Response to Zhao et al., ACPD, 2026

RC1

In “Inefficient Consumption of Natural Gas Drives Methane Emissions from a Megacity,” Zhao et al. utilize multiple measurement platforms to investigate the sources of methane emissions in the New York City Metropolitan Area. The observations presented in this work represent a dense dataset from which much can be learned about the largest metropolitan area in the United States. Understanding the sources of methane emissions in cities is key to successful mitigation strategies. Accordingly, I find the study timely and of interest to a broad audience.

The study is very thoroughly presented with clear figures and method descriptions. However, I believe there are some areas that should be addressed prior to publication.

1. Section 2.6. This section lacks citations for the transport models and their application in this work. Another option could be to reference the later section, 2.6.3.

   We thank the reviewer for this comment. The citations were added in the main text:

   “We calculated hourly simulated methane enhancements (ΔCH_4, ppb) by convolving a series of global, national, and regional methane emission inventories (units: nmol m−2 s−1) with surface influence footprints (ppb (nmol m−2 s−1)−1) derived from a Lagrangian atmospheric transport model described in Section 2.6.3 (Fasoli et al., 2018; Lin et al., 2003).”

2. Line 302: The term “suburban outflow” is first introduced here, but it wasn’t clear to me what this means. It appears to reference the difference between this work and that of Schiferl et al. 2025, but it wasn’t clear how they are different. I assume it has to do with the footprint of the Mineola Tower versus that of what is used in Schiferl at al.

We thank the reviewer for this valuable comment. To clarify our use of the term "suburban outflow": Schiferl et al. (2025) employ a rooftop observation site located within Manhattan's urban core (Upper Harlem), whereas our Mineola Tower site is located in a suburban setting on Long Island. Under westerly wind conditions, Mineola Tower is capable of detecting urban emission signals from New York City (see Figure 1 in the main text for the site footprint). Notably, the footprint of our study overlaps with that of Schiferl et al. (2025) (See the Figure 1 in the main text and Figure 4a in Schiferl et al., 2025), providing a basis for comparison while also highlighting the contrasting perspectives offered by a near-source urban core site versus a downwind suburban receptor site. In the main text, we revised the text to be clearer in line 310:

“The seasonal cycle of NYCMA methane emissions quantified ~~for the suburban outflow~~ using suburban Mineola Tower measurements is consistent with the ~~city core methane~~ emissions from Schiferl et al. (2025) using city core observations, which saw peak methane emissions in January and February, followed by a decline into spring {Schiferl et al.,2025}.

The 75% percentile footprint at Mineola Tower encompasses the majority of the urban areas within the NYCMA (Figure 1), including northeastern New Jersey, Manhattan, the Bronx, and western Long Island. This footprint largely overlaps with the 75% percentile footprint reported by Schiferl et al. (2025) for their urban core site, providing a robust spatial basis for comparison between the two studies {Schiferl et al.,2025}."

3. There is clearly significant added value from concurrent measurements of CH4, CO, and C2H6. However, is it fair to attribute all the variability in observed CH4:CO and CH4:C2H6 to source behavior? If I understand correctly, "plumes" in this work correspond to relatively short time periods (as suggested by Figure 5b) and each undergo the slope analysis. If that is the case, is it possible they have experienced different levels of atmospheric mixing, such that, depending on the sources, that could influence the derived CH4:CO and C2H6:CH4 slopes? So, is it fair to completely attribute all the observed variability to combustion efficiency in the statement below:

Line 332: "Thermogenic plumes with moderate R2 ΔCO:ΔCH4 (between 0.1 and 0.8) are observed more frequently during months with building heating loads (Figure S12), suggesting greater variability in combustion efficiency, likely driven by various natural gas combustion processes for heating purposes."

