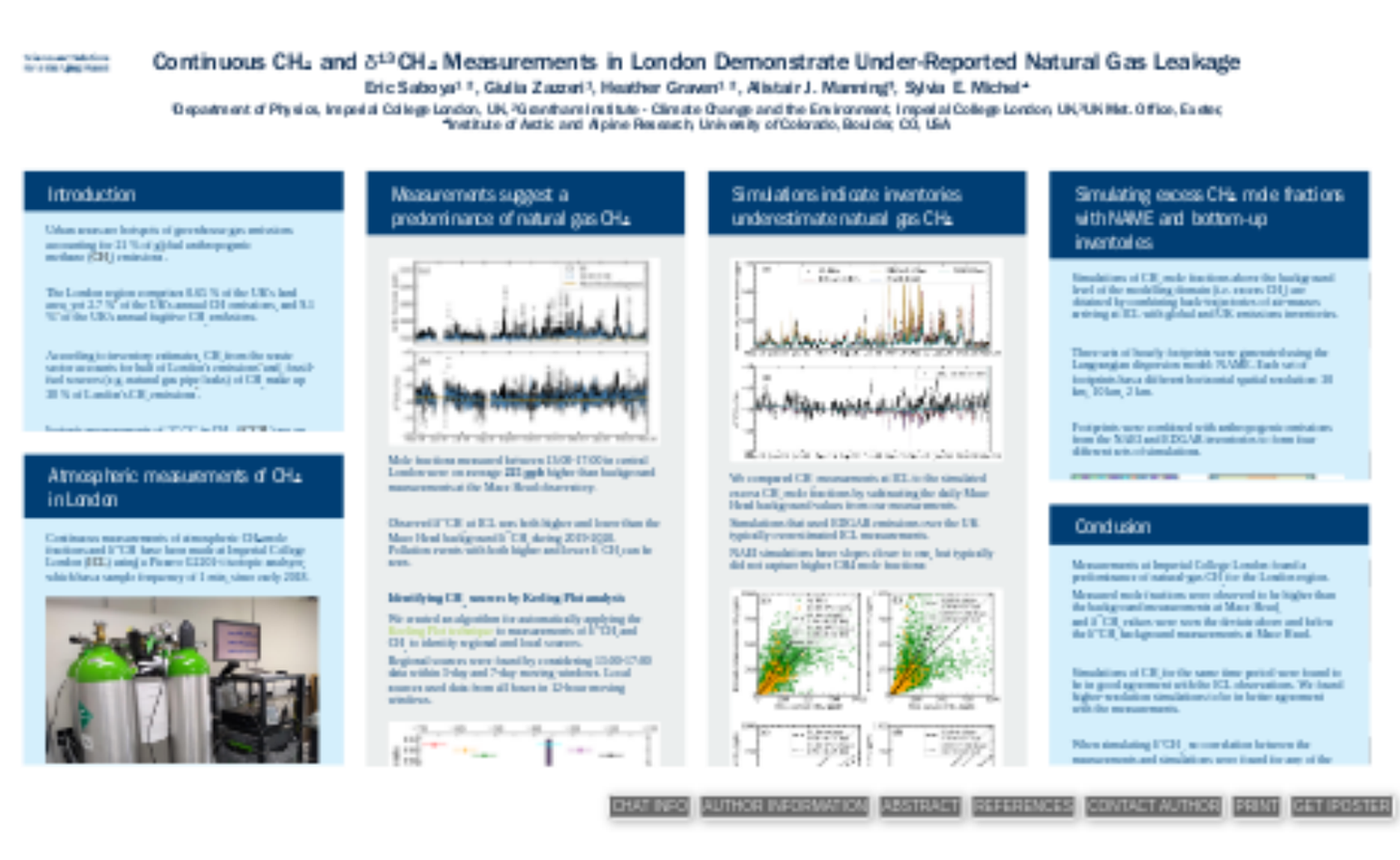

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INTRODUCTION

Urban areas are hotspots of greenhouse gas emissions accounting for 21 % of global anthropogenic methane (CH_4) emissions[1].

The London region comprises 0.65 % of the UK's land area, yet 2.7 %[2] of the UK's annual CH_4 emissions, and 9.1 %[2] of the UK's annual fugitive CH_4 emissions.

According to inventory estimates, CH_4 from the waste sector accounts for half of London's emissions[2] and, fossil-fuel sources (e.g. natural gas pipe leaks) of CH_4 make up 38 % of London's CH_4 emissions[2].

Isotopic measurements of $^{13}C/^{12}C$ in CH_4 ($\delta^{13}CH_4$) are an established means for dinstinguishing between sources[3]. Fossil-fuel sources typically have $\delta^{13}CH_4$ > -50 ‰.

The aim of this study is to develop the use of $\delta^{13}CH_4$ measurements for assessing regional-scale CH_4 emissions and sources. We use a mass-balance framework for comparing regional-scale simulations of $\delta^{13}CH_4$ and CH_4 to observations.

ATMOSPHERIC MEASUREMENTS OF CH_4 IN LONDON

Continuous measurements of atmospheric CH_4 mole fractions and $\delta^{13}CH_4$ have been made at Imperial College London (ICL) using a Picarro G2201-i isotopic analsyer, which has a sample frequency of 1 min, since early 2018.

