Detection and quantification of urban methane emissions in Heidelberg (Germany) using mobile and isotope measurements

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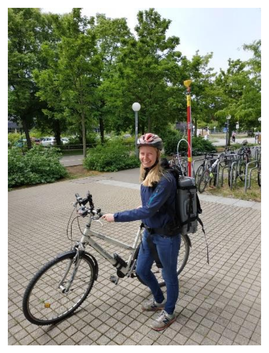
Introduction

The detection and quantification of urban CH_4 emissions is still a challenge due to the complex and heterogeneous distribution of emitters, i.e. leakages in the natural gas grid, waste and wastewater treatment, and biogas plants. Mobile measurements at street level, with cars or bicycles, are a good way to detect and quantify methane emitters, and isotope measurements can help to identify the source type.
In this study we present stationary $\delta^{13}CH_4$ measurements and a careful analysis of the source partitioning in Heidelberg (Germany) together with mobile CH_4 measurements at street level. CH_4 emissions from the natural gas network as well as emissions from the wastewater treatment plant were determined using a Gaussian plume. CH_4 flux chamber measurements were carried out at selected manholes in the sewerage system.

Methods and Sites



In-situ Measurements at Heidelberg Site

- CRDS system for ^{13}C in CH_4 and CO_2 analysis (Picarro G2201-i)
- Regular calibration (every 5 hours)
- Target precision over 6 years: is 0.2 ppb for CH_4 and 0.3 ‰ for $\delta(^{13}CH_4)$ (see Fig. A2).

Mobile Measurement equipment

- Mobile measurements with measuring devices in a van, on a bicycle or on foot
- OF-CEAS analyser for CH_4/CO_2 (LICOR LI-7810)
- 2D weather station installed on a roof system
- 3D sonic anemometer
- Daily calibration with one standard gas

Studied Sites for CH_4 city measurements

- Heidelberg, Schwetzingen, (street level, car and bicycle)

Point sources measurements:

- waste water treatment plants (Heidelberg, Mannheim)

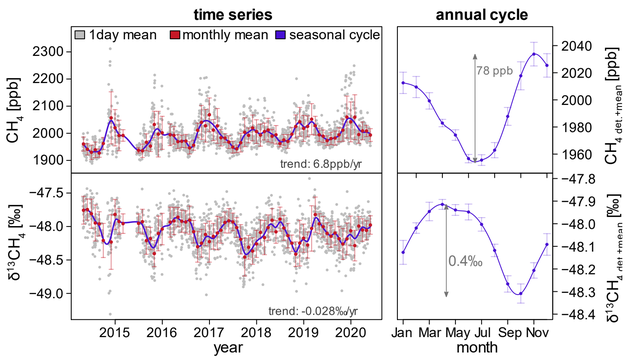
Chamber Measurements

- manholes in the sewerage system (Heidelberg)

Release Experiments:

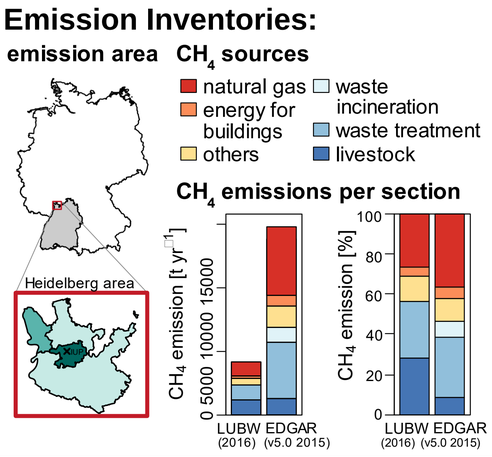
- CH_4 release Experiments to prove methods for Gaussian plume models and city measurements

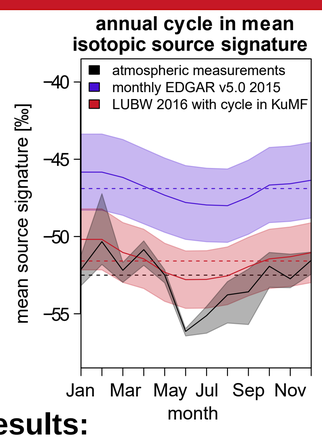
In-situ $\delta^{13}CH_4$ measurement in Heidelberg to study source contribution



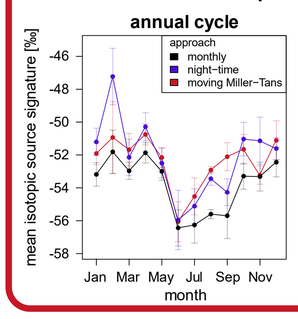
Atmospheric CH_4 and $\delta^{13}CH_4$ show seasonal variations. Annual cycles depend on the height of the boundary layer, but also on seasonal variations in background and in regional and local CH_4 emissions.

