

BULETINUL INSTITUTULUI POLITEHNIC DIN IAȘI

Publicat de

Universitatea Tehnică „Gheorghe Asachi” din Iași

Volumul 70 (74), Numărul 1, 2024

Secția

CHIMIE și INGINERIE CHIMICĂ

DOI: 10.5281/zenodo.11145577

DETECTION OF HIGH CH₄ CONCENTRATIONS AT NATURAL GAS END-USE DISTRIBUTION NETWORK IN URBAN AREAS

BY

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Received: February 16, 2024

Accepted for publication: March 26, 2024

Abstract. Since the industrial revolution, CH₄ emissions have increased by 150%. Urban Areas are responsible for approximately 60% of these emissions, mainly coming from anthropogenic activities. Besides, urbanization caused changes in land use and reduced CH₄ sinks. The sources of CH₄ emissions in Urban Areas still have a high degree of uncertainty. Recent studies have stated that leaks from natural gas distribution networks are significant sources of CH₄ in the atmosphere, and they also represent a potential loss of energy resources. However, emissions from end-use natural gas networks are poorly explored in the literature. The main contributors in the Romanian CH₄ budget are the agriculture and energy sectors. Over the period 1989–2000, methane emissions rate decreased by 34% due to sectoral changes in agriculture and fossil fuels. Nevertheless, the Romanian national inventory doesn't report CH₄ emissions from urban areas.

This study investigates CH₄ concentration from the end-use natural gas distribution networks in Cluj-Napoca, the second-largest city in Romania in terms of population. These points can be identified as part of the natural gas distribution networks that serve natural gas to end users for gas consumption. The detected points were pipeline junctions and natural gas meters. The estimation of CH₄ concentration in the atmosphere was carried out based on a laser CH₄ sensor Tunable Diode Laser Absorption Spectroscopy (TDLAS) with high measuring accuracy of 0.1 ppmv. This CH₄ concentration detection was performed from

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December 2022 to January 2023 at 74 natural gas end-use points. The determination of whether to represent leaks or not was quite after estimating the background level in the city and comparing the obtained concentrations with this background. This study has revealed that 76% of the detected end-use natural gas distribution points have gas leaks and represent continuous contributors to CH₄ annual budget. However, this contribution to the annual budget should be estimated. Moreover, the results of this study indicate the presence of high leaks from natural gas end-use points in urban areas. They suggest in-depth investigation and allocation of all natural gas leaks at the end-use points, in order to take certain reduction measures regarding CH₄ mitigation.

Keywords: greenhouse gas, methane, atmosphere, urban areas, natural gas end-use networks, gas leaks.

1. Introduction

Greenhouse gases (GHG) are atmospheric gases that have global warming potential (GWP) in that they have the ability to absorb terrestrial radiation; thus, they increase the atmospheric temperature (IPCC, 2013a). GHGs include various types of gases, among them carbon dioxide (CO₂), methane (CH₄), water vapor (H₂O), nitrous oxide (N₂O), and some other synthetic chemicals. Basically, the GHGs are considered beneficial for Earth's heat balance. But with the significant increase in anthropogenic activities since the industrial era, their emissions have excessively grown (EPA, 2022).

Methane, as a major GHG, is emitted from natural and anthropogenic sources. Among its natural sources are wetlands, oceans, lakes, and wild animals. The anthropogenic ones can be landfills, the combustion of fossil fuels, agriculture, transportation, or waste water treatment (IPCC, 2013b). Methane is the second most effective GHG, with more than 20% more highly increasing radiative effects in the atmosphere than CO₂ (Cuna *et al.*, 2008). In addition, it has a short lifetime in the atmosphere, up to 12 years; any significant decline in CH₄ emissions, especially from fossil fuels, agriculture, and waste sectors as major emission sources, would mean a reduction in global temperature by 1.5–2°C (UNEP, 2021).

