

Response to the Reviewers' Comments

Thank you for the comments provided and for the careful review of the manuscript. Below, we present my point-by-point responses based on the observations made.

Referee #1, 15 Apr 2025

This paper investigates greenhouse gas emissions associated with residential kitchens. The topic is relevant because it analyzes natural gas and liquefied petroleum gas that are widely used in many countries for cooking and can affect human health and ultimately might have climate impacts. Electric stoves may be ideal to minimize emissions, but there are challenges in their implementation in many countries and Brazil can be a typical example. The manuscript is clear and concise, which is really appreciated, and results are clearly illustrated and supported by the data. I have only one concern before recommending the publication in AMT: The authors claim in the text and in the supplementary material that they have measurements from 30 different kitchens. However, the discussion is focused in SP_CASA02 and SP_CASA03. I miss the link between these two examples and a global conclusion that includes measurements in all kitchens. Saying that, are the results of emission rates and emission factors representative of all cases or only of SP_CASA02 and SP_CASA03?

Response: *Thank you very much for your careful reading and valuable feedback. We appreciate your positive comments regarding the clarity and relevance of the manuscript.*

Regarding your concern: SP_CASA02 and SP_CASA03 were selected as illustrative examples to describe the temporal variation in emissions from the two main types of fuel studied (liquefied petroleum gas and natural gas, respectively). These cases were used only in the initial analysis to exemplify the emission dynamics under each fuel type. Similar temporal patterns were observed across the remaining households, which is why we focused on just these two for this part of the discussion.

However, all subsequent analyses — including variability in concentrations, emission rates, and emission factors — were based on data from the full sample of 30 households. We will clarify this point in the revised manuscript to ensure it is clear that the broader results are representative of the complete dataset.

We will also incorporate your minor suggestions directly into the text. Thank you again for your thoughtful review.

MINOR REVISIONS

- **Introduction: I miss the objectives at the end of the introduction section. This can enrich the manuscript.**

Response: To make the objectives clearer, the statement of the objectives was separated from the final paragraph of the Introduction and presented more explicitly.

The statement is now as represented next: “The objective of this research is to gather data on cooking fuel usage in Brazilian kitchens, focusing on the two most commonly used sources: liquefied petroleum gas and natural gas. The study seeks to evaluate the emissions of CO₂, CH₄, and NO_x, resulting from the use of gas stoves in urban areas,

and to analyze the associated indoor air quality, expanding the analysis of data on indoor concentrations and emissions, which is limited.”

- **Line 119: Give a reference for US EPA**

Response: Reference for US EPA given in Line 141 (U.S. EPA, 2017)

- **Line 120: Give a reference for European Norm (EN)**

Response: Reference for European Norm (EN) given in Line 141 (Ecotech Inc., 2020).

- **Line 125: Give a reference for Integrated Cavity Output Spectroscopy technique.**

Response: The information about the Integrated Cavity Output Spectroscopy technique was taken from the User Manual of the equipment used for greenhouse gas concentration measurement. This reference is cited as suggested as (ABB Inc, 2020) in Line 146.

- **Lines 133-135: Give a reference for equation 1.**

Resp.: IPCC (2006) reference included in the citation of equation 1. Reference: Intergovernmental Panel on Climate Change (IPCC). (2006). 2006 IPCC Guidelines for National Greenhouse Gas Inventories, Volume 2: Energy. Available at: <https://www.ipcc-nggip.iges.or.jp/public/2006gl/>

- **Lines 146-147: Can you explain better how air exchange rate was determined?**

Response: Further details in a new topic 4 “Air Exchange Rate” of the supplement material with more description

Text add: “The air exchange rate was determined by exponential adjustment after kitchen ventilation to ensure similar concentrations to those in the outdoor environment.

As shown in the protocol, in all homes, after ventilation, cycles 1, 2, and 3 consist of first stabilization with the kitchen closed (St_OFF) and then stove stabilization (St_ON). After stabilization, there is a period with the stove turned off (OFF) and the kitchen still closed. During this period of each cycle, an exponential adjustment was made to determine λ , which is the air exchange (ACH).

