White Paper:
Atmospheric Methane, Seasonal Variations, and Space Heating

Executive Summary

A published paper[1] *Atmospheric methane emissions correlate with natural gas consumption from residential and commercial sectors in Los Angeles* (JPL) asserts that winter increases in methane emissions are due to emissions stemming from natural gas consumption during that period of the year in the Los Angeles (LA) metropolitan area. In particular, the JPL paper infers a large impact from natural gas space heating (the largest incremental winter seasonal use of natural gas).

A recently published paper by Merrin and Francisco (M&F) indicates this assertion (i.e., post-combustion methane emissions) in the JPL paper is unlikely to be a primary factor.[2] Our analysis indicates natural gas space heating has up to 0.14 Gg/month of methane emissions in the LA Basin. The JPL paper infers an additional seasonal winter methane emission estimate of 20 Gg/month. This is over 140 times greater than our estimates of post-combustion methane emissions from natural gas space heating equipment. **It is improbable that operation of natural gas space heating is a primary or even secondary contributor to this level of methane emissions.**

The JPL paper also does not include an important mechanism that is a key contributor to seasonal changes in atmospheric methane concentrations – a central element of the investigation. The process of methane oxidation is known to be driven by hydroxyl radical (OH) – which is also a key actor in atmospheric ozone chemistry. **This leads to a process of a summer drop in atmospheric methane concentration on a global basis and is seen independent of methane emissions sources; even locations far from natural gas production and consumption show a summer decline and winter increase in atmospheric methane concentration.** The JPL paper does not include a complete exploration of the possible causation of the seasonal cycles in atmospheric methane concentration; this phenomenon may be enhanced in the LA Basin due to the presence of pollutants such as NO_x and ozone. Further investigation is warranted on this topic.

There is widespread agreement on the need to reduce the level of atmospheric methane and the release of methane from various sources, including natural gas production, delivery, and use. There is also a need to conduct research to improve our understanding of methane emissions and sinks – globally, nationally, and – like the JPL study – regionally. However, there are issues with the JPL study – most notably the likely erroneous conclusion that natural gas space heating is a key contributor to atmospheric methane emissions.

Further research is warranted to improve our understanding of year-round methane emissions and sinks – enabling a better understanding of key factors that can contribute to mitigating methane emissions.

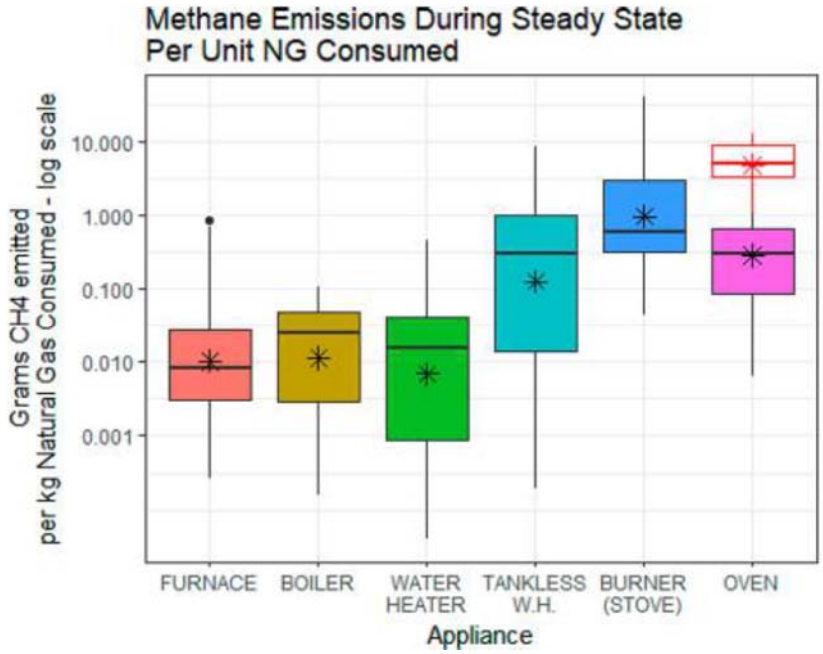
Discussion on Methane Emissions from Natural Gas Space Heating

The JPL paper attempts to make a top-down case for a better understanding of the contribution of methane emissions (bottom-up) sources. In particular, they make an inference that seasonal winter uses of natural gas – which is principally natural gas used for space heating – could be a contributor to seasonal changes in methane.

