

Referee comments. Reviewer #3.

This manuscript quantifies methane and NO_x emission rates from residential natural gas stoves in Chile and Colombia during three operating stages: steady-state off, steady-state on, and the ignition (pulse) phase using direct measurements. The paper's topic is well aligned with Atmospheric Chemistry and Physics. However, I have some concerns regarding the methodology and the instrument calibration procedures that could be addressed with careful editing

We want to thank the reviewer for his/her insightful and constructive comments. They helped us to improve the manuscript significantly not only for the specialized readers but also for the general community. We believe that we addressed all the comments from the reviewer and again we are thankful since they improved the manuscript significantly.

Major comments:

The manuscript provides a very limited description of how the instruments were characterized and calibrated. This is particularly important when sampling trace gases under conditions involving high temperatures and large humidity variations (for example, water vapor can increase substantially in a closed-box environment during boiling experiments). Laser-based spectrometers are sensitive to changes in water vapor when reporting dry mole fractions, which can affect measurement accuracy. The authors should provide more detailed information on the calibration procedures applied for each instrument to ensure the validity and reliability of the reported data.

This point is well taken. We have expanded the description of how the instruments were calibrated and elaborated on the points brought up by the reviewer. Please see the reviewed manuscript with track changes for further clarity. Key points are summarized here:

- In the experiments water was never brought to a boil. The steady-state on segments lasted typically 5 to 9 minutes, which never resulted in the water boiling. Therefore, water vapor production was somehow limited during the sampling campaign.
- The NDIR instruments also measure the water vapor mixing ratio and use it to apply a correction factor due to the cross sensitivity of water vapor on the desired trace gas being quantified.

- The recorded air temperature inside the static flux chamber naturally increased during the experiments, but the absolute change rarely exceeded 3 to 4°C, which is not large enough to negatively impact the measurements.
- Equipment were calibrated using a Teledyne T750 gas dilutor used to dilute standard from Linde (gas supplier) using zero air to levels that were usually found in the field. Here we provide a table with the calibration performed

Date	Gas	Actual Concentration	Measured Concentration	Units	Source Concentration	Method
6/4/2024	CH4	5.0	4.9	PPM	1002	Teledyne T750
6/4/2024	CH4	19.6	19.7	PPM	1002	Teledyne T750
6/4/2024	CH4	40.7	41.3	PPM	1002	Teledyne T750
6/4/2024	CO2	452.0	439.0	PPM	150200	Teledyne T750
6/4/2024	CO2	892.0	862.0	PPM	150200	Teledyne T750
6/4/2024	CO2	1270.0	1226.0	PPM	150200	Teledyne T750
6/4/2024	CO	1.0	1.2	PPM	440	Teledyne T750
6/4/2024	CO	2.2	2.4	PPM	440	Teledyne T750
6/4/2024	CO	12.8	13.5	PPM	440	Teledyne T750

The composition of natural gas varies substantially on a daily/monthly basis and across regions within the distribution network. Using city-scale composition estimates from 10 to 15 years ago is unlikely to be representative of the natural gas being sampled in 2024. Given that the measurements include 1 Hz observations of methane and ethane, which are the major components of natural gas, the authors could use the observed ethane/methane ratios to better constrain the molar composition and corresponding NG flow calculations. These measurements could also help quantify the uncertainty associated with variability in natural gas composition when calculating the relevant emission factors.

This is an excellent point raised by the reviewer. However, during the measurements in Bogotá, for instance, no ethane measurements were available, and the analysis suggested by the reviewer cannot be included in our manuscript. We have made this limitation more explicit in the manuscript. For clarity, the reported NG composition that we used, is the one reported in the fuels database of each country, which in turn is the source for the calculation of GHG emission inventories.

Action:

Now the text says

“Expressing emissions in terms of the emission factor facilitates the comparison of the observed values to those used in local air quality inventories or those for GHG emission inventories. The city of Bogotá is supplied with a natural gas mixture with 82.2% CH₄, 10.2% ethane, with the remaining being propane, butane, pentane, hexane and CO₂ (UPME, 2016). The mean number of carbon atoms in the Natural Gas supplied to Bogotá is $x = 1.14$, and the LHV is 45.05 MJ/kg. For the case of Santiago, natural gas is richer in methane, with 90.0% CH₄, 6.38% ethane, 0.22% propane, with the remaining fraction corresponding to higher-carbon content gases and CO₂. For Santiago, we used $x = 1.05$ and LHV of 43.7 MJ/kg (CNE, 2009).