The Graven lab at ICL where we measure CH_4 and CO_2

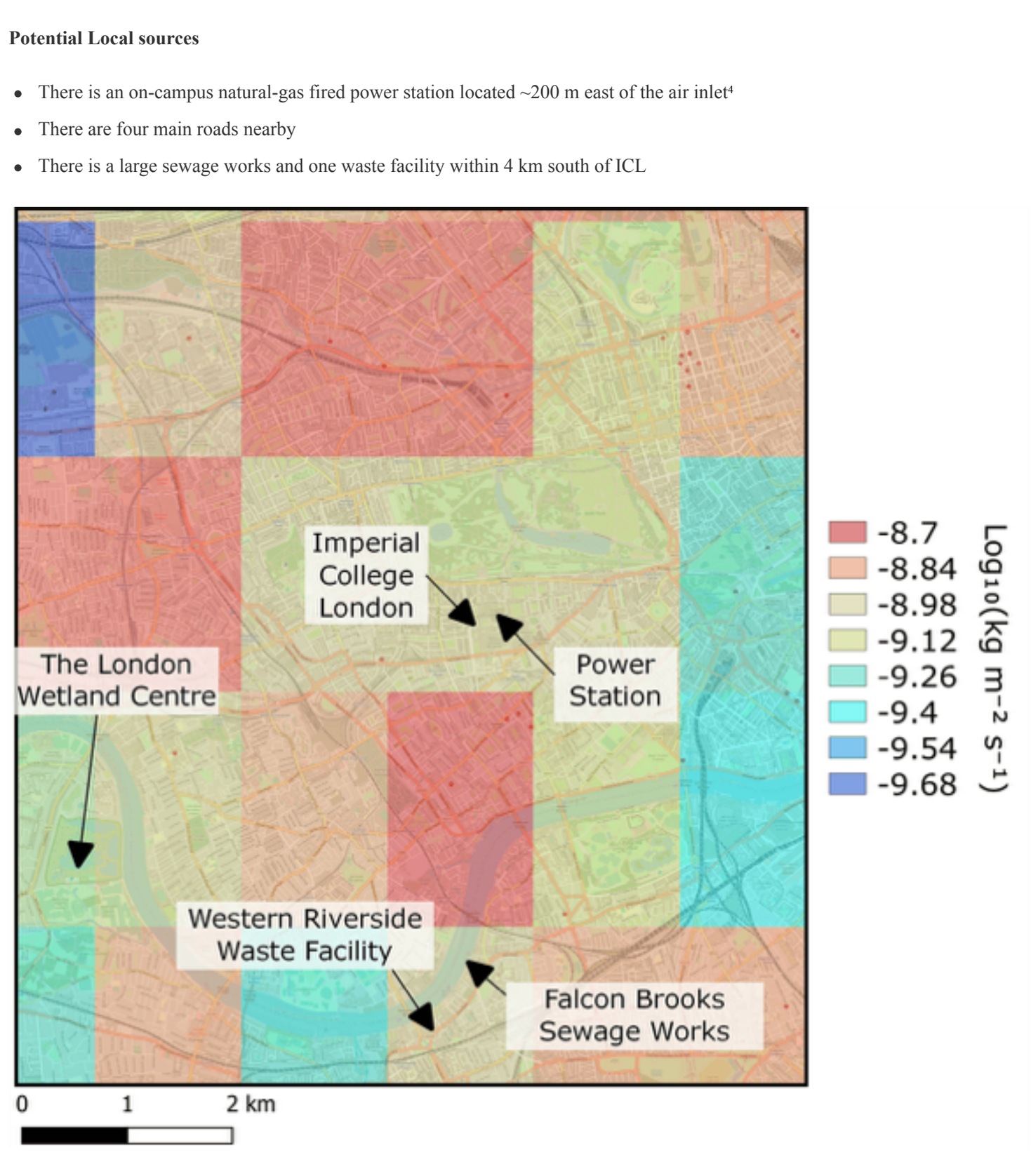
An Allan standard deviation was calculated to measure the noise response of the instrument over different averaging intervals. A precision of 0.2 ‰ (1σ) was achieved for 20-minute data.

In this study we use 20-minute averaged data.

Ambient air is sampled from an inlet mounted on a 2 m mast on the southeast corner of the Huxley building roof (26 magl, 51.4999°N, 0.1749°W).