The best adjustment was chosen for each home, which was not necessarily in the same cycle. The figure exemplifies the exponential adjustment made for home 06, where the best adjustment was found in cycle 2. ...”

- **Give a reference for equation 2**

Response: The same reference of equation 1 (IPCC, 2006), given in equation 2 citation.

- **Line 158: How did you compute normalized concentration profiles?**

Response: The normalized concentration profiles were computed by the standardization method (z-score). This explanation now is included in Line **185**. “The concentrations of the gases analyzed (CH₄, CO₂, and NO_x) were normalized, making it possible to evaluate the influence of each gas on the others and their behavior in

relation to the conditions maintained at the time of measurement, as they had different magnitudes. This normalization was done through the standardization method (z-score), which transforms the data, so it has a mean of zero and a standard deviation equal to one”.

In order to enhance comprehension of the z-score concept, a detailed explanation has been included in the Supplementary Material topic 3 “Normalized Concentration”, line 60-73:

Text add: “To facilitate the analysis and comparative evaluation of the three distinct gases (methane, carbon dioxide, and nitrogen oxides) in a ensembled chart representation, the concentration data were normalized utilizing the z-score methodology. This approach enables the determination of how each measurement deviates from the mean in terms of standard deviations (Brase and Brase, 2016; Willard, 2016). ...”

- **Figure 5a: I recommend reducing y-axis to values of 0.2, even though you miss the outlier in cycle 2. Another recommendation could be reducing to values of 0.3 and including the outlier in cycle 2. Also, Figure 5 caption can be improved.**

Response: The legends for Figures 5 and 6 have been renamed. The y-axis was adjusted in Figure 5a and, in the adjustment, the opportunity to reduce the y-axis to the value of 0.15 was identified.

New Figure 5 renamed - Variability in gas concentrations across the different monitored households using Natural Gas (NG) and Liquefied Petroleum Gas (LPG): (a) methane (CH₄), with the lower explosive limit indicated at 50 ppb; and (b) carbon dioxide (CO₂), with the health effect threshold set by NIOSH at 2000 ppm.

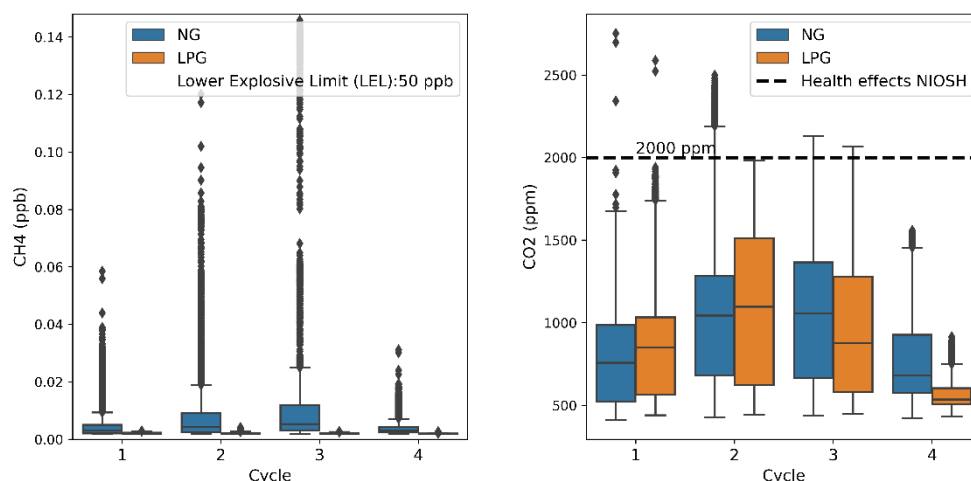
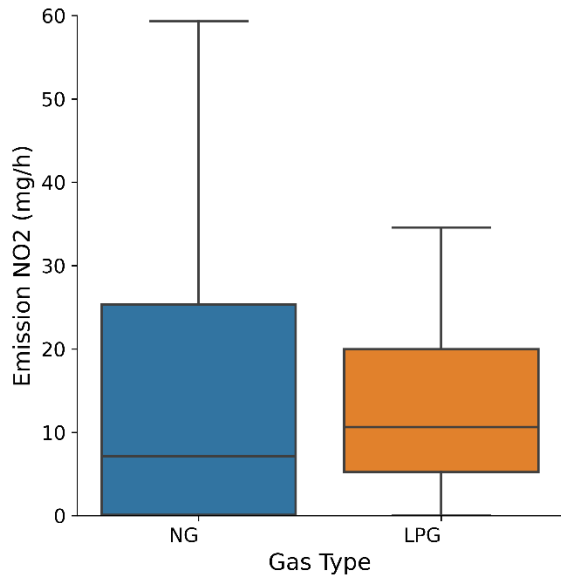