[1] https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2019GL083400

[2] Merrin, Z. and Francisco, P.W., "Unburned Methane Emissions from Residential Natural Gas Appliances," Env. Sci & Tech., March 25, 2019.

The JPL paper asserts there is a winter-based seasonal component of methane emissions on the order of 20 Gg/month in the LA Basin. They imply this is mainly from winter season natural gas use – that is, space heating. To test this assertion, we use the results of a recently published paper on methane emissions from natural gas equipment by Merrin and Francisco (M&F). For furnaces, M&F find the steady-state emission rate is about 0.008 g of methane/kg of natural gas consumed – **this was the lowest emission rate of the different types of natural gas equipment tested** (see below).



From this testing, most methane emissions stem from equipment start-up and shut down. In fact, at steady-state operation M&F find instances where furnace methane emission levels are lower than background ambient methane concentration (i.e., furnaces in some instances consume ambient methane during steady-state operation). Even factoring in on/off cycles, this study finds very low methane emissions from gas furnaces.

Based on estimates from the JPL paper and other GTI analysis, there is an incremental seasonal natural gas use of about 500 Gg/month in the LA Basin during the winter – mainly for space heating. Using the natural gas furnace methane emission data from the M&F paper (and including different on/off cycle time assumptions), we find a range for furnace methane emissions values of 0.11 - 0.28 g/kg natural gas consumed (the upper value uses an unlikely 5-minute cycle time). Using the upper end value and applying that to 500 Gg/month results in a space heating-related monthly emission value of 0.14 Gg/month. The JPL paper asserts an incremental winter-based methane emissions from natural gas of 20 Gg/month; this emission rate is over 140 times greater than the composite furnace emission rate. **It is improbable that seasonal natural gas combustion for winter space heating is a primary (or secondary) contributor to seasonal changes in atmospheric methane concentration.**

Atmospheric Methane Oxidation and Seasonal Changes in Methane

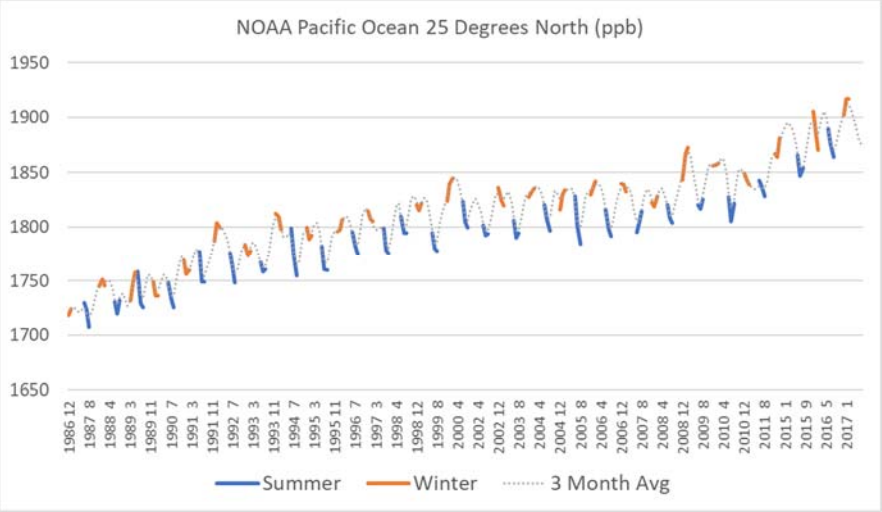
The JPL paper does not discuss the possible contribution of an important mechanism that contributes to seasonal changes in atmospheric methane concentrations. Other similar publications (Townsend-Small)

discussing methane emissions in LA have touched on this important factor.[3] The following are two quotes from the Townsend-Small paper:

> "Methane is also an important component in the overall oxidative capacity of the troposphere"
>
> "This indicates consumption of CH_4 by interaction with OH and other radicals in the urban atmosphere at our study site, which is high in concentration in the summer."