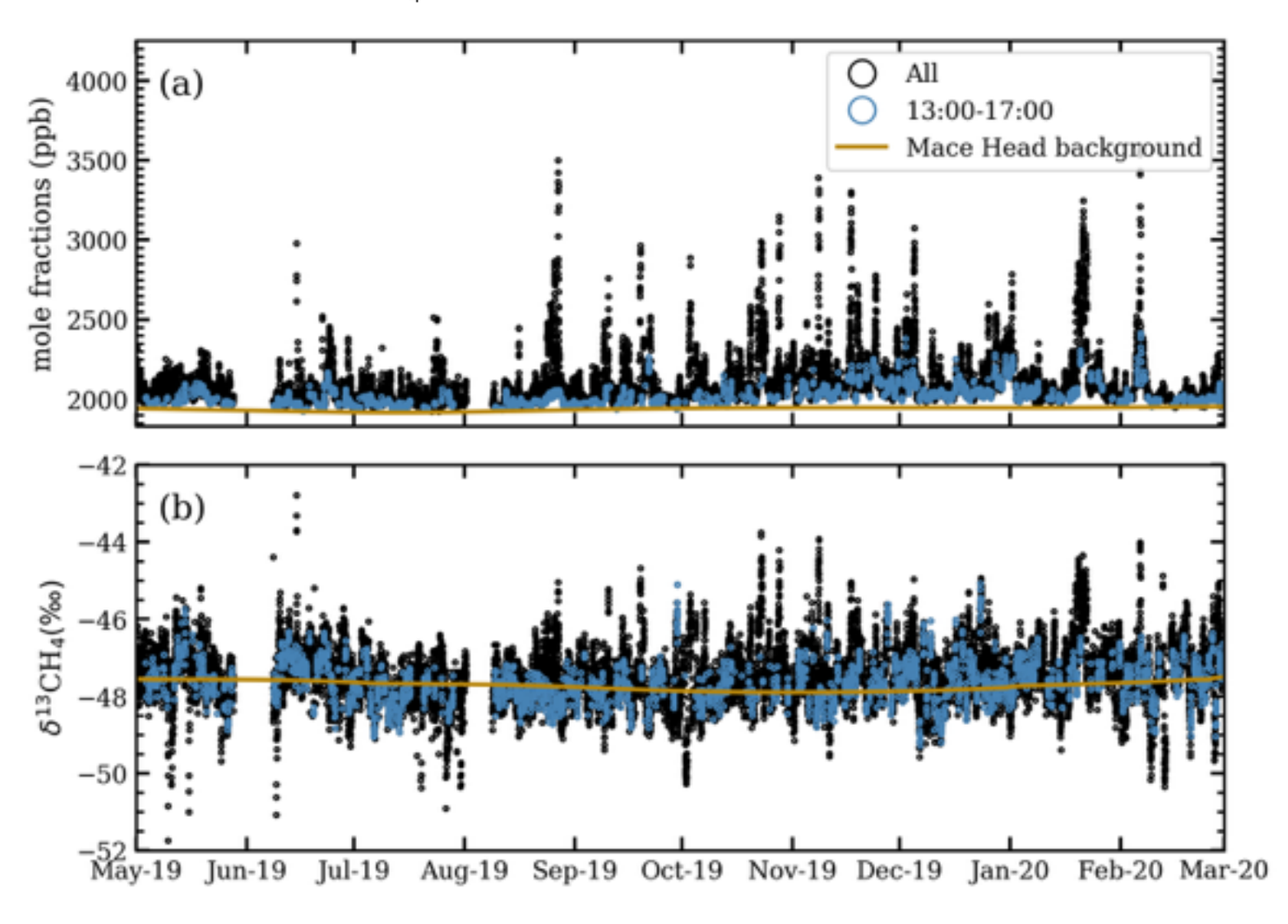
Potential Local sources

- There is an on-campus natural-gas fired power station located ~200 m east of the air inlet[4]
- There are four main roads nearby
- There is a large sewage works and one waste facility within 4 km south of ICL



Sources in the surrounding area of ICL with the NAEI emissions superimposed.

MEASUREMENTS SUGGEST A PREDOMINANCE OF NATURAL GAS CH_4



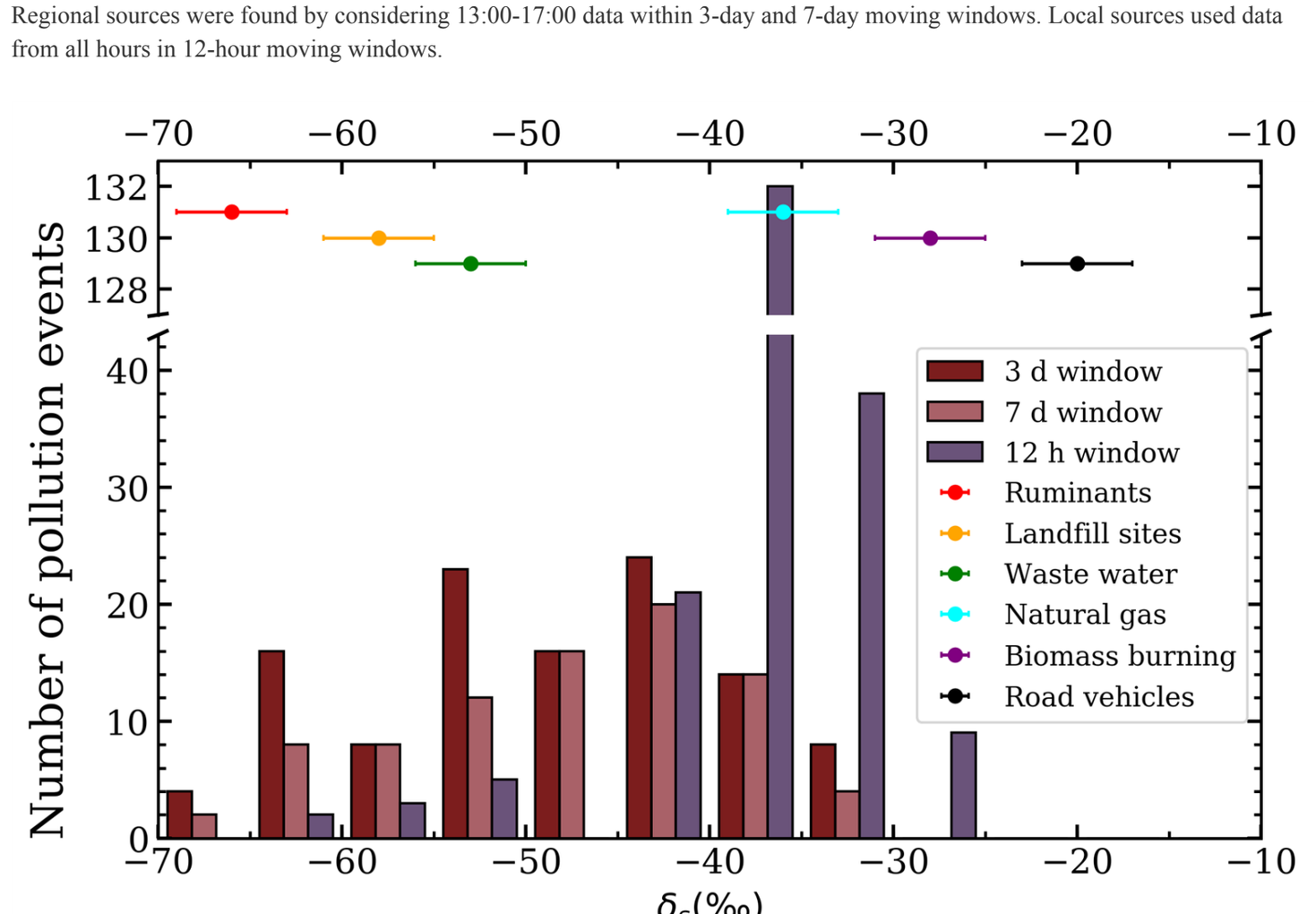
Mole fractions measured between 13:00-17:00 in central London were on average **225 ppb** higher than background measurements at the Mace Head observatory.

