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Emissions from fuel combustion by stoves in residential kitchens in São Paulo – Brazil

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Supplementary Material

S1 Protocol

S1.1 Kitchen Dimensioning and Initial Preparation

To prepare for the sampling process, the kitchen's dimensions (width, height, and length) were measured using a laser measuring device to calculate its volume accurately. During this step, sampling tubes connected to the monitoring equipment were installed on a tripod, which was adjusted to the height of the person cooking and positioned near the stove for optimal data collection.

Following the dimensioning process, initial preparation and ventilation were conducted to stabilize gas concentrations. Two fans and, windows and doors open were used to ventilate the kitchen, ensuring that gas concentrations, such as CO_2, CH_4, and NO_2, reached levels close to the external environment. This procedure allowed for the accurate measurement of baseline gas concentrations, serving as reference points for subsequent stages. Stabilization was continuously monitored until the concentration values remained constant, ensuring reliable baseline measurements.

S1.2 Back_Module (Background State)

After initial stabilization, the kitchen was sealed or closed to begin the "Back" phase. At this stage, all stove burners were turned off, and the room remained closed to assess possible gas infiltration or residual concentrations. For approximately two minutes (a duration determined through preliminary testing), concentrations of CO_2, CH_4, and NO_2 were continuously monitored to ensure the space was completely isolated from external sources, providing an accurate baseline for "off" state gas levels.

S1.3 Inject Gas_Module (Gas Injection)

Immediately after the "Back" period, the gas injection phase, referred to as "Inject Gas," was performed. During this stage, 450 mL of CO_2 was injected into the kitchen using a syringe connected to a CO_2 cylinder. This standardized injection allows the calibration of measurement instruments and helps determine the air exchange rate within the room. CO_2 concentrations were monitored over time until stabilization and subsequent decay, enabling the calculation of correction factors and the assessment of the room's insulation efficiency.

S1.4 ST_OFF_Module

After the Inject Gas phase, the Stated OFF phase was conducted to allow gas concentrations (CO_2, CH_4, and NO_2) to return to baseline levels. This phase lasted approximately two minutes.

S1.5 ST_ON_Module

Following the "Off" phase, one of the stove burners was lit to initiate the "Steady-State On" phase. To simulate typical equipment use, a pot with water was placed on the burner to recreate real cooking conditions. During this phase, CO_2, CH_4, and NO_2 concentrations were monitored while the burner remained lit until stabilization was reached. This stage lasted approximately five minutes, capturing maximum emission levels associated with burner use.

S1.6 OFF_Module

In the OFF phase, the previously lit burner was turned off for about two minutes. This phase was repeated three times in each cycle.

S1.7 ON_Module

After two minutes, the burner that was previously lit during the Stated ON phase was reignited for one minute. This phase was also repeated three times in each cycle.

S1.8 Background Period (Back)

After the "On" phase, a new ventilation period, referred to as "Back," was carried out to reduce gas concentrations in the environment. This step ensures that the measurement space returns to conditions similar to the external environment before proceeding with the next gas injection. This stabilization is crucial to ensure that subsequent measurements are based on a new baseline.

S1.9 Cycle Repetition

The entire measurement cycle was repeated for each stove burner, ensuring the collection of representative data under varying usage intensities. The structured process described ensures standardization and consistency of the data, enabling future comparisons and reliable inferences about GHG emissions associated with cooking gas use in Brazilian households.

S2 Contact with post-selection volunteers

After registering the volunteers, the researcher contacted those interested to confirm their participation, provide additional details, and address any questions they might have. Once the interest was confirmed, further information about the kitchen was requested to prepare for the measurements. Another topic discussed and agreed upon was the date and time of the volunteer's availability to schedule the sampling.

During these discussions, the sampling duration, which ranged from two hours and 30 minutes to three hours, was consistently emphasized. Information about the equipment and the noise they might produce was also shared.

On the day of the visit, upon arriving at the residence, each homeowner was asked to sign the Informed Consent Form, which explains the project details and requests authorization for all activities conducted during the sampling, including taking photographs at the residence.

After the measurements were completed, a copy of the Informed Consent Form was sent to each volunteer.

Table S1 provides detailed information about the homes included in the study, including the number of houses, the study dates, house types, the use of plastic sealing, the type of gas used for cooking, and other relevant data.