The process of methane oxidation is driven by the hydroxyl radical (OH) – also a key actor in atmospheric ozone chemistry. The role of OH in atmospheric chemistry is complex[4], but there appears to be a link to higher rates of OH formation stemming from more solar ultraviolet (UV) radiation – which is greater during the summer. **This process of a summer drop in atmospheric methane concentration occurs on a global basis and is seen independent of methane emissions sources; even locations far from natural gas production and consumption show a summer decline and winter increase in atmospheric methane concentration.** The JPL paper does not include a complete exploration of the possible causation of the seasonal cycles in atmospheric methane concentration – a phenomenon that may be enhanced within the LA Basin due to the presence of pollutants such as NO_x and ozone.

NOAA has a large database of worldwide atmospheric methane concentration data. Below are NOAA data from the Pacific Ocean at 25° north. This shows summer and winter seasons over an extended number of years.



These multi-year data – located far from natural gas production or consumption sources – show clear seasonal changes in atmospheric methane concentration. Concentration rates are consistently lower and steadily decline in the summer and rebound or rise in the winter in all instances.

The following figure shows multi-year seasonal changes in monthly average methane concentrations at the Mt. Wilson Observatory in California – a primary location discussed in the JPL paper. A cyclical pattern is seen with maximum values in the winter and minimum values in the summer. There is a lack of information to fully describe the behavior of methane and OH at lower elevations in the LA Basin – that is, locations below the planetary boundary layer discussed in the JPL paper. This leads to uncertainty about the dynamic behavior of methane emissions and sinks in the LA Basin throughout the year.

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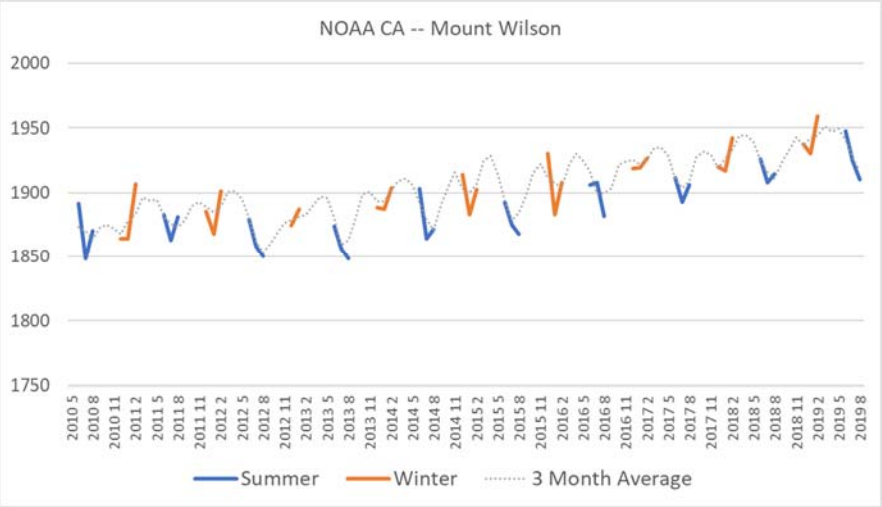
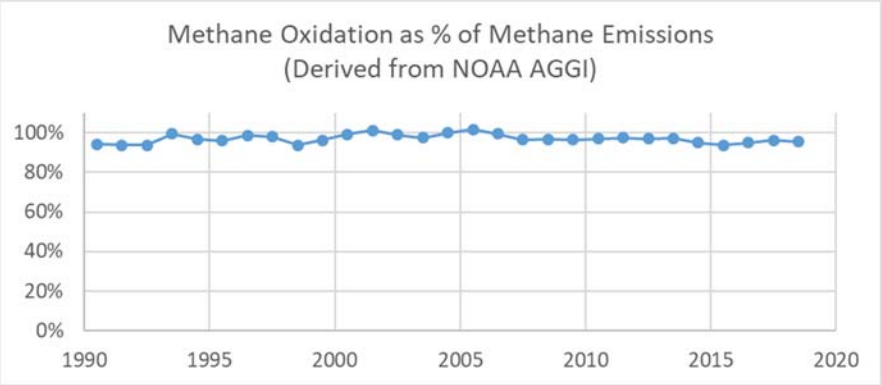
[3] https://www.researchgate.net/publication/231175207_Isotopic_measurements_of_atmospheric_methane_in_Los_Angeles_California_USA_Influence_of_fugitive_fossil_fuel_emissions

[4] https://pubs.acs.org/doi/10.1021/cr500310b#



Methane oxidation is a significant parameter. On an annual basis, the annual amount of methane oxidation is remarkably similar to the amount of methane emissions. Since 1990, methane oxidation has averaged about 97% of methane emissions (GTI calculations from NOAA Annual Greenhouse Gas Index data; assumes a 10-year atmospheric life for methane).



