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Street-level emissions of methane and nitrous oxide from the wastewater collection system in Cincinnati, Ohio[☆]



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ABSTRACT

Recent studies have indicated that urban streets can be hotspots for emissions of methane (CH₄) from leaky natural gas lines, particularly in cities with older natural gas distribution systems. The objective of the current study was to determine whether leaking sewer pipes could also be a source of street-level CH₄ as well as nitrous oxide (N₂O) in Cincinnati, Ohio, a city with a relatively new gas pipeline network. To do this, we measured the carbon ($\delta^{13}\text{C}$) and hydrogen ($\delta^2\text{H}$) stable isotopic composition of CH₄ to distinguish between biogenic CH₄ from sewer gas and thermogenic CH₄ from leaking natural gas pipelines and measured CH₄ and N₂O flux rates and concentrations at sites from a previous study of street-level CH₄ enhancements (77 out of 104 sites) as well as additional sites found through surveying sewer grates and utility manholes (27 out of 104 sites). The average isotopic signatures for $\delta^{13}\text{C}\text{-CH}_4$ and $\delta^2\text{H}\text{-CH}_4$ were $-48.5\text{‰} \pm 6.0\text{‰}$ and $-302\text{‰} \pm 142\text{‰}$. The measured flux rates ranged from 0.0 to 282.5 mg CH₄ day⁻¹ and 0.0–14.1 mg N₂O day⁻¹ (n = 43). The average CH₄ and N₂O concentrations measured in our study were 4.0 ± 7.6 ppm and 392 ± 158 ppb, respectively (n = 104). 72% of sites where fluxes were measured were a source of biogenic CH₄. Overall, 47% of the sampled sites had biogenic CH₄, while only 13% of our sites had solely thermogenic CH₄. The other sites were either a source of both biogenic and thermogenic CH₄ (13%), and a relatively large portion of sites had an unresolved source (29%). Overall, this survey of emissions across a large urban area indicates that production and emission of biogenic CH₄ and N₂O is considerable, although CH₄ fluxes are lower than those reported for cities with leaky natural gas distribution systems.

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1. Introduction

Atmospheric concentrations of the greenhouse gases methane (CH₄) and nitrous oxide (N₂O) are increasing as a result of human activities (Ciais et al., 2013; NOAA, 2016). Methane has a global warming potential 34 times greater than carbon dioxide (CO₂) over 100 years, and 86 times greater than CO₂ over 20 years (Myhre et al., 2013). N₂O has a global warming potential 298 times greater than CO₂ over 100 years (Myhre et al., 2013); N₂O is also the dominant ozone-depleting gas emitted by human activities (Ravishankara et al., 2009). Both CH₄ and N₂O have unique urban point and nonpoint sources. Examples of urban point sources of

N₂O include wastewater treatment plants, fertilized landscapes, and industrial processes; whereas fertilized landscaping soils and, to a lesser extent, vehicle emissions, are diffuse or nonpoint sources of urban N₂O (Townsend-Small et al., 2011a, 2011b; USEPA, 2015a; 2015b). Urban areas have discrete sources of CH₄ from landfills, dairies, wastewater treatment plants, and power plants, but natural gas pipelines and end uses such as homes, power plants, and customer meters are more diffuse CH₄ sources in cities (Lamb et al., 2015, 2016; Townsend-Small et al., 2012, 2016b).

Overall, the oil and natural gas supply chain is the largest anthropogenic CH₄ source nationally, accounting for approximately 30% of U.S. CH₄ emissions (USEPA, 2015a), and is likely the largest anthropogenic source globally (Saunio et al., 2016). The lack of quantitative data on this source in urban areas has motivated research on street-level CH₄ emissions from the underground natural gas distribution pipelines in roadways (Phillips et al., 2013; Jackson et al., 2014; Gallagher et al., 2015; Hendrick et al., 2016; von Fischer et al., 2017). For example, Phillips et al. (2013) and Jackson

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et al. (2014) mapped street-level CH₄ enhancements (≥ 2.50 ppm) over urban roadways in Boston, MA and Washington, D.C., respectively. Both studies suggested that a majority of street-level CH₄ enhancements were from a thermogenic source, such as natural gas distribution pipelines. Using similar methods, Gallagher et al. (2015) surveyed three East Coast cities (Manhattan, NY, Cincinnati, OH, and Durham, NC) and found that cities with accelerated natural gas pipeline replacement programs, such as Durham and Cincinnati, had fewer street leaks per kilometer of roadway (0.14 leaks km⁻¹ and 0.29 leaks km⁻¹, respectively) than metropolitan areas with slower pipeline replacement programs such as Manhattan, Boston, and Washington, D.C., with 2.64 leaks km⁻¹ (Gallagher et al., 2015), 2.66 leaks km⁻¹ (Phillips et al., 2013), and 2.44 leaks km⁻¹ (Jackson et al., 2014), respectively. Similarly, von Fischer et al. (2017) showed that cities with older, more corrosion-prone distribution gas lines had higher leak rates of natural gas than those with more rapid pipeline replacement programs.