Observed $\delta^{13}CH_4$ at ICL was both higher and lower than the Mace Head background $\delta^{13}CH_4$ during 2019-2020. Pollution events with both higher and lower $\delta^{13}CH_4$ can be seen.

Identifying CH_4 sources by Keeling Plot analysis

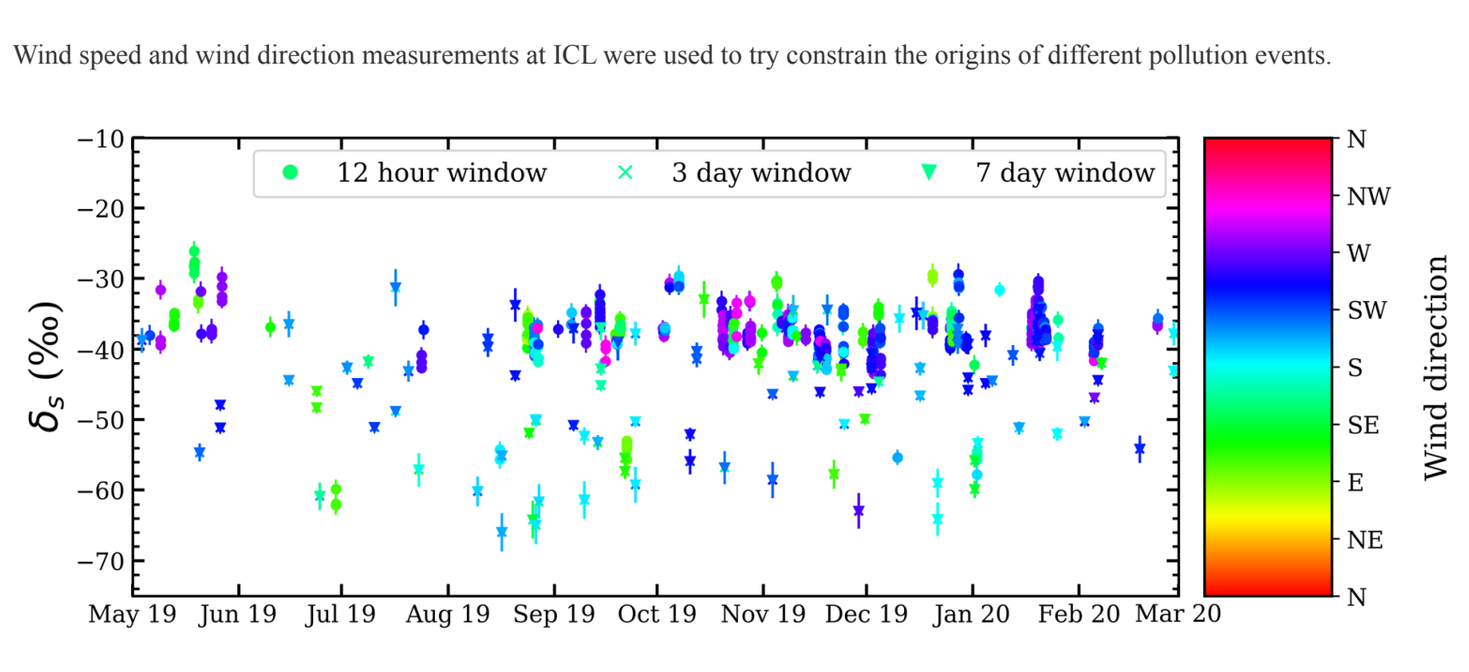
We created an algorithm for automatically applying the Keeling Plot technique to measurements of $\delta^{13}CH_4$ and CH_4 to identify regional and local sources.

Regional sources were found by considering 13:00-17:00 data within 3-day and 7-day moving windows. Local sources used data from all hours in 12-hour moving windows.



Nearly **65 %** of 13:00-17:00 isotopic source values were > -50 ‰ suggesting a predominance of natural gas CH_4.

