Supplementary material

Title:

Methane (CH₄) sources in Krakow, Poland: insights from isotope analysis

Authors:

Malika Menoud, Carina van der Veen, Jaroslaw Necki, Jakub Bartyzel, Barbara Szénási, Mila Stanisavljević, Isabelle Pison, Philippe Bousquet, and Thomas Röckmann

Correspondance: m.menoud@uu.nl



Figure S1: Diagram of the extraction system for methane isotopic measurements in ambient air. PC: preconcentration, F: focus traps, two 10 cm stainless steal tubes (1/8" and 1/16" o.d., respectively) filled with 2 cm HayeSep D in the centre and glass beads at both ends.



Figure S2: Ratio of the peak source signature difference between original and "corrected" χ (CH₄) data, over the original signature uncertainties (standard deviation, σ). For the "corrected" χ (CH₄), we applied an offset to the IRMS data to match the CRDS data in the periods where an offset was observed. Most differences are lower than 2σ for δ^{13} C (A), and δ^{2} H (B).



Figure S3: CH₄ mole fraction hourly averages during the fall (September 14 to November 15, 2018). Size of shaded area is 1σ .







Figure S5: Miller-Tans plots using all the **A**. δ^{13} C-CH₄ and **B**. δ^{2} H-CH₄ data in ambient air collected during the measurement period. The red points show when there was a mismatch in the mole fractions between the IRMS and CRDS instruments, and the black points show the background data (χ (CH₄) < 1986.0 ppb, i.e. 10th lower percentile).



Figure S6: Dual isotope diagrams with signature ranges of specific CH_4 formation processes (background grey patterns), reproduced from Milkov and Etiope (2018). **A.** Source signatures of the sampled sites around Krakow and in the USCB (Kotarba 2001, Kotarba and Pluta 2009 and Kedzior et al. 2013). **B.** Source signatures of the emission peaks measured in ambient air. Red points: mismatch in the mole fractions between the IRMS and CRDS instruments, which peak isotopic signatures did not significantly differ (we retained all the data).



Figure S7: Reported CH₄ emission rates over the domain used in CHIMERE, from two inventories: EDGAR v5.0 (left columns) and CAMS-REG v4.2 (right column).