Although oil and gas systems are a large contributor to CH₄ emissions at a variety of spatial scales, all together, biological CH₄ production is the largest contributor to both natural (e.g., wetlands, lakes, and soils) and anthropogenic (e.g., agriculture and waste) emissions of CH₄ globally (USEPA, 2015a; Saunois et al., 2016). On city streets, CH₄ enhancements could evolve from sewer and natural gas pipelines, both of which can leak from cracks, corrosion, or joint leakage as well as through vents, grates, and infrastructure access points such as manholes. Previous studies have indicated that sewer mains could be a source of atmospheric CH₄ (Guisasola et al., 2008; Liu et al., 2015; Chamberlain et al., 2016; Hopkins et al., 2016a) and/or N₂O (Short et al., 2014). The nutrient-rich wastewater carried in sewer pipelines encounters environmental conditions that produce both biogenic CH₄ and N₂O (Doorn et al., 2006; Townsend-Small et al., 2011a; USEPA, 2015a). The close placement of underground sewer and natural gas conveyances can result in the mixing of biogenic CH₄, N₂O, and natural gas, all of which are lighter than air and travel upwards along pressure gradients to vent as combined emissions into the atmosphere, either through infrastructure access points or via soil diffusion. Therefore, biogenic CH₄ produced in the wastewater collection system could be contributing to overall street-level CH₄ emissions, such that not all CH₄ enhancements are street leaks from the natural gas distribution pipelines. Biogenic CH₄ and thermogenic CH₄ can be distinguished by the carbon ($\delta^{13}\text{C-CH}_4$) and hydrogen ($\delta^2\text{H-CH}_4$) stable isotopic composition of CH₄ (Townsend-Small et al., 2012, 2015), or by analysis of other source apportionment tracers that are co-emitted with thermogenic CH₄, such as ethane or larger alkanes (Simpson et al., 2012; Hopkins et al., 2016b), or with biogenic CH₄, such as N₂O or ammonia (Leytem et al., 2011; Schneider et al., 2015).

The objective of this study was to determine the contribution of biogenic sources to street-level CH₄ and N₂O enhancements and emissions in Cincinnati, OH. We measured $\delta^{13}\text{C-CH}_4$ and $\delta^2\text{H-CH}_4$ and made measurements of CH₄ and N₂O emission rates and concentrations from utility manholes, sewer grates, and CH₄ hotspots identified in a previous study (Gallagher et al., 2015) as well as a variety of other locations on city streets.

2. Material and methods

2.1. Study area

All sampling took place between May and September 2016 within the city limits of Cincinnati, OH, a metropolitan area situated on the north bank of the Ohio River in southwestern Ohio. The city is located within Hamilton County, the primary county of operation for the Metropolitan Sewer District of Greater Cincinnati (MSD) to

collect and treat wastewater. The MSD operates seven wastewater treatment plants (WWTP), and maintains approximately 5000 km of sewer pipeline (MSD, 2017). Our study area (Fig. 1) covers 54% of the entire MSD wastewater collection system (Pittinger and Chen, 2017), and sites selected for sampling were located in the Mill Creek and Little Miami WWTP service areas. Cincinnati has combined sewer infrastructure, where storm sewers are combined with sanitary sewers during high runoff events. However, all sampling in the current study was conducted during dry conditions to avoid sampling combined sewers.

2.2. Site selection

Most of our sampling sites (77 out of 104 total; Table S1) were previously identified as CH₄ enhancements during a street-level survey of CH₄ concentration in Cincinnati (Gallagher et al., 2015); this represents a randomly selected subset (33%) of the 233 CH₄ enhancements originally identified by Gallagher et al. (2015). The remainder of our sites were either located near sites identified in the Gallagher et al. (2015) study; qualitatively identified as emitters of CH₄ by the characteristic odor of mercaptan, the odorizer used in natural gas or the smell of septic sewage; or selected due to the known location of a combined sewage outfall or pipeline. Because 27 out of our 104 sites were selected this way, our emission rate measurements may be skewed high and may not be representative of the true range of emissions across the city. Most samples were collected from sewer grates or manholes (both utility access manholes and sewer manholes), with a small number of samples collected directly from streets or lawns where CH₄ enhancements were identified (Table S1).

2.3. Sample collection

Direct measurements were made from city streets within the study area (Fig. 1). Each site was qualitatively screened with a Gas-Rover™ (Bascom-Turner Instruments, Inc., Norwood, MA; detection limit = 10 ppm CH₄) to indicate the presence or absence of elevated CH₄ levels. We then collected gas samples for CH₄ and N₂O concentration levels and analysis of $\delta^{13}\text{C-CH}_4$ and $\delta^2\text{H-CH}_4$, although not all samples were analyzed for isotopic composition (see further discussion below). We also collected samples of natural gas and sewer gas for analysis of $\delta^{13}\text{C-CH}_4$ and $\delta^2\text{H-CH}_4$ endmembers. Natural gas samples were taken from one residential stove in Cincinnati and from the laboratory in the Department of Geology at University of Cincinnati. We also took samples from throughout a wastewater treatment plant in Cincinnati with preliminary, primary, secondary, and sludge digestion processes.

Air samples were collected by inserting a 100 mL syringe with stopcock that was fitted with a 66-cm long piece of plastic tubing into the manhole or sewer grate. The plastic tubing reached approximately 60 cm into the grate opening to ensure that sewer gas was being sampled. The syringe was filled and cleared three times before filling vials to ensure that sample gas, not ambient air, was sampled, and that the syringe and tubing were cleared between sampling events. This sample was then transferred using a hypodermic needle into a 20 mL pre-evacuated glass vial with gray butyl rubber septa and aluminum crimps for later analysis. Gas samples were taken as described above and transferred into 12 mL pre-evacuated glass vials (Exetainers®, Labco Ltd., Buckinghamshire, UK) for stable isotope analysis. At sites located at ground level (i.e., not in a sewer grate or manhole; Table S1), samples were taken from the spot previously identified in Gallagher et al. (2015) without inserting the tubing underground.

We also measured CH₄ and N₂O emission rates at a subset of sites (n = 43) where access was safe and which were located in

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