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Supplement of

Measurement Report: Methane and NO_x emissions from natural gas cooking stoves, the case of Chile and Colombia

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S1. Method for determining emission rates.

As stated in the main manuscript, we utilized a static flux chamber technique to quantify the cookstove emission rate. The fundamental equation is the mass balance of species "i" within the chamber assuming no significant chemical reaction and perfect mixing,

$$V_0 \frac{dC_i}{dt} = E_i - \lambda\, V_0\, (C_i - C_{i,b}) \qquad \text{(Eq S1)}$$

With C_i (mol/m^3) the molar concentration of the gas within the chamber, $C_{i,b}$ (mol/m^3) the background or ambient concentration of gas "i", V_0 is the volume of the kitchen chamber (m^3), and λ (min^{-1}) is the air exchange rate between the confined kitchen space and the surroundings. Determination of the mean emission rate over a given period, $\bar{E}_i$, is therefore a parameter estimation problem. Several approaches can be used to solve the system for the parameter $\bar{E}_i$ when time series data on C_i is available, and the volume V_0 and exchange rate are known.

Method 1. Following Lebel et al. 2022, one possible approach to determine $\bar{E}_i$ is to rearrange Eq. S1. As follows,

$$E_i = V_0 \frac{dC_i}{dt} + \lambda\, V_0\, (C_i - C_{i,b}) \qquad \text{(Eq S2)}$$

Which, by applying the fundamental theory of calculus, can be further written as:

$$E_i = V_0 \frac{d}{dt}\left[C_i + \lambda \int_{t_0}^{t} (C_i - C_{i,b})dt'\right] \qquad \text{(Eq S3)}$$

The quantity within the square brackets involves the concentration at time t, C_i, and a term proportional to the cumulative mass of species "i" that has been exchanged with the surroundings over the period. For this reason, the term in brackets is referred to as the "corrected" concentration, $\hat{C}_i$. This is a useful quantity, because when written in terms of $\hat{C}_i$, Eq. S3 takes the form of the mass balance equation for a perfectly sealed chamber (i.e., one with no air exchange),

$$E_i = V_0 \frac{d\hat{C}_i}{dt} \qquad \text{(Eq S4)}$$

Therefore, if consecutive observations of $\hat{C}_i$ can be made as a function of time, with an interval between observations Δt_k, the emission rate over a period would be proportional to the slope of the data $\{t\,,\hat{C}_{i,t}\}$. Written, in terms of observed values, the corrected concentration is:

$$\hat{C}_{i,t} = C_{i,t} + \lambda \sum_{k=t0} (C_{i,k} - C_{i,b})\Delta t_k \qquad \text{(Eq S5)}$$

Once the time-series of $\hat{C}_{i,t}$ has been built with the observations, a least-square method can be used to statistically determine the slope of the time series, which should yield an estimate of the emission rate. When $\lambda \times \Delta t_k \ll 1$, which is the case for our high-frequency samples (in this work we measured CO_2 and CH_4 concentrations of species at 1Hz, and NOx and CO at 1 minute), the product of the exchange rate and the sampling interval could be written as $\lambda\, \Delta t_k \approx (1 - e^{-\lambda\, \Delta t_k})$. Replacing this expression into Eq. S5 yields the expression reported in Supplementary Material of Lebel et al., 2022.

Method 2. Equivalently, if the average emission rate of species "i" over a period Δt, can be fond simply by time-averaging Eq S2 over the period in question, i.e.,

$$\bar{E}_i = V_0 \frac{\overline{\Delta C_i}}{\Delta t} + \lambda\, V_0\, (\bar{C}_i - C_{i,b}) \qquad \text{(Eq S6)}$$

In this approach, the average emission rate consists of two contributions, the first term, is simply the average accumulation rate of species "i" over the period Δt. In fact, this can be simply calculated as the difference between the concentration at the end and at the beginning of the period and divided by the time interval. The second term accounts for the mass of "i" that is lost to the surroundings and can be calculated as the exchange rate times the difference between the average concentration over the period to the background concentration, which is assumed constant over the sampling period.