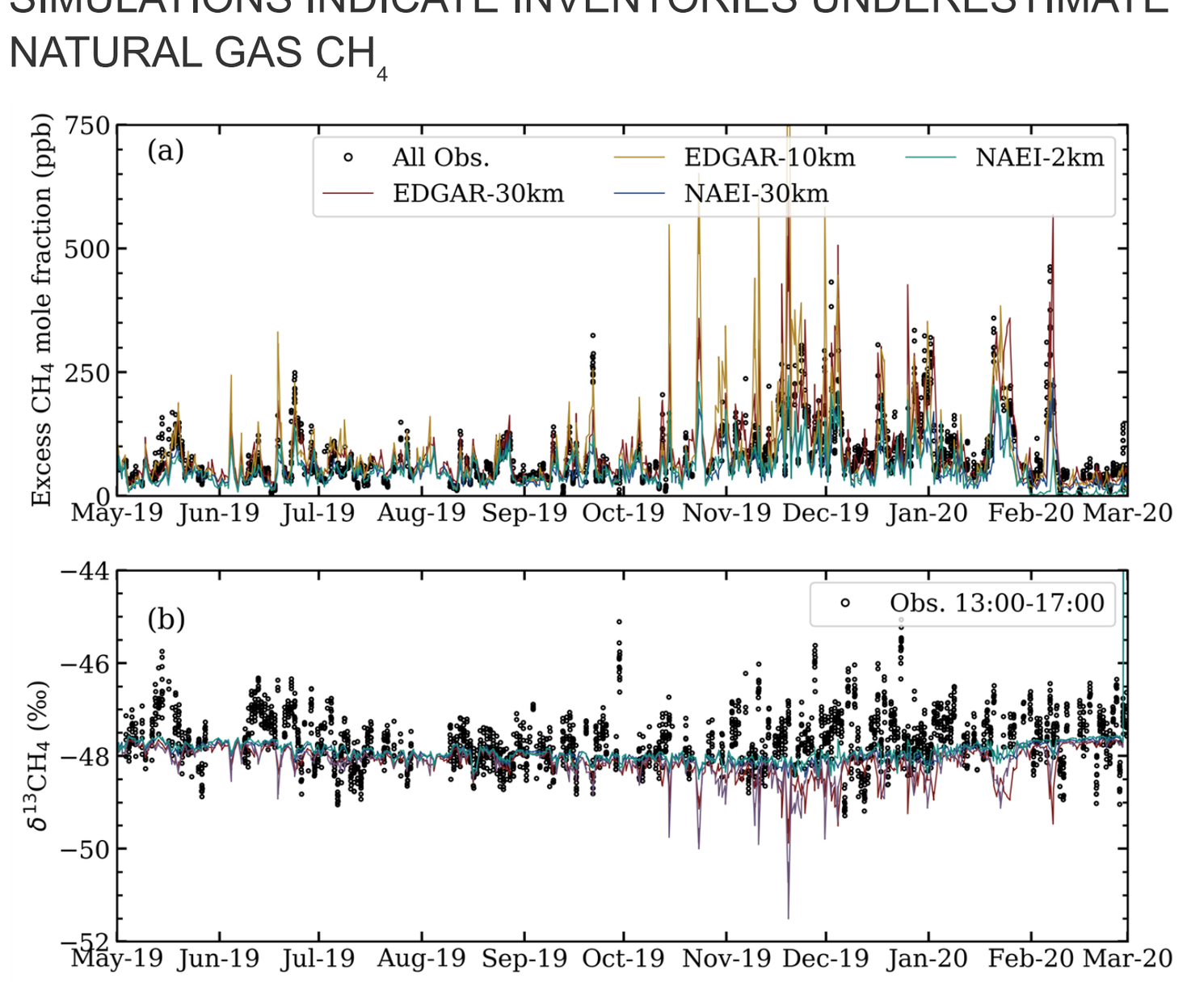
Wind speed and wind direction measurements at ICL were used to try constrain the origins of different pollution events.



We did not find consistent patterns between the wind direction and isotopic source values. This reflects the collocation and heterogeneity of sources in London.

Some events with low isotopic signatures and wind directions southerly or southwesterly may be influenced by the London Wetland Centre of by the sewage or landfill sites south of south-west of the ICL