Figure 6 also renamed - Variability in gas concentrations across the different monitored households using Natural Gas (NG) and Liquefied Petroleum Gas (LPG): (a) nitrogen oxide (NO); and (b) nitrogen dioxide (NO₂), with the health WHO recommendation for 1 hour at 106 ppb

- **Figure 7c: Again, I think that y-axis should be re-scaled to enhance the Box-Whisker plot.**

Resp.: Graph adjusted to the y-axis

New Figure 7c:



- **I recommend joining Discussion and Conclusion in just one section.**

Response: The Discussion and Conclusion sections were merged into a single section. The text was revised to improve readability and coherence, as well as to eliminate redundancy.

- **Line 225: Give link/reference to Abregas**

Response: Reference for Abregas given in Line 258

ASSOCIAÇÃO BRASILEIRA DAS EMPRESAS DISTRIBUIDORAS DE GÁS CANALIZADO (ABEGÁS). Uso de gás natural canalizado no Brasil – <https://www.abegas.org.br/arquivos/78359>

- **Line 274: Give link/reference to Sistema de Estimativas de Emissões e Remoções de Efeito Estufa**

Response: The SEEG reference has been properly incorporated into the text, and its complete bibliographic information has been added to the References list, as recommended.

SEEG: Sistema de Estimativas de Emissões de Gases de Efeito Estufa. Lacunas de Dados Primários para Estimativas de Emissões de Gases de Efeito Estufa: Avanços Necessários e Recomendações., Observatório do Clima, <https://seeg.eco.br/wp-content/uploads/2024/03/SEEG-LACUNAS-DADOS.pdf>, accessed: 23 Jul. 2025, 2021.

Referee #3, 07 Jun 2025

The manuscript presents methods and results from an investigation of indoor greenhouse gas (GHG) emissions from household kitchens in the metropolis of São Paulo, Brazil. Measurements of indoor GHG concentrations and emissions are very scarce, especially in developing countries, hindering the assessment of their impacts on national GHG inventories and human health. Despite the limited number of households investigated (30), the study provides valuable information, reporting high variability in CH₄ emission factors and values up to three times higher than IPCC estimates. The manuscript requires minor corrections, particularly in the description of the methods.

Response: *Thank you very much for the detailed and constructive comments, which will undoubtedly contribute to the improvement of our manuscript. We agree with the points raised and will incorporate the suggested revisions and corrections one by one. In particular, we will focus on improving the description of the measurement protocol, standardizing units and terminology, enhancing the presentation of data and figures, and addressing the technical corrections indicated.*

Once again, we sincerely appreciate the reviewer's careful evaluation and valuable contributions.