S3 Normalized Concentration

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). The z-score is calculated based on the Equation 1:

$$z = \frac{x - \mu}{\sigma} \tag{1}$$

where:

z is the z-score;

x is the measured concentration;

μ is the mean of the variable;

σ is the standard deviation of the variable.

A z-score in close proximity to zero indicates that the measurement aligns closely with the mean value. Furthermore, a positive z-score signifies that the measurement exceeds the mean, while a negative z-score indicates that it falls below the mean. This normalization process is pivotal for appropriately analyzing and comparing the concentrations of the gases in question.

S4 Air Exchange Rate

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

Table S1. Information about the volunteer residences.

City	Region (Zone)	Nomenclature	Date	Kitchen Concept*	Stove Age	Residence Type	Gas Type
São Paulo	Western	SP_CASA01	08-21-24	Open	6	House	NG
São Paulo	Western	SP_CASA02	08-22-24	Closed	10	House	LPG
São Paulo	Western	SP_CASA03	08-22-24	Closed	11	Apartment	NG
São Paulo	Western	SP_CASA04	08-23-24	Closed	15	House	LPG
São Paulo	Southern	SP_CASA05	08-23-24	Closed	10	Apartment	NG
São Paulo	Southern	SP_CASA06	08-23-24	Closed	3	Apartment	NG
São Paulo	Southern	SP_CASA07	08-27-24	Closed	20	Apartment	NG
São Paulo	Southern	SP_CASA08	08-27-24	Closed	10	Apartment	NG
São Paulo	Southern	SP_CASA09	08-27-24	Closed	10	Apartment	NG
São Paulo	Western	SP_CASA10	08-28-24	Open	25	Apartment	NG
São Paulo	Southern	SP_CASA11	08-29-24	Closed	40	House	LPG
São Paulo	Western	SP_CASA12	08-29-24	Closed	7	Apartment	NG
São Paulo	Western	SP_CASA13	08-29-24	Closed	3.5	Apartment	NG
São Paulo	Southern	SP_CASA14	09-02-24	Closed	7	House	LPG
São Paulo	Western	SP_CASA15	09-02-24	Closed	10	Apartment	NG
São Paulo	Western	SP_CASA16	09-04-24	Closed	10	Apartment	NG
São Bernardo do Campo	MASP**	SP_CASA17	09-05-24	Closed	5	House	LPG
São Bernardo do Campo	MASP**	SP_CASA18	09-05-24	Closed	10	House	LPG
São Paulo	Southern	SP_CASA19	09-05-24	Closed	1	Apartment	NG
São Paulo	Western	SP_CASA20	09-06-24	Closed	14	House	NG
São Paulo	Western	SP_CASA21	09-10-24	Open	10	House	LPG
São Paulo	Western	SP_CASA22	09-10-24	Open	4	Apartment	NG
São Paulo	Western	SP_CASA23	09-10-24	Closed	10	House	LPG
Mogi das Cruzes	MASP**	SP_CASA24	09-11-24	Open	5	Apartment	NG
Poá	MASP**	SP_CASA25	09-11-24	Closed	11	Apartment	LPG
São Paulo	Western	SP_CASA26	09-12-24	Open	5	House	NG
São Paulo	Western	SP_CASA27	09-12-24	Open	6	Apartment	NG
São Paulo	Western	SP_CASA28	09-12-24	Open	10	Apartment	NG
São Paulo	Southern	SP_CASA29	09-13-24	Open	3	Apartment	NG
São Paulo	Southern	SP_CASA30	09-13-24	Open	10	House	LPG

* For open concept kitchens, plastic was used to enclose and isolate the kitchen area. For closed concept kitchens, sealing was achieved simply by shutting doors and windows. ** MASP: Metropolitan Area of São Paulo.

As shown in the protocol, in all homes, after ventilation, Cycles 1, 2, and 3 consist of 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 γ, which is the air exchange (ACH).

The best adjustment was chosen for each home, which was not necessarily in the same Cycle. Figure S1 exemplifies the exponential adjustment made for Home 06, where the best adjustment was found in Cycle 2.