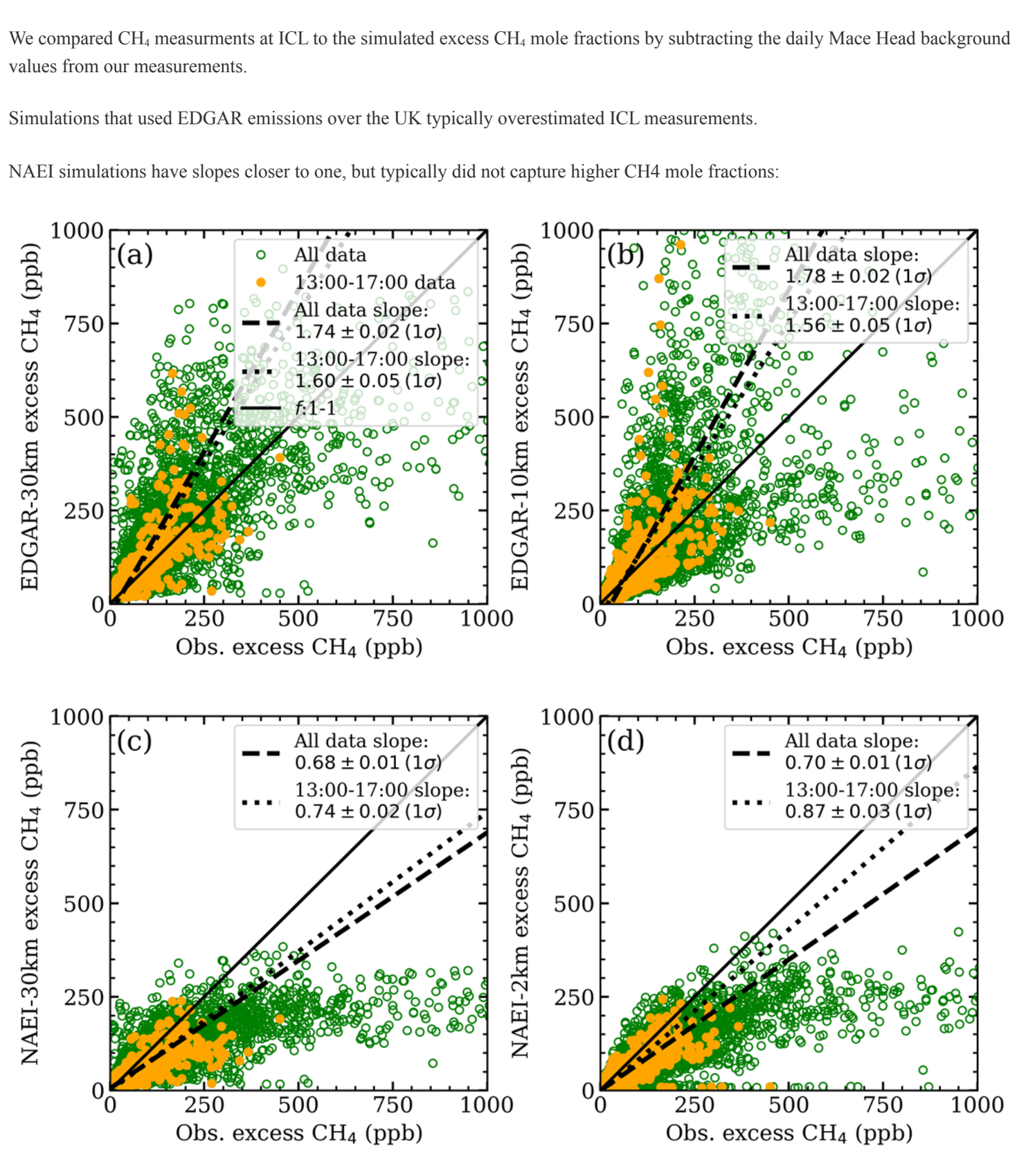
SIMULATIONS INDICATE INVENTORIES UNDERESTIMATE NATURAL GAS CH_4



We compared CH_4 measurments at ICL to the simulated excess CH_4 mole fractions by subtracting the daily Mace Head background values from our measurements.

Simulations that used EDGAR emissions over the UK typically overestimated ICL measurements.

NAEI simulations have slopes closer to one, but typically did not capture higher CH4 mole fractions:



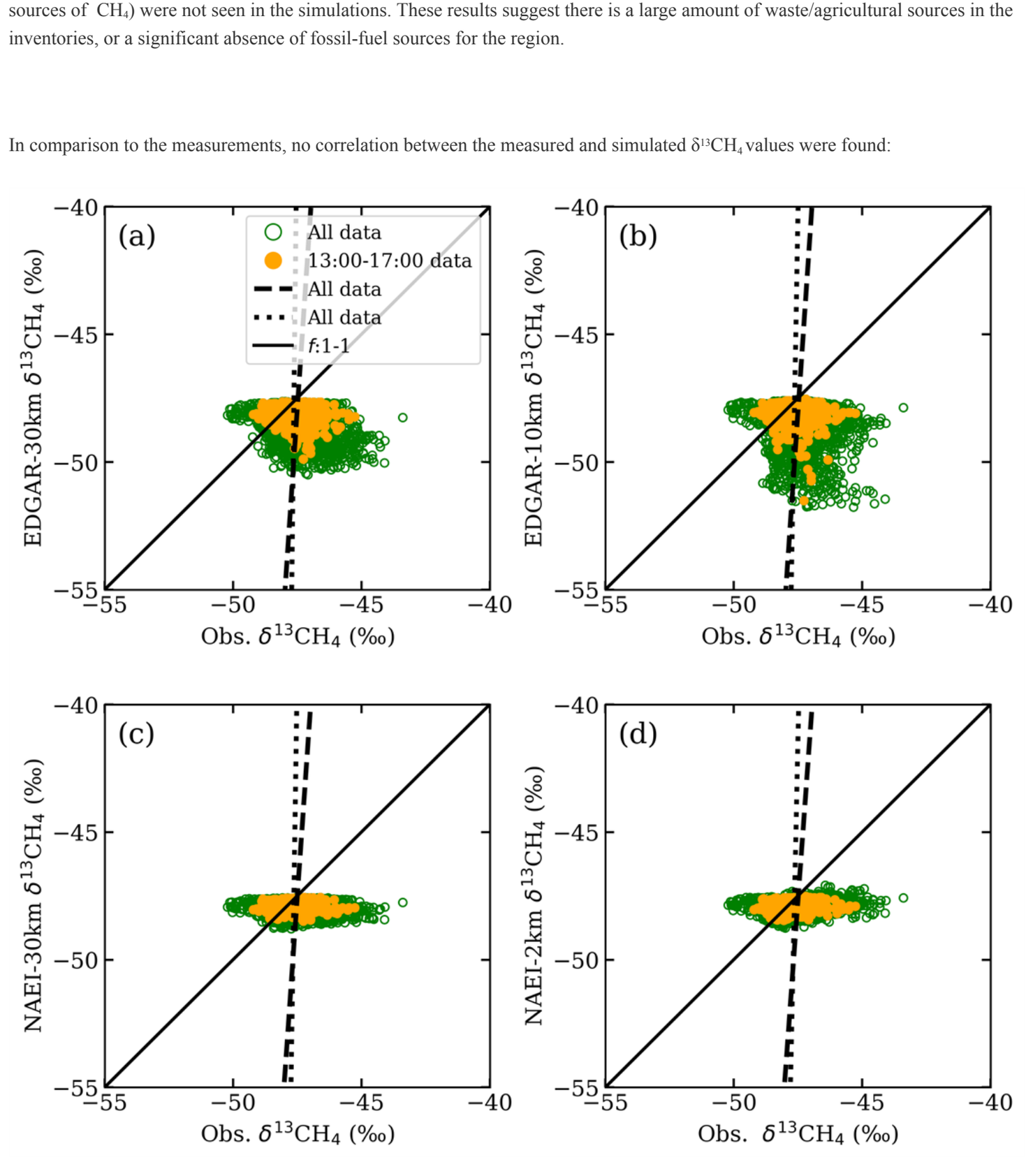
Measurement-model correlations improved when using 13:00-17:00 data. NAEI-2km 13:00-17:00 simulations have a slope of 0.87 i.e. underestimating observations by ~14.9 %

Simulated $\delta^{13}CH_4$ values

Measured $\delta^{13}CH_4$ values were compared to simulated values. The EDGAR and NAEI emissions provide sectoral estimates. Simulated CH_4 for the different sectors, and the background values were multiplied by their UK isotopic signature, summed and then divided by the total simulated CH_4 to simulate $\delta^{13}CH_4$ at ICL to compare the inventory source distributions to our measurements.