Urbanization and land use change have influenced CH₄ concentration in the ecosystem (Wong *et al.*, 2020). Therefore, estimating CH₄ emissions is the first step towards understanding their sources in order to take appropriate mitigation measures afterwards. Recent studies in urban areas found that CH₄ emissions are being underestimated by national inventories in these areas and the degree of uncertainty is quite high in this concern (Cambaliza *et al.*, 2015; Foster *et al.*, 2017).

In Romania, the main estimated contributors to the Romanian CH₄ budget between 1989 and 2018 were agriculture and fossil fuels. Between 1989–2000, the CH₄ emissions rate decreased by 34% due to changes in these

two sectors (ANPM, 2020). Nevertheless, the Romanian national inventory still didn't report emissions categorized by urban areas.

Urban areas are important sources of CH₄ emissions that come from natural gas (NG) distribution systems, especially from old natural gas (NG) distribution infrastructure. However, the data available in this context is quite limited, which makes the process of identifying these emissions and allocating them an important mission for achieving the purpose of continuously combating against global warming effects (von Fischer *et al.*, 2017). Recent studies have estimated significant CH₄ emissions in urban areas attributed to NG leaks (Cambaliza *et al.*, 2015; Foster *et al.*, 2017; McKain *et al.*, 2015; Phillips *et al.*, 2013; von Fischer *et al.*, 2017; Wunch *et al.*, 2016; Zazzeri *et al.*, 2015).

Particularly, leaks from NG networks are potential contributors to CH₄ fluxes in the urban atmosphere (von Fischer *et al.*, 2017; Zazzeri *et al.*, 2015). Nevertheless, data limitations represent a big challenge for understanding the actual contribution of urban areas to the CH₄ annual budget (Heimbürger *et al.*, 2017; Plant *et al.*, 2019). For example, anthropogenic emissions from NG network failure are the most important sources for CH₄ in the USA. The obtained results showed a signature of natural gas sources. By detecting these emissions and identifying their sources, a wide range of advantages can be brought to the atmosphere and the economy (Phillips *et al.*, 2013).

On a national scale, determining exactly where the points of system failure and would definitely guide future maintenance projects for solving these kinds of leakages, which would then cut the loss of gas resources and protect the atmosphere from the burden of emissions. Our study aims to preliminarily determine if the end-use points in the urban area of Cluj-Napoca, the second largest city in Romania in terms of population, represent leaking points and if they also contribute to CH₄ emissions to the urban atmosphere. Accordingly, future research will be based on estimating the contribution of these NG end-use points, in order to provide decision-makers with indicators for appropriately applying mitigation measures on a city-scale.

2. Methods and Materials

This study investigates CH₄ atmospheric concentration at the NG end-use points network in Cluj-Napoca, the second largest city in Romania in terms of population. These end-use points can be identified as part of the NG distribution networks that serve NG to end users for gas consumption. Figure 1 illustrates the location of these points, in which were classified into two areas: the city center and a residential neighborhood. The detected points were pipeline junctions, NG meters, and regulators, as shown in Fig. 2.