Methane oxidation is the largest single parameter that affects well-mixed atmospheric methane concentrations and highly influential in seasonal changes in methane concentration. The phrase “methane oxidation” never appears in the JPL paper and its influence on monthly or seasonal changes in atmospheric methane concentration is not explicitly discussed (in contrast to the Townsend-Small LA methane emission paper that discusses this topic). The atmospheric conditions over LA in the summer may create conditions that cause a larger fluctuation in atmospheric methane concentrations between summer and winter than what would be encountered naturally. The JPL paper does not explicitly discuss how this phenomenon factors into their analysis.

Methane Sources, Sinks, and Measurement Challenges

There are several salient points to consider with respect to quantitatively assessing methane emission sources, sinks, and measurement: (1) methane emissions are generally small in the context of the total inventory of methane in the atmosphere, (2) **net methane emissions are substantially smaller** (when factoring in continuing reductions from methane oxidation), and (3) large-scale mixing of methane is an ongoing process in the atmosphere, driven by ever-changing winds and gaseous molecular diffusion.

The JPL paper includes atmospheric measurements as well as analyses based on estimates from emission inventory data (which have their own uncertainty). The translation from primary measurements (e.g., ppb of

methane) to emissions has further uncertainties. These uncertainties are further compounded by the lack of explicit discussion regarding variable seasonal methane oxidation in the LA Basin (which is unique to other regions of the US) and the challenges with accounting for mixing and diffusion effects.

The JPL paper does not include primary measurements of methane ppb concentrations in the LA Basin (other than the Mt. Wilson background data). Instead, data are shown as a ratio of excess methane to carbon dioxide (CO_2). This analytical approach of coupling methane and CO_2 is curious in that it adds an additional factor (CO_2) that has its own seasonal variability and ties to other fuel sources and factors (e.g., vehicles, plant photosynthetic activity).[5] Further, the vast majority of methane emissions are not from post-combustion emissions (tied to CO_2 emissions), but from pre-combustion emissions from landfills, wastewater treatment facilities, natural gas delivery systems, agricultural operations such as dairy farms, etc. In our estimation, the paper would be better served by focusing specifically on methane measurements as opposed to a convoluted metric (i.e., ratio of excess methane to excess CO_2).

Conclusions

The assertion in the JPL paper that seasonal winter natural gas use (which is predominantly for space heating) is a principal contributor to seasonal changes in atmospheric methane concentration is improbable, based on equipment-level measurements. Post-combustion methane emissions from natural gas furnaces and other similar equipment are much smaller than what JPL infers.

The JPL paper appears incomplete in terms of addressing all possible causes of seasonal changes in atmospheric methane concentrations. An alternative cause for the enhanced seasonal methane changes in the atmosphere may be related to the higher methane oxidation rates that occur during the summer when the combined effects of longer daylight hours (i.e., greater UV radiation) and higher OH formation rates. The unique atmospheric chemistry in the LA Basin – as evidenced by very high ozone concentrations – is an indicator this may be a key parameter for consideration.

It is important to continue to make impactful strides in reducing the methane emission sources – including natural gas production, delivery, and use – and to lower atmospheric methane concentrations. However, the basis and conclusions of the JPL report are not comprehensive and appear to lead to erroneous conclusions. They do not incorporate a potentially large chemical mechanism influencing an enhanced summer time decrease and winter rebound in methane levels – a process which can be seen to occur in remote regions far from natural gas production and consumption. Independent test data from natural gas space heating equipment provides no evidence for methane emission levels approaching those cited in the JPL paper.

Further studies are needed to more fully understand seasonal atmospheric methane concentration changes and refine estimates of methane emission sources and quantities. This is a challenging field of scientific study. Further refinements in measurement technology and methodology are needed to fully account for methane emissions, sinks, and the effects of atmospheric mixing and diffusion.

For More Information

Contact methane@gti.energy

[5] https://scripps.ucsd.edu/programs/keelingcurve/2013/05/07/why-are-seasonal-co2-fluctuations-strongest-in-northern-latitudes/