Instantaneous release. For instantaneous releases of methane during pulse-on or pulse-off operations, the emission rate can be considered as a delta function: $E_i = n_i \delta(t - t_0)$, with n_i the total number of moles released at time t_0. With that approach, Eq A6 for this case is $n_i = V_0 \Delta C_i$. For this type of measurement, we disregard the correction air exchange correction term, as the timescale of air exchange is much larger than the time-interval considered for the pulse-on/off measurement.

Method 3. Solving Eq S1 for the initial condition $C_i(t = 0) = C_{i,0}$ results in the following closed-form solution for C_i,

$$C_i = C_{i,b} + \frac{E_i}{\lambda V_0} + \left(C_{i,0} - C_{i,b} - \frac{E_i}{\lambda V_0}\right) e^{-\lambda t} \;\; , \;\; \lambda \neq 0 \qquad \text{(Eq S7a)}$$

$$C_i = C_{i,0} + \frac{E_i}{V_0} t \;\; , \;\; \lambda = 0 \qquad \text{(Eq S7b)}$$

Given paired observed data for C_i over time, parameters E_i and $C_{i,0}$ can be estimated using non-linear Ordinary Least Squares (OLS) with known values for $C_{i,b}$ and λ. In OLS, the vector of parameters θ is estimated by minimizing the sum of squared residuals,

$$\theta_{OLS} = \underset{\theta \in \Theta}{\text{argmin}} \sum\nolimits_{j=1}^{n} \left[C_{i,obs_j} - C_{i,sim}(t_j, \theta)\right]^2 \qquad \text{(Eq S8)}$$

where C_{i,obs_j} is the observed concentration of species "i" at time "j", $C_{i,sim}(t_j, \theta)$ is the simulated concentration of species "i" at time "j" using Eq S7, n is the total number of observations, and Θ is the admissible parameter space. In this study we implemented a constrained approach since $E_i \geq 0$ and $C_{i,0} \geq 0$. We used the *least_squares* function from the *optimize* module of the *scipy* library (version 1.15.2) in Python 3.10, with the Trust Region Reflective minimization algorithm.

Parameter uncertainty was quantified using residual bootstrapping. In this method, the vector of residuals $R = C_{i,obs_j} - C_{i,sim}(t_j, \theta)$ obtained when fitting the model of Eq S7 to the observations is resampled with replacement hundreds of times to generate synthetic residual vectors (here we used 1000 replicates). Each resampled vector is added to the fitted model simulations using the original data to generate synthetic observations. Then, model parameters (i.e., E_i and $C_{i,0}$) are estimated using non-linear OLS for each set of synthetic observations. The resulting bootstrap distributions of model parameters provide percentile-based 95% confidence intervals accounting for potential heteroscedasticity and non-normality.

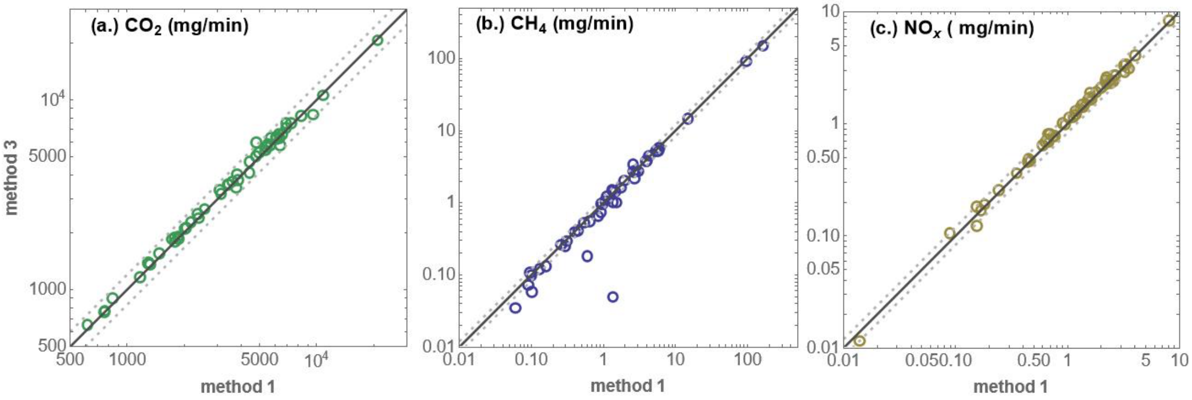

Figure S1. *One-to-one plots comparing the estimated emission rates for the SS-on state for (a.) CO_2 (b.) CH_4, and (c.) NOx. All emission rates are expressed in mg/min. The dashed lines in the plots show the 20% region.*

The analysis demonstrates that the true variability in emission rates, which for all species analyzed span several orders of magnitude, is much larger than both the method accuracy and the relative differences between methods to estimate the emission rates. This analysis shows the method is reliable for estimating individual burner emission rates, with only the sample size limiting the ability to characterize the real distribution of emission rates for the population of stoves analyzed in both cities.

