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Human metabolic emissions of carbon dioxide and methane and their implications for carbon emissions

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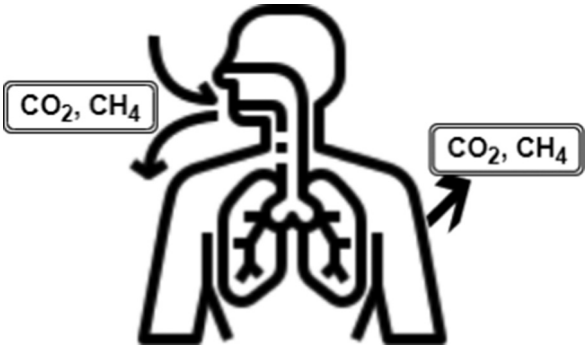
HIGHLIGHTS

- Chamber experiments measured the whole-body, breath and dermal CO_2 and CH_4 emissions from humans.
- CO_2 emissions were strongly influenced by temperature.
- Individual differences influenced CH_4 emissions the most.
- CH_4 producers (third of the subjects) exhaled 10 times more CH_4 than non-CH_4 producers.
- Dermal emissions contributed ~3.5% (CO_2) and ~ 5.5% (CH_4) to the whole-body emissions.

GRAPHICAL ABSTRACT



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ABSTRACT

Carbon dioxide (CO_2) and methane (CH_4) are important greenhouse gases in the atmosphere and have large impacts on Earth's radiative forcing and climate. Their natural and anthropogenic emissions have often been in focus, while the role of human metabolic emissions has received less attention. In this study, exhaled, dermal and whole-body CO_2 and CH_4 emission rates from a total of 20 volunteers were quantified under various controlled environmental conditions in a climate chamber. The whole-body CO_2 emissions increased with temperature. Individual differences were the most important factor for the whole-body CH_4 emissions. Dermal emissions of CO_2 and CH_4 only contributed ~3.5% and ~5.5% to the whole-body emissions, respectively. Breath measurements conducted on 24 volunteers in a companion study identified one third of the volunteers as CH_4 producers (exhaled CH_4 exceeded 1 ppm above ambient level). The exhaled CH_4 emission rate of these CH_4 producers (4.03 ± 0.71 mg/h/person, mean ± one standard deviation) was ten times higher than that of the rest of the volunteers (non-CH_4 producers; 0.41 ± 0.45 mg/h/person). With increasing global population and the expected large reduction in global anthropogenic carbon emissions in the next decades, metabolic emissions of CH_4 (although not CO_2) from humans may play an increasing role in regional and global carbon budgets.

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1. Introduction

Carbon dioxide (CO_2) and methane (CH_4) are among the most important radiatively active trace gases in the Earth's atmosphere. To a large degree, the biosphere governs the emissions of these compounds through the balance between primary production, respiration and biomass burning in the case of CO_2 and via wetlands, freshwaters and domestic animals in the case of CH_4. In addition, there are also important contributions to both budgets from anthropogenic sources such as fossil fuel use. Human metabolism serves as a major source of these compounds in indoor environments (Alberts, 1994), with potential importance also for the local and regional outdoor air and global budgets (Polag and Keppler, 2019). The amount of CO_2 produced by intracellular metabolism is dependent on the amount of proteins, carbohydrates and fats metabolized and the metabolic rate (Azuma et al., 2018; Persily, 1997; Qi et al., 2014; Kuga et al., 2021). CH_4 is largely produced through anaerobic fermentation of carbohydrates and fiber in the large intestine (Cummings, 1983; Sahakian et al., 2010; Bond et al., 1971; de Lacy Costello et al., 2013; Nose et al., 2005; Polag et al., 2014; Roccarina et al., 2010; West et al., 2009). The CO_2 and CH_4 produced in the human body are transported in the blood to the lungs, where they are exhaled into the ambient air. Thus CO_2 and CH_4 are common breath components (Dryahina et al., 2010; Keppler et al., 2016; Levitt et al., 2006; Polag and Keppler, 2018; Polag et al., 2014; Qi et al., 2014; Szabó et al., 2016; Yang et al., 2020).