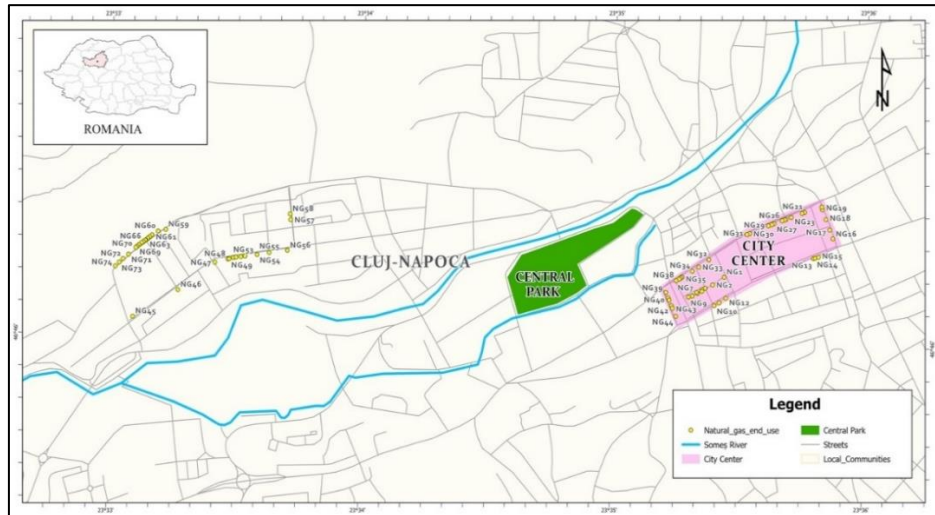


Fig. 1 – The location of NG end-use points at which the estimation of CH₄ concentrations was carried out.



Fig. 2 – Example of the end-use points at which CH₄ concentration was estimated from the city center area and the residential area.

The NG end-use points were selected in two different areas in order to understand the situation of NG end-use points from different locations, especially since the city center area is characterized by restaurant activities based on high gas consumption. The residential area is characterized mostly by its use for households.

The selection of these NG end-use points was constrained by their accessibility from the street in the two areas. The estimation of CH₄ concentration in the atmosphere was carried out based on laser CH₄ sensors via Tunable Diode Laser Absorption Spectroscopy (TDLAS), which records gas concentration parts per million in volume (ppmv) every second and precisely up to 0.1 ppmv. The survey was performed between December 2022 and January 2023 at 74 NG end-use points.

Moreover, for the purpose of evaluating the concentration of CH₄ and to characterize the values for whether there were leaks or not, a background level of CH₄ was also estimated. The background value was determined by continuously conducting street-level atmospheric concentration measurements in the primary streets and in the central park area CH₄ via TDLAS, and then the mean value represents the background.

3. Results and Discussion

The atmospheric concentration of CH₄ was determined at each of the 74 end-use points, as shown in Fig. 3 (a, b). The results of the estimation are represented in Fig. 4, where they are also categorized according to their concentration values.

The measurements revealed that 66% of the end-use points have a CH₄ concentration value between 1.5 and 5.0 ppmv, and 19% of them have values between 5.1 and 15.0 ppmv (Fig. 4). All NG end-use points in the residential area were almost within these two categories.

However, higher values are also detected during the measurement, of which 11% are between 15.1 and 50 ppmv. Moreover, values between 50 and 100 ppmv and 150.1 and 500.0 ppmv represent 1% and 3%, respectively. These highly detected concentrations were found at NG end-use points in the city center. From a statistic perspective, the minimum and maximum values were estimated 1.5 and 482.0 ppmv, respectively. Also, the mean value of all end-use points is 15.2 ppmv, with a standard-deviation of 58.1 ppmv (Table 1).

In addition to CH₄ concentration at the end-use points, the background was determined to be 2.2 ppmv after continuous street-level detection at primary streets over more than 3.5 km. By comparing the background value with the results obtained from the end-use points, the data revealed that 76% of the NG end-use points are higher than the background level, in which more than 84% and 77% of the concentrations in the city center area and the neighborhood are higher than the background.



Fig. 3 – End-use points categorized according to the estimated value of CH₄ concentration (in ppmv). a) NG end-use points in the city center; b) NG end-use points in the residential neighbourhood.

This study displays that NG network systems at end-use points are potential sources of CH₄ into the atmosphere in urban areas. Recent studies (von Fischer *et al.*, 2017; Zazzeri *et al.*, 2015) have also estimated high CH₄ emissions from the NG network in urban areas.

Also, Plant *et al.* (2019) studied **five urban centers** in the US East Coast from Washington to Boston, and they found that CH₄ emissions in these centers are underestimated in the national inventory estimated by the EPA inventory area. This can be clearly compared to our values from the city center, which have significant values due to the highly pressured operation in regard to NG

consumption on one hand. On the other hand, given the results of this study, the high percentage of leaks from almost 76% of the end-use points is an obvious indicator that the national inventory should consider these values within their annual reports.