S2. Characteristics of Natural Gas for Chile and Colombia.

The information regarding the composition of the Natural Gas (NG) supplied to the locations sampled was collected from publicly available information. In the case of Colombia, the Energy Planning Agency (UPME), a government agency, keeps and publishes the information for all fuels supplied nationwide, including gas from different basins. The NG composition for Bogotá, which is a mixture from two different basins, is the following:

Table S1. *Physical and chemical characteristics of the natural gas supplied to the city of Bogotá*[1].

Natural gas characteristics (Bogotá)

Components		%	Properties	
CH_4	Methane	82.27%	Density (kg/Nm^3)	0.83
C_2H_6	Ethane	10.23%	#C atoms	1.14
C_3H_8	Propane	1.12%	#H atoms	4.19
C_4H_{10}	Butane	1.67%	Mol weight. (g/mol)	19.55
C_5H_{12}	Pentane	0.24%	LHV (MJ/Nm^3)	37.25
C_6H_{14}	Hexane	0.02%	HHV (MJ/Nm^3)	41.18
CO_2	CO2	3.43%		
N_2	Nitrogen	0.70%		

The natural gas supplied to the city of Santiago de Chile is, in contrast, a little richer in methane, with 90.0% CH_4, 6.38% ethane, 0.22% propane, with the remaining fraction corresponding to higher-carbon content gases and CO_2. The LHV used for the natural gas in Santiago was 43.7 MJ/kg (CNE, 2009).

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[1] Obtained from Table 3 of the Report on Emission factors for Colombian Fuels. A technical report produced by the UPME. Last accessed in July 2025.

S3. Chamber set-up and typical chamber volume.