Specific comments

- **Introduction: I suggest to include a brief discussion about similar studies around the world. What are the main references supporting, for example, the IPCC estimates on GHG emissions by residential kitchens?**

Response: Brief discussion included in the Introduction about similar studies around the world. "Lebel et al. estimated total emissions of 28 Gg CH₄/year (28,000 tonnes) from residential sources in the United States, which exceeds the estimates provided by the U.S. EPA. Their detailed breakdown revealed that burners emit 2.7 Gg CH₄/year (2,700 tonnes) during steady-state-on conditions and 1.1 Gg CH₄/year (1,100 tonnes) from on/off pulses, while 21.2 Gg CH₄/year (21,200 tonnes) is attributed to steady-state-off emissions from stoves, indicating substantial leakage even when appliances are not in active use. Complementary findings by Merrin and Francisco estimated 2.7 Gg CH₄/year (2,700 tonnes) from burner use, aligning closely with Lebel et al., but also reported emissions of 3.3 Gg CH₄/year from stoves (3,300 tonnes) and 5.0 Gg CH₄/year from ovens (5,000 tonnes), suggesting that multiple components of cooking systems contribute significantly to overall methane release. Similarly, Fischer et al. identified 1.6 Gg CH₄/year (1,600 tonnes) from general cooking equipment in California alone, reinforcing the relevance of regional assessments."

- **Section 2.3: the description of the measurement protocol should be improved, in the main text of the manuscript. The scope of this journal leans toward the development and validation of measurement techniques. As such, to enable reproducibility of the protocol you have adapted, it should be better described. For example, you have used the terms "cycles", "modules", "Inject Gas" without a clear description of what they mean, at least in the main text. What is the difference between the "St_ON" and "ON" modules? The reader should be able to understand the measurement protocol from the main text. Consider including a diagram to illustrate the steps of the measurement protocol.**

Response:

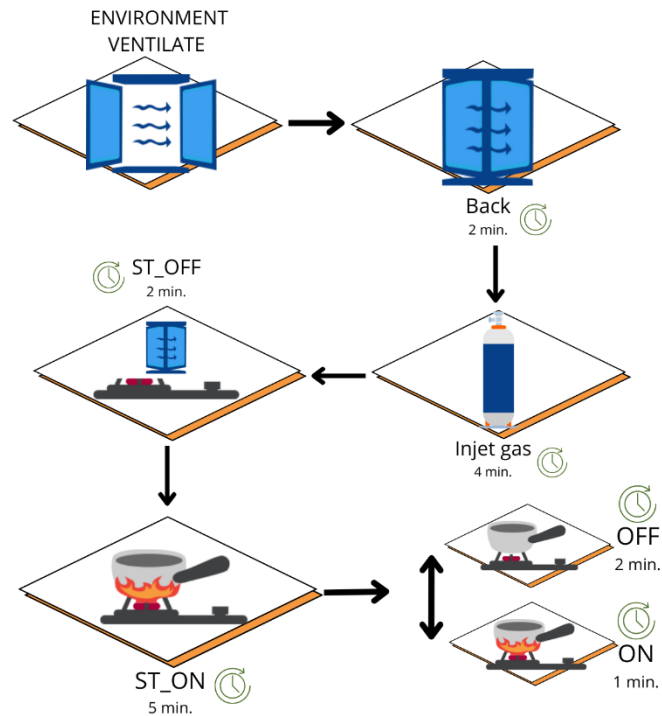
Original text: The cycles and durations of each module used in the experiments are detailed in Table 1, along with the specific modules assigned to each cycle. The cycle durations were adapted from the study conducted by Lebel et al. (2022) and tailored to then context of Brazilian residences. Preliminary tests conducted prior to the measurements identified patterns that influenced the timing of each module (Lebel et al., 2022). For example, the "Inject Gas" module was performed over a period of 4 minutes at the beginning of the measurements. This duration was selected based on observations that CO₂ concentration values stabilized within this timeframe, allowing for accurate calibration and air exchange rate assessment. In the "State ON" module, a duration of 5 minutes was determined to be sufficient for CO₂ and CH₄ concentrations to stabilize while the burner was active. For the "ON" and "OFF" modules, distinct gas behavior patterns were observed: a rapid increase in gas concentrations during the "ON" phase, followed by a gradual decay during the "OFF" phase. These changes were effectively captured within 1 minute for the "ON" module and 2 minutes for the "OFF" module.

Text changed: The experimental modules were organized based on the methodological framework proposed by Lebel et al. (2022), with adaptations made to accommodate the specific context of Brazilian households. The nomenclature and respective cycle durations of each module are detailed in Table 1. These durations were refined through preliminary tests conducted before data collection, which identified behavioral and operational patterns influencing the temporal configuration of each module (Lebel et al., 2022).

