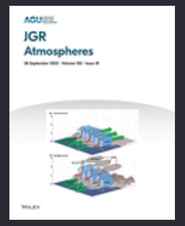

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Methane Sources in Cluj-Napoca, Romania: Insights From Isotopic Analysis

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Key Points:

- Eight months of continuous measurements of methane mole fraction and stable isotopes ($\delta^{13}C$, $\delta^{2}H$) in an Eastern European urban environment
- Source mix was dominated by fossil emissions (57%), plus biogenic emissions (38%) in summer and pyrogenic sources in winter (5%)
- Atmospheric Lagrangian particle dispersion model and used emission inventory lack spatial resolution to successfully reproduce observations

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract Increased emissions of methane (CH_4) have contributed 0.3–0.8°C to global temperature rise since preindustrial times. Reducing these emissions is crucial to mitigate climate change. Measurements of the isotopic composition of CH_4 ($\delta^{13}C$ and $\delta^{2}H$) can be used to distinguish various sources of CH_4. This study reports continuous measurements of CH_4, $\delta^{13}C$ and $\delta^{2}H$ for 8 months in Cluj-Napoca, Romania. An automated extraction and a purification system, coupled to an isotope ratio mass spectrometer alternately measured $\delta^{13}C$ and $\delta^{2}H$ of CH_4 with 20-min time resolution at the campus of the Babeş-Bolyai University. In addition, point source samples were measured to isotopically characterize CH_4 sources in the region. The time series show regular CH_4 elevations during the night, occasionally superimposed on multiday events. From these elevations, we identified three main CH_4 emission categories: Transylvanian biogenic gas (75%); biogenic emissions from rivers and wastewater (38%), predominantly observed during the summer; and a third source emitting ^{13}C-enriched CH_4 in winter, likely of pyrogenic origin (5%). We simulated the CH_4 mole fraction at the measurement site using Lagrangian footprints generated from the FLEXPART-COSMO model convolved with emissions from the TNO-CoCO2 inventory. The simulations show that the emission inventory is not granular enough to represent the city center. The strong underestimation in winter suggests that the emission inventory did not include the pyrogenic winter source. When the model accurately estimated the CH_4 mole fraction, it also predicted the isotopic compositions well.