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## Continuous CH<sub>4</sub> and δ<sup>13</sup>CH<sub>4</sub> measurements in London demonstrate under-reported natural gas leakage

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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<sub>4</sub>) in London revealed that inventories substantially underestimated the amount of natural gas CH<sub>4</sub><sup>1,2</sup>. We report atmospheric CH<sub>4</sub> concentrations and δ<sup>13</sup>CH<sub>4</sub> measurements from Imperial College London since early 2018 using a Picarro G2201-i analyser. Measurements from Sept. 2019-Oct. 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<sub>4</sub> concentration and δ<sup>13</sup>CH<sub>4</sub> 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 22 % for all data, and by 16 % when using only 13:00-17:00 data. There was no correlation between the measured and simulated δ<sup>13</sup>CH<sub>4</sub> values. On average, simulated natural gas mole fractions accounted for 28 % of the CH<sub>4</sub> added by regional emissions, and simulated water sector mole fractions accounted for 32 % of the CH<sub>4</sub> 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. Nearly 70 % of isotopic source values were higher than -50 ‰, suggesting the primary CH<sub>4</sub> sources in London are natural gas leaks. The model-data comparison of δ<sup>13</sup>CH<sub>4</sub> and Keeling plot results both indicate that emissions due to natural gas leaks in London are being underestimated in the UK NAEI and EDGAR.

<sup>1</sup> Helfter, C. et al. (2016), Atmospheric Chemistry and Physics, 16(16), pp. 10543-10557

<sup>2</sup> Zazzeri, G. et al. (2017), Scientific Reports, 7(1), pp. 1-13