Large excursions towards more-negative $\delta^{13}CH_4$ values are seen in the simulations. Higher $\delta^{13}CH_4$ values (indicative of fossil-fuel sources of CH_4) were not seen in the simulations. These results suggest there is a large amount of waste/agricultural sources in the inventories, or a significant absence of fossil-fuel sources for the region.

In comparison to the measurements, no correlation between the measured and simulated $\delta^{13}CH_4$ values were found:



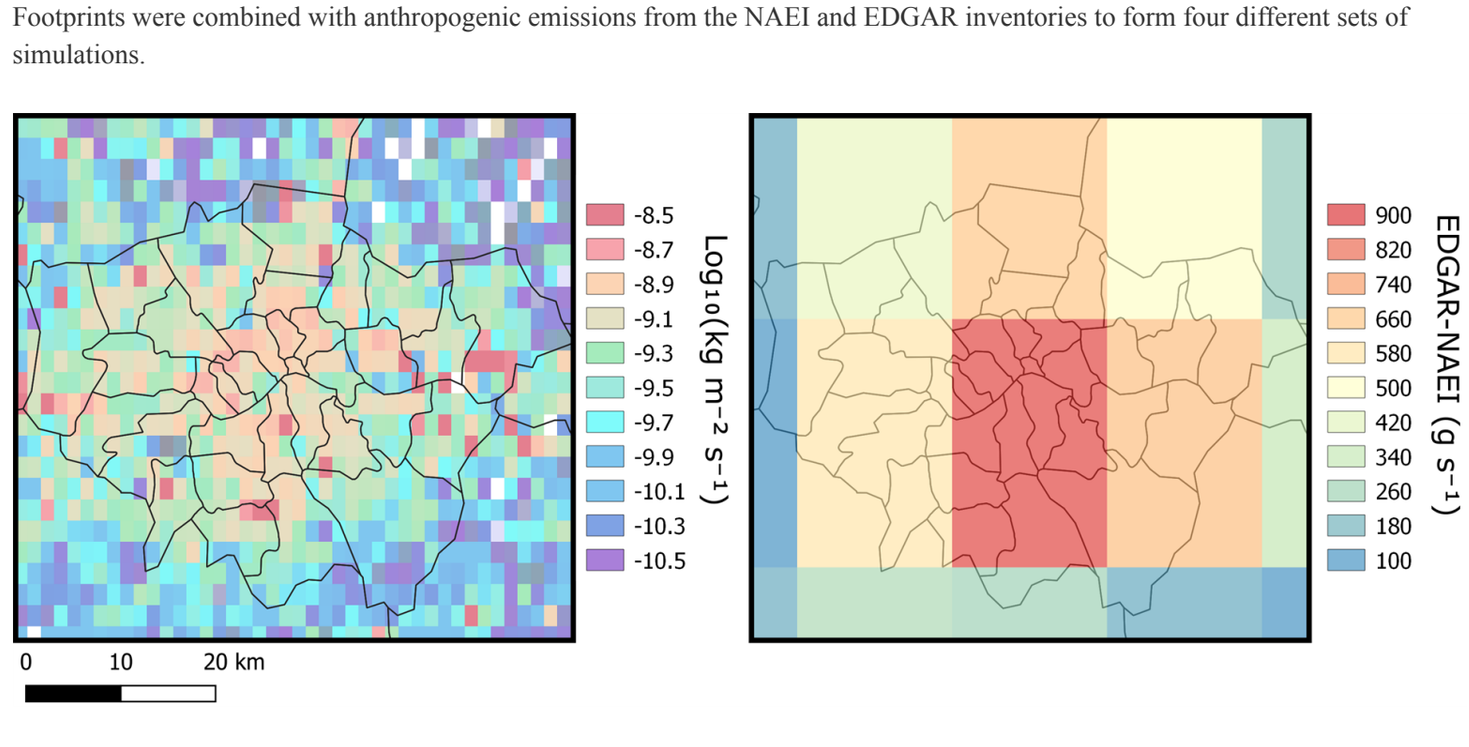
Whilst positive CH4 obs.-sim. correlations were found, the lack of $\delta^{13}CH_4$ obs.-sim. correlation suggests discrepancies in the inventory source apportionments.

SIMULATING EXCESS CH_4 MOLE FRACTIONS WITH NAME AND BOTTOM-UP INVENTORIES

Simulations of CH_4 mole fractions above the background level of the modelling domain (i.e. excess CH_4) are obtained by combining back-trajectories of air-masses arriving at ICL with global and UK emissions inventories.

Three sets of hourly footprints were generated using the Langrangian dispersion model: NAME. Each set of footprints has a different horizontal spatial resolution: 30 km, 10 km, 2 km.

Footprints were combined with anthropogenic emissions from the NAEI and EDGAR inventories to form four different sets of simulations.