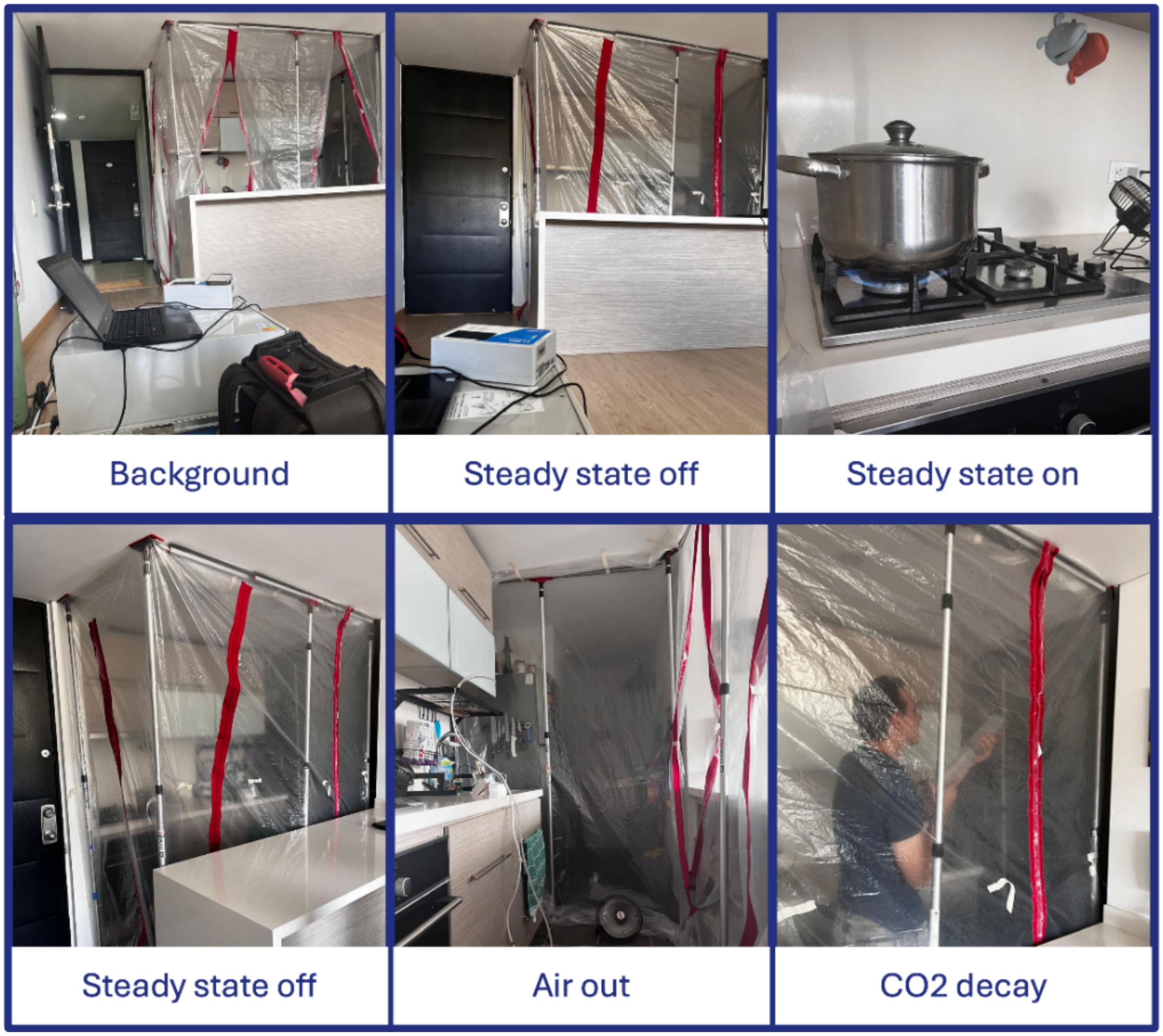

Figure S2*. Example of the enclosing of a kitchen space in Bogotá. In this case, as is common in the city, the kitchen is a relatively small space, but it is open to other rooms in the apartment. In that case, the enclosing must be done entirely using plastic sheets and supports. Fans used to ensure rapid mixing within the chamber are visible in the pictures.*

Given Bogotá's high population density of 24,000 people per km^2, amongst the highest in the world (e.g. Wheeler, 2015), most of the households are relatively small apartments in apartment buildings. The mean enclosed kitchen volume for the houses sampled in Bogotá was 11.2 m^3 (largest volume was 21.2 m^3, and the smallest was 5.65 m^3) and 22.9 m^3 for Santiago. For typical ceiling height of 2.4 m, those volumes correspond to an area of 4.7 m^2 and 9.5 m^2 for Bogotá and Santiago, respectively. Several fans, typically one near the stove and one in another location, (see Figure S2), were used to ensuring rapid mixing within the chamber, aiming to achieve homogeneous gas concentration within the chamber.

S4. Sampling schedule.

Table S2. *Sampling Schedule followed in Chile. Houses were sampled in the capital city of Santiago ("SANT"). The table includes the volume of the enclosed kitchen space, and the species sampled*