We thank the reviewer for this valuable comment and apologize for the lack of clarity in the original manuscript. To clarify our methodology: we use the linear correlation (R^2) of methane and ethane enhancements, rather than the slopes, to infer different sources. We also use the correlation between trace gas enhancements calculated at 1 Hz after removing the background (defined as the 5th percentile of the 1 Hz dry mole fractions over a centered 48-hour running window) to reduce the influence of regional background changes, which we assume vary on timescales longer than 5 minutes. Furthermore, when discussing the CO-to-ethane relationship, where we also only discuss the correlation rather than slope, we only use thermogenic plumes identified by RMA regression with $R^2_{\Delta C2H6:\Delta CH4}$≥0.95, which indicates air masses with limited atmospheric mixing. This two-step approach minimizes the influence of background variability and dilution when quantifying thermogenic air masses.

We clarified the method in lines 151 to 155:

"*To identify thermogenic methane plumes, we calculated the Reduced Major Axis (RMA) ~~fit~~ correlation between 1 Hz ethane enhancements (ΔC2H6) and methane enhancements (ΔCH4) for 5-minute tower measurement intervals ($R^2_{\Delta C2H6:\Delta CH4}$), which considers variability in both x and y axes. To identify times of incomplete combustion, we calculated the ~~fit~~ RMA correlation between 1Hz CO enhancements (ΔCO) and ΔCH4 ($R^2_{\Delta CO:\Delta CH4}$).*"

Line 336: “In contrast, only 7% (summer) and 6% (winter) of the identified thermogenic plumes were observed to have no combustion signature (R2 ΔCO:ΔCH4 < 0.1), showing that natural gas emissions from pipeline leaks were less frequently observed at the Mineola tower compared to combustion-related natural gas sources.” What was the reasoning for using 0.1 as the cutoff here? For lower correlations between CO and CH4, one might expect noise/uncertainty to have a larger effect on the calculated R2 value. Would the conclusions change if you used a larger threshold for these conclusions?

We thank the reviewer for the insightful comment regarding the $R^2_{\Delta CO:\Delta CH4}$ threshold, which was also mentioned by Reviewer 2. The $R^2_{\Delta CO:\Delta CH4}$ threshold was selected to be consistent with our thermogenic plume identification criterion of $R^2_{\Delta C2H6:\Delta CH4}> 0.95$. To evaluate the robustness of this threshold, we examined the distribution of $R^2_{\Delta CO:\Delta CH4}$ for all 5-minute thermogenic plumes. As shown in the attached histogram and in Figure S12, the distribution peaks near $R^2_{\Delta CO:\Delta CH4}=1$, indicating that many thermogenic plumes exhibit strong $R^2_{CO:CH4}$ correlations and therefore likely include a combustion influence. A smaller peak is observed near $R^2=0$ in summer; however, when the cutoff for identifying plumes without a combustion signature is increased above 0.1, the overall conclusion does not change: most thermogenic plumes remain associated with a combustion-related ΔCO:ΔCH4 relationship, while a smaller fraction shows no clear combustion signal.

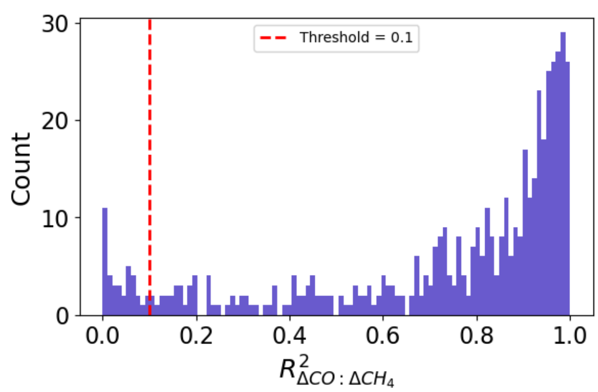

Figure 1. Histogram of $R^2_{CO:CH4}$ between 1 Hz ΔCO:ΔCH4 within each 5 minute interval for summer 2023.