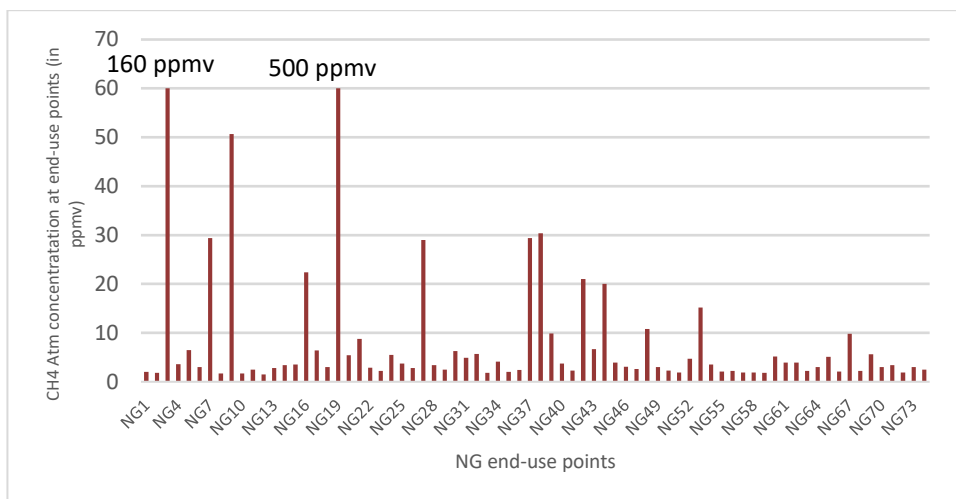


Fig. 4 – The values of CH₄ atmospheric concentration at the NG end-use points.

Table 1

Results from CH₄ concentration detection at NG end-use points

Counts	Leaks	Min CH ₄ (ppmv)	Max CH ₄ (ppmv)	Mean CH ₄ (ppmv)	Standard Deviation
74	76%	1.5	482.2	15	58.1

From another perspective, the results obtained by Keyes *et al.* (2020) showed that the min and max values of the survey in Hartford were 1.97 and 10.99 ppmv, respectively. Despite using Keyes *et al.* (2020) street-level detection of emissions, our study focused on carrying out gas concentration recording starting and ending close to the potential sources at each one of the end-use points for approximately one minute. Nevertheless, this simply and explicitly confirms that the NG network is a significant source of atmospheric methane in urban areas.

4. Conclusions

The study preliminary estimated that methane leaks in the urban area characterized by NG end-use points have values that exceeded background and reached even more than 400 ppmv, indicating gas leaks at more than 70% of the

points in the survey. Given the results of this study, recent urban surveys have also detected high values of CH₄ atmospheric concentration in urban areas, which are underestimated by national inventories.

Shortly, this study found that urban areas are an important source for methane atmospheric emissions, and NG end-use networks are a significant source, in particular confirming results obtained by recent urban studies.

For future surveys, it is highly recommended to conduct a large-scale survey on NG at both end-use points and to map all CH₄ release points to effectively introduce mitigation measures for the purpose of reducing CH₄ emission sources.