Emission Inventories:





Mean and $\delta^{13}CH_4$ Source signature (Keeling)



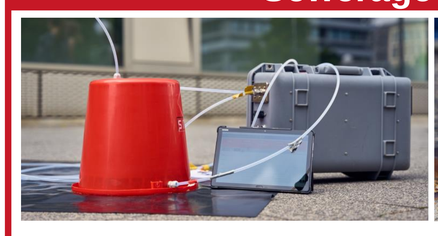
Three approaches:
- monthly (all minute values)
- night-time (17 and 7 CET)
- moving Keeling (1-12h)

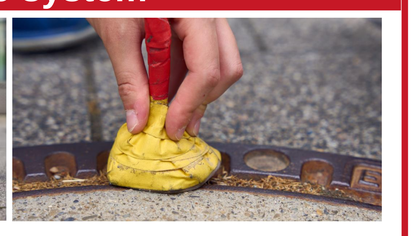
⇒ average around -53 ‰

⇒ seasonal cycle with -52 ‰ in winter and -56 ‰ in summer

Results:
EDGAR: on average -47 ±3 ‰
→ overestimation of emissions from enriched sources such as waste incineration + natural gas
LUBW: on average -52 ±2 ‰
→ represent the mean isotopic signature well
Hoheisel & Schmidt [2024]

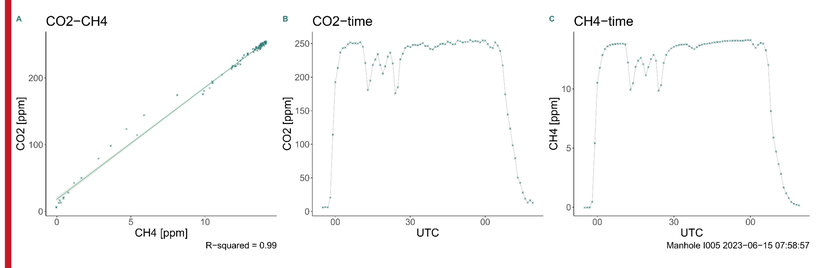
Sewerage system





Measurement sites and methods

- Flux chamber with in-situ CH_4 measurements
- CH_4 enhancement measurements above manholes
- CO_2/CH_4 ratio and $\delta^{13}CH_4$ source signature to attribute between biogenic and fossil fuel (natural gas leakage)
- 55 manholes in three Heidelberg districts

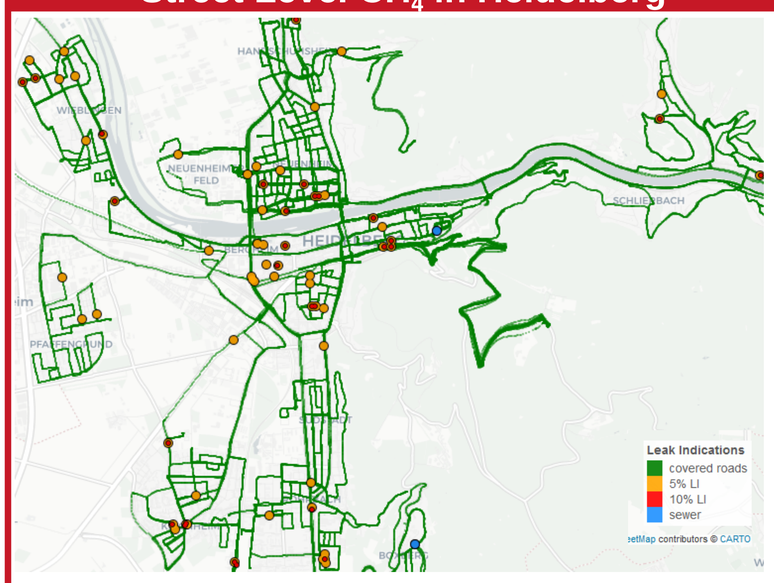


Results:

- Mean CH_4 enhancements @ manholes: 12 ± 21 ppm
- 59 % of the manholes: CH_4 enhancements <0.2 ppm
- all measured CH_4 samples origin from a biogenic source
- flux chamber measurement at selected manholes showed CH_4 fluxes below 500 mg CH_4 /d.
- Mean CH_4 flux 241±9 mg CH_4 /d

=> **0.74 t CH_4/a**
(estimated for 15000 manholes in Heidelberg)

Street Level CH_4 in Heidelberg



Methods

- We adapted the method of Weller (2019) for smaller cities with narrow streets with CH_4 release experiments (Wietzel and Schmidt 2023)
- CH_4/CO_2 ratio to distinguish natural gas and sewer
- 24 measurement days, 588 total driven km

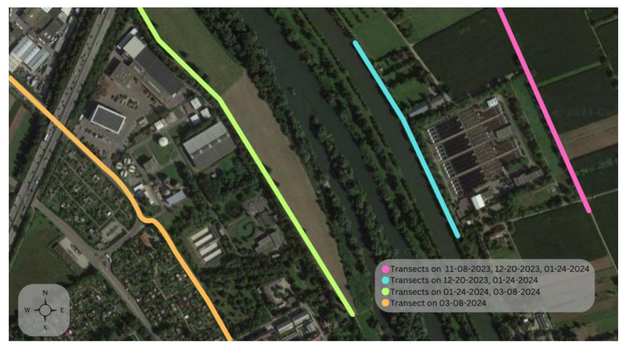
Results

- 70 Leak indication (LI) with CH_4 > 5% above BG
- 2 LIs from sewer, 68 natural gas leaks
- 2.1 km covered per LI
- Heidelberg: **42 t CH_4/a** => 0.26 kg/a per capita

Comparison to emission inventory
EDGAR (2015) -> 197.8 t CH_4/a natural gas
LUBW (2016) -> 201.4 t CH_4/a natural gas

WWTP Heidelberg

Studied Sites and methods



- Mobile CH_4 measurements at 4 waste water treatment plants (WWTP) close to Heidelberg
- CH_4 emission rates calculated with Gaussian Plume model

Results of studied WWTP:

WWTP	P.E	Emission (tCH_4/a)	Emission Factor [kgCH4/a/P.E]
HEIDELBERG	250000	60 ± 12	0.24 ± 0.05
SANDHAUSEN	15000	28 ± 7	1.90 ± 0.50
WEINHEIM	188000	7 ± 4	0.04 ± 0.03
MANNHEIM	570000	154 ± 16	0.27 ± 0.03

⇒ The WWTP in Heidelberg result in an annual CH_4 emission of **60 t CH_4/a**

Emission Factor national emission inventory from UBA
0.26 kg CH_4/a per capita

Measured CH_4 emission factors in this study:
0.04 - 1.9 kg kg CH_4/a per capita

=> Further measurements should be performed at smaller WWTP like Sandhausen.

References and Acknowledgements

- **Wietzel and Schmidt [2023]**:Methane emission mapping and quantification in two medium-sized cities in Germany: Heidelberg and Schwetzingen, Atmos. Environ. X, 20, 100228
- **Hoheisel and Schmidt [2024]:** Six years of continuous carbon isotope composition measurements of methane in Heidelberg (Germany) – a study of source contributions and comparison to emission inventories, Atmos. Chem. Phys., 24, 2951–2969, https://doi.org/10.5194/acp-24-2951-2024, 2024.
- **Weller et al. [2019];** An open source algorithm to detect natural gas leaks from mobile methane survey data.

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Summary and Outlook

- Long term in-situ $\delta^{13}CH_4$ measurements can help to improve and validate emission inventories on regional and city scale
- Mobile street level CH_4 measurements show smaller CH_4 emission rates from natural gas infrastructure compared to emission inventories
- CH_4 emission rates from WWTS are in a similar range compared to natural gas infrastructure

The sewerage system in Heidelberg is a small, negligible CH_4 emitter.

⇒ Further work will be carried out in the city of Mannheim and smaller cities in the Rhine- valley

⇒ More representative measurements on sewerage system and WWTP