Humans exhale CO_2 at approximately 4–6% of total breath volume (Mochalski et al., 2015). In the case of CH_4, only a fraction of the population (38 ± 19%; Polag and Keppler, 2019) have been identified as significant CH_4 producers via breath (here termed "CH_4 producers"). It is a generally accepted criterion that a subject is considered to be a CH_4 producer if the CH_4 concentration in exhaled breath exceeds the ambient air level by 1 ppm (Basseri et al., 2012; Dryahina et al., 2010; Polag and Keppler, 2018). Several studies have indicated possible influencing factors for exhaled CO_2 and CH_4 production, such as gastrointestinal diseases (de Lacy Costello et al., 2013; Furnari et al., 2012; Kunkel et al., 2011; Roccarina et al., 2010), sex (Polag et al., 2014; Qi et al., 2014; Stönner et al., 2018; Triantafyllou et al., 2014), ethnic background (Levitt et al., 2006; Mello et al., 2012; Segal et al., 1988), body mass index (BMI) or visceral fat area (VFA) (Basseri et al., 2012; Fernandes et al., 2013; Mathur et al., 2013; Ozato et al., 2020), age (Keppler et al., 2016; Polag et al., 2014; Stönner et al., 2018), and exercise status (Qi et al., 2014; Szabo et al., 2015). CH_4 can be emitted via intestinal gases and both CO_2 and CH_4 can also emanate from the skin through the cutaneous microcirculation (blood capillaries). However, few studies have investigated dermal emissions of CO_2 and CH_4 and they mostly performed small-scale measurements on specific body parts (Frame et al., 1972; Evans and Rutter, 1986; Carlson et al., 1992; Nose et al., 2005).

The global warming potential of CH_4 is much higher than of CO_2, i.e. one ton of atmospheric CH_4 is equivalent to around 86 tons of atmospheric CO_2 over a 20-year period (IPCC, 2014). Although biogenic emissions from vegetation are considered in global budgets along with emissions stemming from human activities, emissions from human beings are usually not considered. With the increasing international efforts to reduce anthropogenic greenhouse gas emissions and to reach net-zero carbon targets in the coming decades, it is intriguing to consider whether human beings with their rapidly increasing global population projected to be 10.9 billion in 2100 (United Nations, 2019) can emerge, especially locally, as more significant sources of atmospheric CO_2 and CH_4 (Polag and Keppler, 2019).

The Indoor Chemical Human Emissions and Reactivity (ICHEAR) study was designed to utilize advanced technologies to examine the impact of exhaled and dermally emitted human bioeffluents on indoor air chemistry under different conditions comprising human and environmental factors impact. In this study, we investigate human whole-body, dermal and exhaled CO_2 and CH_4 emission rates in a controlled environment under various conditions. The analysis of the CO_2 data is an extension to the work by Sakamoto et al. (2022). The data are used to provide an estimate of the innate human contribution of CO_2 and CH_4 to the global atmosphere.

2. Material and methods

2.1. Data collection

In total, 20 volunteers were divided into five groups with each group consisting of four people (two males, two females, except one group with three males, one female). Each group sat in a stainless steel climate chamber (22.5 m^3, ventilated with outdoor air at an air change rate of ~3.2 h^{-1}) under various indoor environmental conditions (temperature (moderate vs. high with set points 25 or 31 °C), relative humidity (RH, low vs. high, set points ~ 25% or 65%), clothing level (long or short set of provided new clothes), ozone level (none or ~ 40 ppb at steady state in the occupied chamber)). Three groups of young adults (A1, A2, A3) had an average age of 25.1 (range 19–30) years and an average BMI of 21.6 (range 20.0–23.9). The average age of the fourth group (teenagers, T4) was 13.8 (range 13–15) years and their average BMI was 19.5 (range 19.1–20.4). The fifth group (seniors, S5) had an average age of 70.5 (range 68–72) years and an average BMI of 25.6 (range 22.5–28.1).