The integrated emission rates of methane, CO₂, and NO_x per residential natural gas stove should be calculated and applied to emission inventories and future bottom-up estimates. It would also be useful to assess combustion efficiency using the measured concentrations of CO, CO₂, CH₄, and C₂H₆. In particular, examining how combustion efficiency relates to methane emissions under steady-state operating conditions would provide valuable additional insight.

Contrary to other studies, where the authors have had access to extensive data on stove use (i.e., hours per day of use and number of burners used), our data is not detailed enough to carry out such calculation. We have included an estimate based on the retrieved natural gas consumption registered on the natural gas company reading (which is included in the monthly bill). We still consider that our data can inform emission inventories in two ways:

By providing energy based emission factors (which could be paired with energy demand information to estimate total emissions for a given jurisdiction).

By providing estimates of the contribution from different mechanisms (i.e., continuous leaks, and combustion-related emissions).

Regarding the relationship between emissions and combustion efficiency. With the limited CO emission data available (which were only available for Bogotá), we estimated emission efficiency through the CO/CO₂ ratio, and evaluated if CH₄ emissions showed a relation with this variable. However, no clear trend was observed from the data, and therefore, was not included in the manuscript.

Note that the paper talked about the CO emission rate in the conclusion, but showed no data/analysis at all in the results/discussion. Please add this to the main paper.

Results from the CO measurements were included in the manuscript.

Minor Comments:

Abstract 35 to 40:

“We found that methane emissions from residential stoves in Bogotá and Santiago are over six times higher than the Tier 1 IPCC emission factors currently used in national inventories.”

The authors are conflating methane emissions with emission factors, which are distinct quantities. It would be helpful to clarify which specific quantity you are talking about here. Additionally, corresponding values (with associated uncertainties) should be included here.

We apologize for this mistake we corrected this in the text.

Action: We rephrased in included this information in the text

“The mean methane emission rate during combustion was 410.2 (63.9; C.I. 18.8 – 78.3) mg/h in Bogotá and 331.2 (30.7; C.I. 4.8 – 27.9) mg/h in Santiago, respectively. The equivalent energy-based methane emission factors derived from the data for residential stoves in Bogotá and Santiago are many times higher than the Tier 1 IPCC emission factors currently used in national inventories”

In Abstract 40:

“Notably, continuous leaks and ignition-related emissions, which are excluded from official estimates, contribute significantly to total methane emissions.”

This sentence can be strengthened by using more precise terminology than “official estimates.” For example, “current national/local inventory estimates” would better

convey that results from this study can have a broad impact on closing the emission gap. And again, relative contribution/emission rate, and associated uncertainties should be included here in the abstract.

We agree with the comment from RC 1. We rephrased the text as follows:

Action: We rephrased the text as suggested

Notably, our data suggests that continuous leaks and ignition-related emissions, which are excluded from current national and local emission inventories, contribute significantly to total methane emissions, with around 50% of total methane emissions likely coming from continuous and episodic leaks, with the remaining half of emissions generated during combustion

Introduction:

The current ordering of concepts in the introduction reads disjointed. For example, around Line 60, it would be helpful to provide a brief description of natural gas composition here, rather than in the following paragraph (the first three paragraphs could potentially be combined to form a more concise and clear introduction). First, note that natural gas consists primarily of methane with smaller fractions of other hydrocarbons, and natural gas combustion produces additional greenhouse gases CO₂ and other air pollutants. This would create a clearer logical flow into the later discussion of their respective climate and health impacts.

Additionally, it reads awkwardly to discuss the climate impacts of greenhouse gases without first stating how they are linked to natural gas use. The subsequent transition to “In addition, natural gas combustion generates carbon monoxide (CO), an air toxic...” therefore feels abrupt and not well connected logically.

We agree with this comment. Similar comments were brought up by both reviewers. As a result, we completely reorganized and modified the introduction, attempting to improve clarity and conciseness.