In the " STATE ON" module, a duration of five minutes was established as adequate for the stabilization of CO₂ and CH₄ concentrations while the burner remained active. In the "ON" and "OFF" modules, distinct gas emission dynamics were observed: a rapid increase in concentrations during the "ON" phase, followed by a gradual decay during the "OFF" phase. These changes were effectively captured within 1 minute for the "ON" module and 2 minutes for the "OFF" module.

Figure 4 illustrates the sequence and duration of each module applied during the first measurement cycle. The process begins with environmental ventilation, followed by a 2-minute background stabilization phase ("Back"). Next, a 4-minute CO₂ injection is performed ("Inject Gas"), after which the cycle proceeds through the ST_OFF and ST_ON modules. During the ST_OFF phase, the burner remains off, allowing gas concentrations to return toward baseline levels over 2 minutes. In contrast, the ST_ON phase involves igniting the burner under typical cooking conditions, with emissions monitored for 5 minutes to ensure stabilization of CO₂ and CH₄ concentrations. Finally, the ON and OFF phases are alternated to capture transient emission dynamics, lasting 1 and 2 minutes, respectively. The injected gas (in Inject Gas step) was used to ensure accurate CO₂ concentration values for assessing the enclosed environment and the air exchange rate. Additional methodological details are provided in the Sections 1 and 4 in the Supplementary Material.

Figure 4. Sequence and durations of the measurement protocol applied to cycle 1. The ON/OFF phases were repeated three times within each cycle, followed by a ventilation phase to reset the environment.



- **Did you encounter problems following this measurement protocol? Were you able to use data from the 30 households in the calculation of emission rates? Would you recommend changing something in the protocol? This is important information that may provide guidance for future studies.**

Response: In our experience, we did not encounter many difficulties in applying the protocol, other than those incorporated in the text. The measurement cycles were completed in all 30 households, and the data collected were incorporated into the calculation of emission rates. In this context, some data with negative values and possible external interference were observed, which was reflected in the difficulty in calculating the decay rate. For future applications, it would be advisable to consider methodological refinements, as the use of CO₂ in the Inject Gas module - originally intended for assessing air exchange rates - may not have been the most appropriate choice. As highlighted by Lebel et al. (2022), employing a tracer gas such as ethane, which is not emitted during combustion, would minimize potential interference with background CO₂ levels and improve the accuracy of air exchange calculations. Therefore, we recommend adopting a similar approach using ethane or another inert tracer gas in future studies.

- **Line 130 you have mentioned a CO₂ cylinder, what was it used for? In line 147 you have mentioned a controlled release of CH₄.**

Response: In all homes, during cycle 1 and under closed conditions, 450 mL of CO₂ were injected using a cylinder (Inject gas). The goal was to calculate the decay rate based on the decrease in gas concentration within the environment. However, this volume proved insufficient in some residences, making it impossible to estimate the

decay rate. As a result, during subsequent cycles, after the stabilization of the flame being turned on (St_ON), the most suitable decay curve was identified, and the emission rate was determined based on the cycle that showed the best curve fit. More description in section 4 of **Supplementary Material**.

Text added in topic “4 Air Exchange Rate” - line 83 SM: “It is worth mentioning that preliminary tests were conducted to validate the methodology prior to the start of measurements in the households. During these tests, the decay curve was simulated by injecting CO₂ into the environment using a pressurized cylinder. The injected volume was intentionally kept low to avoid saturating the environment and to allow a quick return to normal conditions, thus ensuring greater agility in conducting the sampling procedures in the homes.

However, given the typical conditions of Brazilian houses, which are generally naturally ventilated, the injected volume in some residences was not sufficient to accurately determine the decay rate of the gas. The injection was carried out in all homes during Cycle 1, immediately after the first kitchen closure, also serving to assess the sealing effectiveness. In some cases, the sealing was compromised due to air infiltration through windows, even when closed.