ID	DATE (dd/mm/yyyy)	VOLUME (m^3)	SPECIES
SANT_CASA01	13/06/2024	30.94	CH4, CO2, CO
SANT_CASA02	13/06/2024	41.49	CH4, CO2, CO
SANT_CASA03	14/06/2024	12.77	CH4, CO2, CO
SANT_CASA04	14/06/2024	39.36	CH4, CO2, CO
SANT_CASA05	15/06/2024	14.39	CH4, CO2, CO
SANT_CASA06	15/06/2024	28.16	CH4, CO2, CO
SANT_CASA07	16/06/2024	53.91	CH4, CO2, CO
SANT_CASA08	16/06/2024	40.96	CH4, CO2, CO
SANT_CASA09	18/06/2024	12.76	CH4, CO2, CO, NOX, C6H6
SANT_CASA10	18/06/2024	30.38	CH4, CO2, CO, NOX
SANT_CASA11	19/06/2024	22.03	CH4, CO2, CO, NOX, C6H6
SANT_CASA12	19/06/2024	31.52	CH4, CO2, CO, NOX
SANT_CASA13	20/06/2024	37.54	CH4, CO2, CO, NOX, C6H6
SANT_CASA14	20/06/2024	6.05	CH4, CO2, CO, NOX, C6H6
SANT_CASA15	21/06/2024	14.40	CH4, CO2, CO, NOX
SANT_CASA16	21/06/2024	19.41	CH4, CO2, CO, NOX
SANT_CASA17	22/06/2024	11.69	CH4, CO2, CO, NOX, C6H6
SANT_CASA18	22/06/2024	12.41	CH4, CO2, CO, NOX, C6H6
SANT_CASA19	23/06/2024	5.88	CH4, CO2, CO, NOX, C6H6
SANT_CASA20	23/06/2024	19.30	CH4, CO2, CO, NOX, C6H6
SANT_CASA21	24/06/2024	10.73	CH4, CO2, CO, NOX, C6H6
SANT_CASA22	24/06/2024	9.60	CH4, CO2, CO, NOX, C6H6
SANT_CASA23	25/06/2024	11.44	CH4, CO2, CO, NOX, C6H6
SANT_CASA24	24/08/2024	41.06	CH4, CO2, CO, NOX, C6H6
SANT_CASA25	26/08/2024	38.03	CH4, CO2, CO, NOX
SANT_CASA26	26/08/2024	16.63	CH4, CO2, CO, NOX, C6H6
SANT_CASA27	27/08/2024	19.24	CH4, CO2, CO, NOX, C6H6
SANT_CASA28	27/08/2024	14.27	CH4, CO2, CO, NOX, C6H6
SANT_CASA29	28/08/2024	46.37	CH4, CO2, CO, NOX, C6H6
SANT_CASA30	28/08/2024	34.49	CH4, CO2, CO, NOX, C6H6
SANT_CASA31	29/08/2024	31.34	CH4, CO2, CO, NOX, C6H6
SANT_CASA32	29/08/2024	13.66	CH4, CO2, CO, NOX, C6H6
SANT_CASA33	30/08/2024	14.47	CH4, CO2, CO, NOX, C6H6
SANT_CASA34	30/08/2024	16.38	CH4, CO2, CO, NOX, C6H6

Table S3. *Sampling Schedule followed Colombia. All houses were sampled in the capital city of Bogotá. The table includes the volume of the enclosed kitchen space, and the species sampled*

ID	**DATE (dd/mm/yyyy)**	**VOLUME (m3)**	**SPECIES**
M1_Casa4	09/09/2024	13.3	CO2, NOx, CH4
M2_Casa17	09/09/2024	21.2	CO2, NOx, CH4
M3_Casa22	10/09/2024	5.7	CO2, NOx, CH4
M4_Casa33	10/09/2024	11.9	CO2, NOx, CH4
M5_Casa23	11/09/2024	7.8	CO2, NOx, CH4, CO
M6_Casa37	11/09/2024	7.4	CO2, NOx, CH4, CO
M7_Casa13	12/09/2024	9.4	CO2, NOx, CH4, CO
M8_Casa2	12/09/2024	13.3	CO2, NOx, CH4, CO
M9_Casa1	13/09/2024	8.6	CO2, NOx, CH4, CO
M10_Casa27	13/09/2024	10.2	CO2, NOx, CH4, CO
M11_Casa9	14/09/2024	9.6	CO2, NOx, CH4, CO
M12_Casa8	14/09/2024	10.8	CO2, NOx, CH4, CO
M13_Casa6	15/09/2024	11.8	CO2, NOx, CH4, CO
M14_Casa28	16/09/2024	18.6	CO2, NOx, CH4, CO
M15_Casa34	16/09/2024	7.7	CO2, NOx, CH4, CO
M16_Casa26	17/09/2024	11.0	CO2, NOx, CH4, CO
M17_Casa25	17/09/2024	8.3	CO2, NOx, CH4, CO
M18_Casa20	18/09/2024	18.9	CO2, NOx, CH4, CO
M19_Casa40	18/09/2024	8.2	CO2, NOx, CH4, CO
M20_Casa38	19/09/2024	10.6	CO2, NOx, CH4, CO
M21_Casa39	19/09/2024	7.3	CO2, NOx, CH4, CO
M22_Casa41	25/09/2024	10.3	CO2, NOx, CH4, CO
M23_Casa42	25/09/2024	12.1	CO2, NOx, CH4, CO