Action: The CO₂ emissions associated with CH₄ combustion are mentioned in the first paragraph. *“Natural gas is widely used for household cooking, with millions of homes globally relying on it for daily meal preparation in developing countries (Quinn et al., 2018, WHO 2014) and with future use projected to increase further (Stoner et al., 2021). It is primarily composed of methane (CH₄), with small amounts of other short-chain hydrocarbons such as butane, propane, and benzene (Chang et al., 2000, Rowland et al., 2024). In addition to carbon dioxide (CO₂), the main driver of anthropogenic climate*

change, natural gas combustion generates air toxics and air pollutants such as carbon monoxide (CO), and nitrogen oxides (NO_x = NO + NO₂). Both, long-term (e.g., Huangfu and Atkinson, 2020), and short-term exposure to NO₂ (Oreallano et al., 2020; Zeng et al., 2021) have been associated with asthma and other deleterious health effects. NO_x is also a key ozone precursor. Unlike other gas appliances that vent emissions outdoors, stoves release combustion byproducts directly indoors, which can impact both household air quality and contribute to overall air pollutant emissions (Barros and Fontes, 2024, Balmes et al., 2023).”

In line 70:

If you are stating that “many” studies exist, it would be helpful to include additional citations, as only one reference is currently provided.

We agree with the comment, and we included two more references regarding the fugitive methane leaks supporting the statement.

Action: Now we included two more references and the text says:

“Most studies on methane emissions from the energy sector have focused on fugitive emissions from oil and gas exploitation, as well as leaks from natural gas distribution networks (Boothroyd et al., 2016; Hendrick et al., 2016; Staniaszek et al., 2022).

Refs cited:

Boothroyd, I. M., Almond, S., Qassim, S. M., Worrall, F., and Davies, R. J. (2016). Fugitive emissions of methane from abandoned, decommissioned oil and gas wells. *Science of the Total Environment*, 547, 461-469. Chan, S. H., and Wang, H. M.: Effect of natural gas composition on autothermal fuel reforming products, *Fuel Process. Technol.*, 64, 221–239, 2000.

Hendrick, M. F., Ackley, R., Sanaie-Movahed, B., Tang, X., and Phillips, N. G. (2016). Fugitive methane emissions from leak-prone natural gas distribution infrastructure in urban environments. *Environmental Pollution*, 213, 710-716.

In line 80:

“While some studies have reported emission factors for specific countries and fuel sources (Smith et al., 1993. Lebel et al. 2022), these datasets remain limited, creating uncertainties in global emission inventories.”. The statement “these datasets remain limited” would benefit from additional clarification. It is not clear

what the limitation refers to. Is it the limited number of appliances sampled, large uncertainties in the reported emission rates, restricted geographic coverage, or any other factor, like too few studies being published? Clarifying these limitations would strengthen the argument and more clearly motivate how the present study helps address the resulting uncertainties in global emission inventories.

This was expanded in the manuscript and the whole paragraph was re-written in the newer version of the manuscript taking into account the comments provided by both reviewers.

Action:

Now the text says “Natural gas appliances emit methane through incomplete combustion as well as through small but continuous methane leaks, even when the appliance is off (Lebel et al., 2022). Household cookstoves, although small in the quantity of gas burned when compared to gas furnaces, play a crucial role in domestic energy use and contribute to GHG emissions. In developing countries, household fuel consumption represents a significant share of total energy use, yet the emission factors associated with small-scale fuel combustion are often poorly quantified compared to larger-scale sources (Levine, 1996). While some studies have reported emission factors for specific countries and fuel sources (e.g., Smith et al., 1993. Lebel et al. 2022), these datasets remain limited due to the overall scarcity of studies and the corresponding lack of geographic diversity, creating uncertainties in global emission inventories. These uncertainties hinder the accuracy of climate models and the development of effective mitigation and adaptation strategies. As research has shown in other natural gas applications, underestimating leakage of methane emissions can lead to significant misrepresentations of its full contribution to warming (e.g., Alvarez et al., 2012; Comer et al., 2024)

In line 90:

It is unclear whether this statement refers only to methane emissions or whether it also applies to combustion byproducts. Seems like you are only talking about methane, but please clarify which species are being discussed to avoid ambiguity.

See below comment since this and the next corresponds to the whole paragraph and we corrected the whole paragraph

In line 95:

17% and 15% do not appear particularly large on their own. If the intention is to emphasize a substantial expansion, it would be helpful to provide supporting data on how rapidly these shares have changed over time, rather than citing only the current percentages.