Considering these limitations, the analyses were based on the decay curve observed after the stove was turned off, following the stabilization of the concentration while the stove was in operation. This approach proved to be more effective, as the concentrations reached during combustion were higher than those achieved through artificial CO₂ injection, as previously described and illustrated in the figure.”

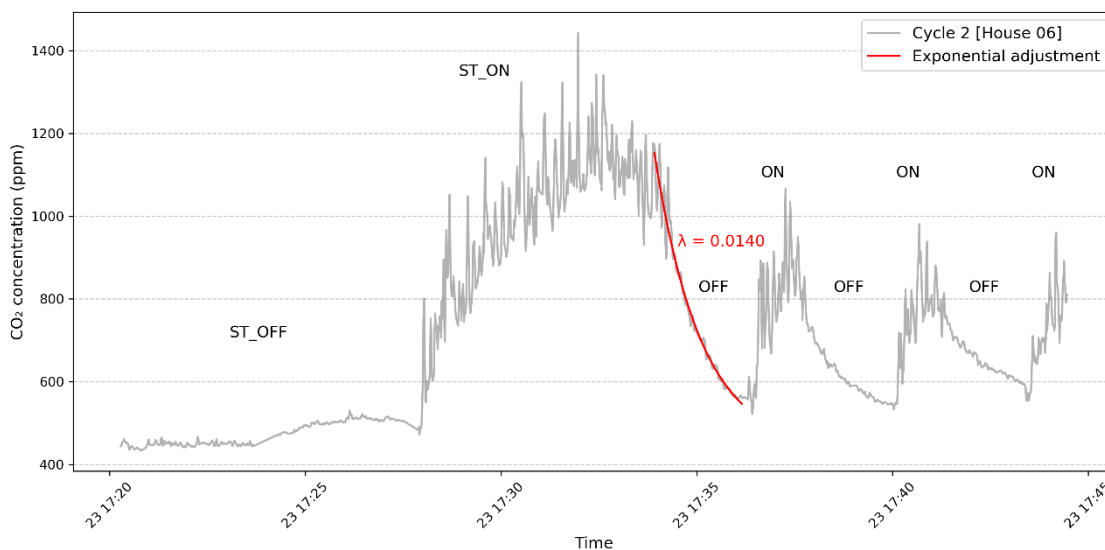
- **Section 2.5: equations 1 and 2 should be better described. Please inform the units of the quantities involved in the calculation of emission rates and emission factors. In which cycle/module was the background concentration determined? How was the concentration rate of change ($\Delta C/\Delta t$) calculated, in which cycle/module? Did you fit a line to obtain the rate of change, or have you considered the conditions at start and end times only? What values of LHV have you considered in equation 2?**

Response: More details of the equations, as well as the values, were inserted into the text of the article in the rewrite of the topic "2.5 Emission Estimation Methodology". New topics ("4 Air Exchange Rate" and "5 Characteristics of Natural Gas and LPG in Brazil") with more details were also inserted in the supplementary material.

- **Lines 146-147: Consider including a plot to illustrate the concentration decay and calculation of the ACH. The calculation of $\Delta C/\Delta t$ could also be illustrated with an example. That would be informative to the readers.**

Response: Further details in a new topic 4 of the supplement material – text with more description and an illustrative figure

Figure S1. Example of decay curve with House 06 in Cycle 2 using the exponential adjustment.



- **Line 188: what do you mean by “steady state ON”? Is it the same as “St_ON”? Please standardize the terminology.**

Response: The term “steady state ON” was replaced by “St_ON”.

- **Table 2 and Table 3: There are too many significant figures in the reported emission rates. Considering the measurement uncertainties, are you really able to provide 2 decimal figures in the emission rates? Also, I recommend reporting median and interquartile values, instead of mean values, since the data distribution is very skewed. In Table 3, I recommend including the IPCC and the National emission factor estimates, for a direct comparison.**

Response: New tables 2 and 3 with median, quartiles 1 and 3. Table 3 with the factor emission, including the IPCC and National emissions factors.