REFERENCES

- ANPM, N. E. P. A., Romania's Greenhouse Gas Inventory 1989-2018 (National Inventory report (NIR) No 28), https://unfccc.int/sites/default/files/resource/BR4_Romania.pdf (2020).
- Cambaliza M.O.L., Shepson P.B., Bogner J., Caulton D.R., Stirn B., Sweeney C., Montzka S.A., Gurney K.R., Spokas K., Salmon O.E., Lavoie T.N., Hendricks A., Mays K., Turnbull J., Miller B.R., Lauvaux T., Davis K., Karion A., Moser B., Richardson S., *Quantification and source apportionment of the methane emission flux from the city of Indianapolis*, *Elementa: Science of the Anthropocene*, **3**, 000037, <https://doi.org/10.12952/journal.elementa.000037>, (2015).
- Cuna S., Pendall E., Miller J.B., Tans P.P., Dlugokencky E., White J.W.C., *Separating contributions from natural and anthropogenic sources in atmospheric methane from the Black Sea region, Romania*, *Applied Geochemistry*, **23**(10), 2871-2879, <https://doi.org/10.1016/j.apgeochem.2008.04.019> (2008).
- EPA, Greenhouse Gases, United States Environmental Protection Agency, <https://www.epa.gov/report-environment/greenhouse-gases> (2022).
- Foster C.S., Crosman E.T., Holland L., Mallia D.V., Fasoli B., Bares R., Horel J., Lin J.C., *Confirmation of Elevated Methane Emissions in Utah's Uintah Basin with Ground - Based Observations and a High - Resolution Transport Model*, *Journal of Geophysical Research: Atmospheres*, **122**(23), <https://doi.org/10.1002/2017JD027480> (2017).
- Heimbürger A.M.F., Harvey R.M., Shepson P.B., Stirn B.H., Gore C., Turnbull J., Cambaliza M.O.L., Salmon O.E., Kerlo A.-E.M., Lavoie T.N., Davis K.J., Lauvaux T., Karion A., Sweeney C., Brewer W.A., Hardesty R.M., Gurney K.R., *Assessing the optimized precision of the aircraft mass balance method for measurement of urban greenhouse gas emission rates through averaging*, *Elementa: Science of the Anthropocene*, **5**, 26, <https://doi.org/10.1525/elementa.134> (2017).
- IPCC, Annex III: Glossary, In: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA (2013a).

- IPCC, *Observations: Atmosphere and Surface. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA (2013b).
- Keyes T., Ridge G., Klein M., Phillips N., Ackley R., Yang Y., *An enhanced procedure for urban mobile methane leak detection*, *Heliyon*, **6**(10), e04876, <https://doi.org/10.1016/j.heliyon.2020.e04876> (2020).
- McKain K., Down A., Raciti S.M., Budney J., Hutyra L.R., Floerchinger C., Herndon S.C., Nehrkorn T., Zahniser M.S., Jackson R.B., Phillips N., Wofsy S.C., *Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts*, *Proceedings of the National Academy of Sciences*, **112**(7), 1941-1946, <https://doi.org/10.1073/pnas.1416261112> (2015).
- Phillips N.G., Ackley R., Crosson E.R., Down A., Hutyra L.R., Brondfield M., Karr J.D., Zhao K., Jackson R.B., *Mapping urban pipeline leaks: Methane leaks across Boston*, *Environmental Pollution*, **173**, 1-4, <https://doi.org/10.1016/j.envpol.2012.11.003> (2013).
- Plant G., Kort E.A., Floerchinger C., Gvakharia A., Vimont I., Sweeney C., *Large Fugitive Methane Emissions from Urban Centers Along the U.S. East Coast*, *Geophysical Research Letters*, **46**(14), 8500-8507, <https://doi.org/10.1029/2019GL082635> (2019).
- UNEP, *Emissions Gap Report 2011: The Heat Is On—A world of climate promises not yet delivered*, Nairobi (2021).
- von Fischer J.C., Cooley D., Chamberlain S., Gaylord A., Griebenow C.J., Hamburg S.P., Salo J., Schumacher R., Theobald D., Ham J., *Rapid, Vehicle-Based Identification of Location and Magnitude of Urban Natural Gas Pipeline Leaks*, *Environmental Science & Technology*, **51**(7), 4091-4099, <https://doi.org/10.1021/acs.est.6b06095> (2017).
- Wong G.X., Hirata R., Hirano T., Kiew F., Aeries E.B., Musin K.K., Waili J.W., Lo K.S., Melling L., *How do land use practices affect methane emissions from tropical peat ecosystems?* *Agricultural and Forest Meteorology*, 282-283, 107869, <https://doi.org/10.1016/j.agrformet.2019.107869> (2020).
- Wunch D., Toon G.C., Hedelius J.K., Vizenor N., Roehl C.M., Saad K.M., Blavier J.-F.L., Blake D.R., Wennberg P.O., *Quantifying the loss of processed natural gas within California's South Coast Air Basin using long-term measurements of ethane and methane*, *Atmospheric Chemistry and Physics*, **16**(22), 14091-14105, <https://doi.org/10.5194/acp-16-14091-2016> (2016).
- Zazzeri G., Lowry D., Fisher R.E., France J.L., Lanoisellé M., Nisbet E.G., *Plume mapping and isotopic characterisation of anthropogenic methane sources*, *Atmospheric Environment*, **110**, 151-162, <https://doi.org/10.1016/j.atmosenv.2015.03.029> (2015).