Done. We made it now explicit to remove the ambiguity on the sentence and we also corrected the paragraph regarding to the fact of the expansion Natural Gas usage in Colombia and Chile; now we corrected the text.

Action:

We specified supported and accurate refs and numbers for this statement, now the text says:

“As evidence of emissions from natural gas stoves in the global south are scant, building on previous research, this study aims to quantify both GHG emissions as well as other air pollutant emissions, from natural gas cookstoves in Chile and Colombia. The two countries have experienced a rapid expansion in the use of natural gas as household fuel. In Colombia, natural gas currently supplies 17% of residential energy demand, growing from just 2.8 PJ in 1991 to 56.4 PJ per year in 2021, a 20-fold increase, with emissions from its residential use currently accounting for 2.44 MtCO_{2eq}. According to government estimates, most of its residential use is in direct-heat applications, overwhelmingly in cooking. Similarly, in Chile, adoption of natural gas has increased in recent years, and currently supplies some 13% of the residential energy demand, and accounts for 1.08 MtCO_{2eq} (MMA, 2018). According to Chile’s official energy balances, final natural gas consumption increased from 73.8 PJ in 2004 to approximately 83.7 PJ in 2020. Although the increase in total final natural gas demand was moderate over this period, the residential segment became progressively more relevant (CNE, n.d.). In 2020, aggregated commercial, public and residential (CPR) use accounted for 12.7% (32.5 PJ) of final natural gas consumption, while in 2004 accounted for 5.4% (18.2 PJ).”

In line 100:

When people refer to a “cleaner” fuel, they are often considering a broader range of pollutants beyond NO_x, such as PMs, CO, SO₂, etc. This sentence gives the false impression that natural gas is not necessarily cleaner than oil or wood.

We agree with this comment we polished the sentence to be more clear:

Action:

Now the text says

“Because natural gas combustion emits significantly lower amounts of particulate pollution, methane, and other harmful species compared to biomass and other solid and liquid fuels, the quantification of the emission rates of air pollutants from residential gas stove use has been overlooked, resulting in limited data to characterize this atmospheric pollution source. By measuring emissions during different operational phases -including while the stove is off, during ignition, and during combustion- this research seeks to refine household emission estimates, providing evidence to improve GHG inventories, indoor air quality, and inform policy on residential energy use and climate mitigation strategies.

In line 208: missing a period:”...and are likely a lower bound estimate of the true value. Our results show that...”

We apologized for this mistake.

Action:

Now the text says “Following the procedure outlined in the methods, the mean (median) energy-based emission factors from the observations are equivalent to 80.8 (16.2) kg CH₄/TJ for Bogotá and 41.2 (3.66) kgCH₄/TJ for Santiago. Our results suggest that the true variability in emission rates, which spans several orders of magnitude, is much larger than the uncertainties associated with the method used (see Supplementary Material). “

Additionally, what is “true value” referred to here?

We removed the sentence and change this part of the discussion to conclusions section.

Action:

Now this paragraph was corrected as follows:

“These values are over many times higher than those used in the construction of national inventories (i.e., the IPCC Tier 1 emission factor for natural gas combustion is 5.0 kg CH₄/TJ) for Colombia and Chile respectively. Over 42% of the samples in Santiago were above this Tier 1 emission factor, with a similar percentage observed in Colombia. Furthermore, this first-of-its-kind study in Latin America, supports the idea that continuous methane leaks might be larger and more commonplace than previously thought, and are likely to emit significant amounts of methane. These types of continuous releases were only recently incorporated into IPCC GHG emission guidelines, but further evidence to constrain its true value is still needed. Despite the

potential limitation from the sample size, the findings of this work suggest that the contribution to national methane emissions from residential use of natural gas are significantly underestimated in both countries. The findings for both cities are consistent with recent methane and NO_x emission estimates carried out in the U.S. (Lebel et al., 2022).”

In line 205: What are the causes of the two outliers, and is it a reasonable judgment to exclude them?

After carefully evaluating the comments from both reviewers, we have decided to leave those outliers in the analysis, as we think those were valid datapoints, except for one specific datapoint for the case of Chile. We now emphasize in the manuscript the fact that, due to the small sample size, those outliers have a strong impact on the mean, and we report the mean and the median, together with confidence intervals.

In conclusion: Need to add some description of the analysis/value for CO emission since the authors are discussing them here.

We have now included a description of the CO emission rates for both cities.