Table 2. Emission rate for CO₂, CH₄, and NO₂ for NG and LPG with Median, First and Third quartile (Q1 and Q3).

Compound	Natural Gas (NG)			Liquefied Petroleum Gas (LPG)		
	Median	Q1	Q3	Median	Q1	Q3
CO ₂ (g/h)	238	124	322	282	190	404
CH ₄ (mg/h)	121	0	504	0	0	2
NO ₂ (mg/h)	1	0	9	1	0	11

Table 3. Emission factor for CO₂, CH₄, and NO₂.

Natural Gas (NG)					
Compound	Median	Q1	Q3	Brazil Factor	IPCC
CO ₂ (kg/TJ)	23543	12315	31834	56100	56100
CH ₄ (kg/TJ)	12	0	50	1.1	5
NO ₂ (kg/TJ)	0	0	1	-	-
Liquefied Petroleum Gas (LPG)					
Compound	Median	Q1	Q3	Brazil Factor	IPCC
CO ₂ (kg/TJ)	27337	18403	39078	63100	63100
CH ₄ (kg/TJ)	0	0	0	1	5
NO ₂ (kg/TJ)	0	0	1	-	-

- **Line 195: what could result in “negative emissions”?**

Response: The calculation considers the decay rate, which varies according to the level of kitchen sealing. To simulate more enclosed kitchens in open-plan layouts — such as American-style or integrated kitchen-dining areas — additional sealing materials were used. In other homes, closure was achieved through windows and doors. The experiment was conducted in real households, within the Brazilian context, where homes are generally not tightly sealed. In some cases, negative values were observed, indicating that even with the kitchen closed, effective sealing was not achieved. This represents a limitation of the study, as it was conducted under real-life conditions to reflect the typical Brazilian housing reality.

Add text in line 229: “Negative emissions values indicate that, even with the kitchen closed, there was a gas leak, highlighting 230 the difficulty of achieving proper sealing in some homes with high ventilation, which is a common characteristic in Brazilian residences.”

- **Line 201: NO₂ emission rates were calculated based on the concentration rate of change ($\Delta C/\Delta t$). They should not be affected by the background concentrations, unless the kitchens were not properly sealed.**

Response: Same reason of the “negative values”.

Add Text 235: "External interference may also occur for the same reason, due to inadequate sealing and high air exchange in some homes."

- **Line 205 (for example): standardize the emission factor units for each pollutant, to facilitate the comparison. For example, here you have used kg CH₄/TJ, while in Table 3 you have used mg/MJ.**

Response: New table 3 with the standardized emission factor for the kg/TJ unit.

Technical corrections

Line 10: Reformulate the sentence. “Close values” is not a scientifically sound term.

Correction: Line 10: The term “close values” reformulated and replaced by “consistent values”.

Fig1: Include the data source in the figure caption (SVMA, 2022).

Correction: Figure 1: Data source (SVMA, 2022) included in the figure caption.

Line 76: Rephrase the sentence.

Correction: Line 86: The sentence “Considering to analyze the emissions of CO₂, CH₄, and NO_x, emitted by the use of gas stoves in cities” reformulated and replaced by “The study seeks to evaluate the emissions of CO₂, CH₄, and NO_x resulting from the use of gas stoves in urban areas”.

Line 150: “M” is not included in Equation 2.

Correction: "M" is not actually used in Equation 2, so it has been removed from the explanation of terms in Equation 2.

Standardize the usage of “LPG” and “NG” along the whole text and figures. For example, in Figures 5, 6, 7 you have used “GLP” and “GN”.

Correction: All instances where the terms 'LPG' or 'NG' were previously used have been reviewed and corrected to consistently reflect the appropriate terminology, as exemplified in Lines 239 and 240, and Figures 5, 6, and 7.

Figure 5: Consider using a y axis log scale for CH₄ concentrations.

Response: A logarithmic scale on the y-axis for CH₄ concentrations in Figure 5 was considered. However, this approach reduced the visibility of the concentration sensitivity in the results, so we opted to retain the linear scale to better represent the data.