DETECȚIA CONCENTRAȚIILOR MARI DE CH₄ LA
REȚEAUA DE DISTRIBUȚIE PENTRU UTILIZARE FINALĂ DE GAZ
NATURAL ÎN ZONE URBANE

(Rezumat)

De la revoluția industrială, emisiile de CH₄ au crescut cu 150%. Zonele urbane sunt responsabile pentru aproximativ 60% din aceste emisii, provenind în principal din activitățile antropice. În plus, urbanizarea a provocat schimbări în utilizarea terenului și a redus rezervele de CH₄. Sursele de emisii de CH₄ din zonele urbane au încă un grad ridicat de incertitudine. Studiile recente au afirmat că scurgerile din rețelele de distribuție a gazelor naturale sunt surse semnificative de CH₄ în atmosferă și reprezintă, de asemenea, o potențială pierdere de resurse energetice. Cu toate acestea, emisiile de la rețelele de gaze naturale destinate utilizării finale sunt puțin explorate în literatură. Principalii contributivi în bugetul CH₄ al României sunt sectoarele agricultură și energie. În perioada 1989–2000, rata emisiilor de metan a scăzut cu 34% din cauza schimbărilor sectoriale din agricultură și combustibilii fosili. Cu toate acestea, inventarul național al României nu raportează emisiile de CH₄ din zonele urbane.

Acest studiu investighează concentrația de CH₄ din rețeaua de gaze naturale destinate utilizării finale din Cluj-Napoca, al doilea oraș ca mărime din România ca populație. Aceste puncte pot fi identificate ca parte a rețelelor de distribuție a gazelor naturale care deservesc gaze naturale utilizatorilor finali pentru consumul de gaze naturale. Punctele detectate au fost joncțiunile conductelor și contoarele de gaze naturale. Estimarea concentrației de CH₄ în atmosferă a fost realizată pe baza unui senzor laser CH₄ Tunable Diode Laser Absorption Spectroscopy (TDLAS) cu o precizie ridicată de măsurare de 0,1 ppmv. Această detectare a concentrației de CH₄ a fost efectuată din decembrie 2022 până în ianuarie 2023 la 74 de puncte de utilizare finală a gazelor naturale. Determinarea fie a reprezentat scurgeri, fie a nu a fost după estimarea nivelului de fond în oraș și compararea concentrațiilor obținute cu acest fond. Acest studiu a relevat că 76% dintre punctele de distribuție a gazelor naturale cu utilizare finală detectate au scurgeri de gaze și reprezintă contributivi continui la bugetul anual CH₄. Cu toate acestea, această contribuție în bugetul anual ar trebui estimată. În plus, rezultatele acestui studiu indică prezența unor scurgeri mari de la punctele de utilizare finală a gazelor naturale din zonele urbane. Aceștia sugerează investigarea aprofundată și alocarea tuturor scurgerilor de gaze naturale la punctele de utilizare finală, pentru a lua anumite măsuri de reducere în ceea ce privește atenuarea CH₄.