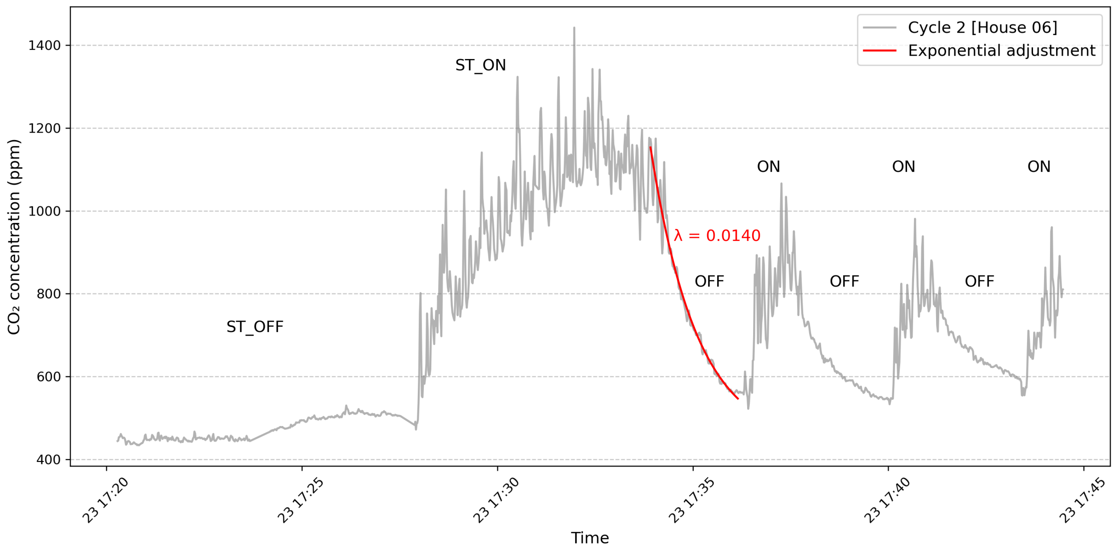

Figure S1. Example of decay curve with House 06 in Cycle 2 using the exponential adjustment.

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 CO_2 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 CO_2 injection, as previously described and illustrated in the figure.

Within this context, periods of decreasing indoor gas concentrations were observed due to air exchange between the kitchen and the outdoor environment. Under such conditions, ventilation influences the indoor mass balance, leading to a reduction in indoor concentrations. When emission rates are inferred from concentration changes under these conditions, the calculation may yield negative values, which do not represent physical emissions but rather reflect limitations of the estimation method.

Negative emission values occurred in a small fraction of the dataset, corresponding to approximately 9 out of 103 calculated emission rates (about 8%). These cases were converted to zero during data processing and did not affect the overall emission patterns discussed in the main manuscript.

S5 Characteristics of Natural Gas and LPG in Brazil

Table S2. Technical characteristics of LPG (referred to in articles 1, 2, 4, 6 and 9 of ANP Resolution Nº 825, of August 2020; ANP (2020))

CHARACTERISTIC	UNIT	PROPANE/BUTANE MIXTURE
Higher Heating Value (4)	MJ/kg	46
Maximum Vapor Pressure at 37.8°C (1)	kPa	1430
Butanes and heavier, max. (2)	% vol.	-
Pentanes and heavier, max. (2)	% vol.	2,0
Propane, min.	% vol.	-
Propene, max.	% vol.	-
Residue, 100 mL evaporated, max.	ml	0,05 (3)
Evaporation Residue, max. (4)	mg/kg	350
Total Sulfur, max.	mg/kg	140

Table S3. Technical characteristics of natural gas (As amended by ANP Resolution Nº. 7/2010; ANP (2010))

CHARACTERISTIC	UNIT	LIMIT (2) (3) North, Central - West, Southeast * and South
	kJ/ m^3	35,000 to 43,000
Higher Heating Value (4)	MJ/kg	45
	kWh/m^3	9.72 to 11.94
Wobbe Index (5)	kJ/m^3	46,500 to 53,500
Methane Number, min. (6)		65
Methane, min.	% mol.	85.0
Ethane, max.	% mol.	12.0
Propane, max..	% mol.	6.0
Butanes and heavier, max.	% mol.	3.0
Oxygen, max. (7)	% mol.	0.5
Inerts (N2+CO2), max.	% mol.	6.0
CO2, max.	% mol.	3.0

* São Paulo - Southeast

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