On the left are the high-resolution NAEI emissions for the London region. On the right are the NAEI emissions gridded at 30 km subtracted from the EDGAR emissions for London. Over the UK, NAEI emissions are generally higher but in London EDGAR emissions are greater than the NAEI.

We considered four sets of simulations:

1. **EDGAR-30km:** 30 km footprints combined with the EDGAR v4.3.2 (2012) emissions.
2. **EDGAR-10km:** 10 km footprints over Europe nested in 30 km footprints over the rest of the modelling domain all combined with EDGAR.
3. **NAEI-30km:** NAEI emissions for the UK and EDGAR emissions for rest of the domain.
4. **NAEI-2km:** 2 km footprints combined with NAEI emissions for the UK nested in 10 km footprints for Europe, nested in 30 km footprints.

In each set of simulations we considered **wetland contributions** by combining the 2015 WetCHARTs mean extended ensemble emissions with the 30 km footprints.

CONCLUSION

Measurements at Imperial College London found a predominance of natural-gas CH_4 for the London region.

Measured mole fractions were observed to be higher than the background measurements at Mace Head, and $\delta^{13}CH_4$ values were seen the deviate above and below the $\delta^{13}CH_4$ background measurements at Mace Head.

Simulations of CH_4 for the same time period were found to be in good agreement with the ICL observations. We found higher-resolution simulations to be in better agreement with the measurements.

When simulating $\delta^{13}CH_4$, no correlation between the measurements and simulations were found for any of the simulations. This suggests discrepancies in the source apportionment of the inventories, where waste emissions are likely being overestimated and natural gas emissions are being under-reported.

Previous measurement campaigns in London[4,5] found natural gas emissions were underestimated in the national inventory. It is likely leaks from natural gas pipes were not entirely accounted.

We have demonstrated that $\delta^{13}CH_4$ measurements can be used to infer sources of CH_4 at a regional-level, and are an effective method for comparing the source allocation in emissions inventories.

AUTHOR INFORMATION

Eric Saboya is a third year PhD student based in the Space and Atmospheric Physics group at Imperial College London, UK and is part of the highly-competitive Science and Solutions of a Changing Planet DTP. Eric uses a range of measurement and modelling techniques to research the distribution and evolution of sources of methane across the UK and London. Eric also works with a team aiming to reduce the amount of air travel made by staff and students at Imperial College through policy recommendations, and has designed and lead several public-engagement activies to raise awareness of aviation emissions.

Feel free to contact me if you have any questions or just want to chat! My email is ess17 [at] ic.ac.uk

Giulia Zazzeri is a postdoctoral researcher in the Space and Atmospheric Physics group at Imperial College London, UK. Giulia primarily focuses on measuring carbon dioxide and methane, with a recent focus in radiocarbon measurements.

Heather Graven is the PI of the Carbon Cycle group in the Space and Atmospheric Physics group at Imperial College London, UK

ABSTRACT

Assessment of bottom-up greenhouse gas emissions estimates through independent methods is needed to demonstrate whether reported values are accurate or if bottom-up methodologies need to be refined. Previous studies of measurements of atmospheric methane (CH_4) in London revealed that inventories substantially underestimated the amount of natural gas CH_4 [1,2]. We report atmospheric CH_4 concentrations and $\delta^{13}CH_4$ measurements from Imperial College London since early 2018 using a Picarro G2201-i analyser. Measurements from May 2019-Feb. 2020 were compared to the values simulated using the dispersion model NAME coupled with the UK national atmospheric emissions inventory, NAEI, and the global inventory, EDGAR, for emissions outside the UK. Simulations of CH_4 concentration and $\delta^{13}CH_4$ values were generated using nested NAME back-trajectories with horizontal spatial resolutions of 2 km, 10 km and 30 km. Observed concentrations were underestimated in the simulations by 12 %, and there was no correlation between the measured and simulated $\delta^{13}CH_4$ values. CH_4 from waste sources and natural gas comprised of 32.1 % and 27.5 % of the CH_4 added by regional emissions. To estimate the isotopic source signatures for individual pollution events, an algorithm was created for automatically analysing measurement data by using the Keeling plot approach. Over 70 % of source signatures had values higher than -50 ‰, suggesting large amounts of natural gas CH_4. The analyses based on model-data comparison of $\delta^{13}CH_4$ and on Keeling plot source signature emission both indicate that emissions due to natural gas leaks in London are being under-reported in the NAEI. These results suggest that estimates of CH_4 emissions in urban areas need to be revised in the CH_4 emissions inventories.

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REFERENCES

[1] Tang, Z., Brody, S. D. Quinn, C. et al. Moving from agenda to action: Evaluating local climate change action plans, J. Environ. Plan. Manag. 53(1), 41-62, 2010.

[2] NAEI: The UK National Atmospheric Emissions Inventory, [online] Available from: https://naei.beis.gov.uk/ (Accessed 23 June 2020), 2017.

[3] Zazzeri, G., Lowry, D., Fisher, R. E., France, J. L., Lanoisellé, M., Grimmond, C. S. B. and Nisbet, E. G.: Evaluating methane inventories by isotopic analysis in the London region, Sci. Rep., 7(1), 1–13

[4] Spark s, N.and Toumi, R: Remote sampling of a CO2 point source in an urban setting. Atmos. Environ., 44(39), 5287-5294, 2010

[5] Helfter, C., Tremper, A. H., Halios, C. H., Kotthaus, S., Bjorkegren, A., Grimmond, C. S. B., Barlow, J. F. and Nemitz, E.: Spatial and temporal variability of urban fluxes of methane, carbon monoxide and carbon dioxide above London, UK, Atmos. Chem. Phys., 16(16), 10543–10557, doi:10.5194/acp-16-10543-2016, 2016.