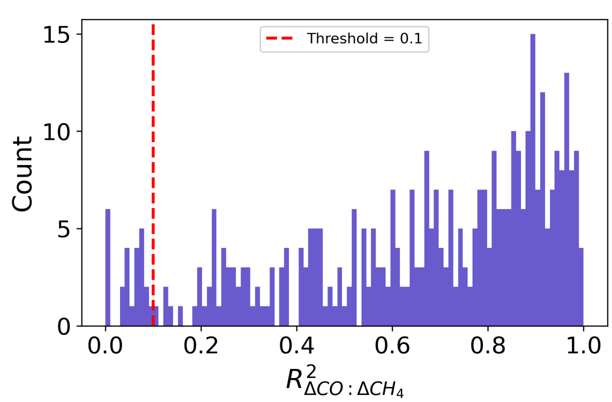

Figure 2. Histogram of $R^2_{CO:CH4}$ between 1 Hz ΔCO:ΔCH4 within each 5-minute interval for winter 2024.

In supplemental section 3, the authors examine street-level data of two pipeline leaks, with high correlation between CH4 and C2H6, but not CO. Were there other instances of strong correlations between CH4, C2H6, and CO in the street-level data? This would indicate incomplete combustion of natural gas and support the attribution conclusions since the transport time would be relatively short. In the current manuscript, it wasn't clear to me how the street-level observations were incorporated into the larger study.

We thank the reviewer for this comment. The mobile measurements were conducted to serve three specific purposes:

1. **Quantifying the local pipeline ethane-to-methane ratio.** The mobile drive allowed us to directly measure the ethane:methane ratio of local natural gas pipeline leaks, providing a local tracer signature for comparison with tower observations.
2. **Assessing the presence of natural gas combustion signals at street level.** Exhaust from natural gas combustion is typically emitted from elevated heights, such as building chimneys, rather than at street level. Mobile sampling was conducted during daytime conditions when the boundary layer is relatively well-mixed, meaning hot combustion plumes are unlikely to descend and be sampled at street level. Consistent with this expectation, we measured natural gas leaks but no natural gas related combustion signals were detected in our street-level mobile dataset.
3. **Evaluating the method that distinguishes between combustion and natural gas leak sources using the correlation between trace gases.** Street-level sampling is more sensitive to near-surface sources, including natural gas pipeline leaks and CO emissions from traffic. Traffic-related CO is not co-emitted with methane, but different sources can be mixed during mobile sampling. The presence of nearby CO sources and methane enhancements from different sources does not usually produce strong correlations at high temporal resolution, indicating that even in environments where nearby CO sources and methane enhancements from different sources are common, **we do not observe strong methane and CO correlations at 1 Hz level observations.** This further supports our method of using 1 Hz tracer–tracer correlations to inform potential incomplete combustion of natural gas.

We clarified and revised our manuscript about mobile sampling in the main text Line 105:

"*To complement our tower-based observations, we conducted street-level sampling of 1 Hz methane, ethane, CO, and N2O dry mole fractions around the Mineola tower using the New York Atmospheric Composition and Air Quality (NYAAQ) Mobile Laboratory on August 16th, 2023. More details are described in Section S3 and Figure S2. Trace gas correlations were used to characterize methane sources in the area around the tower. We used mobile sampling to assess if natural gas combustion signals could be detected at* *street level and to evaluate whether the 1 Hz tracer–tracer correlation method (Section 2.5) can distinguish natural gas signals from nearby sources, such as traffic-related CO (detailed information can be found in supporting information Section S3).*"

And in the Supporting Information Line 55:

"*During the mobile sampling drive (Figure \ref{Fig:S2}), we observed two pipeline leaks with strong correlations between methane and ethane, with no correlations between methane and CO despite nearby sources such as traffic. We did not see consistent street-level co-peaks of CO and natural gas (methane and ethane). Since natural gas combustion exhaust is typically emitted at heights far above street level (e.g., from building chimneys), daytime street-level mobile sampling rarely interceptes these elevated plumes, and we therefore did not observe methane plumes strongly correlated with CO. Instead, our measurements primarily captured near-surface sources, and the absence of strong 1 Hz* $R^2_{\Delta CO:\Delta CH4}$*, despite CO being a near-surface traffic tracer, supports the use of high-frequency tracer–tracer correlations to distinguish natural gas leaks from incomplete natural gas combustion.*"