Because the ICHEAR study originally focused on the impact of human emissions on subsequent indoor air chemistry and chemical reactivity, restrictions on the selection of volunteers and their behavior applied. All volunteers were Caucasian (from Denmark, Greece, Italy, Slovakia, Hungary, Spain), non-smokers, and did not suffer from asthma, allergies, or any chronic disease. They were instructed not to drink alcohol, eat spicy food or garlic or significantly alter their diet throughout the experimental period. Eating or chewing gum was not allowed during the measurements. All participants showered the night before each experimental day and used only the personal care products provided by the experimenters (fragrance-free liquid soap and shampoo, and toothpaste). Experiments lasted 3 h in the morning. On most experimental days the subjects returned to the chamber for another 2.5 h after a short lunch break (~15 min, a light meal consisting of toast bread, butter, sliced cheese). In a few experiments with one of the groups, the volunteers sat in one chamber wearing breathing masks and exhaled into another identical chamber, isolating dermal and exhaled emissions. They sat either in the chamber where the measurements were made and exhaled into the adjacent chamber (dermal emissions, 2 experiments), or they sat in the adjacent chamber and exhaled into the measured chamber (breath emissions, 1 experiment). The breathing masks (Sperian ValuAir Plus 6100 V series RP155) covered the mouth and nose and were attached to Teleflex medical tubes tightly connected to the other chamber. One-way valves in the mask ensured that the volunteers inhaled air from the chamber where they were seated, and exhaled into the other chamber. A miniature fan mounted at the end of each tube facilitated the movement of all exhaled air through the tube into the second chamber. The measured CO_2 concentrations and tracer gas measurements confirmed the tightness of the breathing arrangement (Bekö et al., 2020).

A cavity ring-down spectrometer (Picarro G2401, time resolution 2–3 s) was connected to the exhaust of the chamber and measured CO_2 and CH_4. The instrumental precision for CO_2 and CH_4 was 0.05 ppm and 1 ppb, respectively. The CO_2 and CH_4 data were calibrated with a standard calibration gas from NOAA (National Oceanic and Atmospheric Administration, USA). Detailed description of the ICHEAR study design and the environmental conditions can be found in Bekö et al., 2020. Detailed analyses of the whole-body CO_2 emissions have been described in Sakamoto et al. (2022). Supplementary data for comparison was obtained from direct measurements of CH_4 in breath of 24 volunteers in a companion project conducted in Porto, Portugal. Details can be found in the Supplementary Material (SM).

2.2. Emission rate estimation

We estimated CO_2 or CH_4 emission rates under steady-state conditions in the chamber. Steady-state condition was defined as the period during which the variation of CO_2 or CH_4 was smaller than 10% (last ~30 min before the volunteers exited the chamber; this was slightly different from our earlier analysis of whole-body CO_2 emissions where the last 15 min were

used; Sakamoto et al., 2022). Under steady-state, occupants were the only source in the chamber, and chamber ventilation and reaction with the hydroxyl radical (OH) (for CH_4 only) were the sinks. Thus, the steady-state emission rates can be calculated as follows:

$$E_{ss} = V \times \left[\left(C_{chamber} - C_{background}\right) \times ACR + C_{chamber} \times k_{OH+CH4} \times OH\right]/N \quad (1)$$

E_{ss} is the estimated steady-state CO_2 or CH_4 emission rates (g/h/person), V is the chamber volume (22.5 m^3), $C_{chamber}$ is the average concentration under steady-state condition (g/m^3, converted from ppm assuming a temperature of 25 °C and 1 standard atmospheric pressure; same for $C_{background}$), $C_{background}$ is the pre-experiment background concentration (g/m^3) for each experiment corrected by a time-dependent diurnal variation correction factor observed on a non-experiment day. ACR (air change rate, h^{-1}) was calculated from the decay of CO_2 after each experiment (Bekö et al. (2020)). k_{OH+CH4} (cm^3/molecules/h) is the reaction rate of CH_4 and OH radical under the steady-state temperature (Atkinson et al., 1997). OH (molecules/cm^3) is the actual OH concentration inferred for each experiment from direct measurements of all individual gas-phase VOCs using a PTR-TOF-MS and from measurements of total OH reactivity (Zannoni et al., 2022). N is the number of volunteers in the chamber.

2.3. Data analyses

The inter-day variation of whole-body CO_2 and CH_4 emission rates were analyzed by comparing individual replicate experiments in the ICHEAR study. For the eight pairs of replicate experiments, average emission rates and their standard deviations were calculated. Due to the small number of experiments with specific experimental conditions, such direct comparison of results was also used to look for associations between whole-body emission rates and temperature, humidity and volunteer group, as well as for comparison of dermal and breath emissions.

The dominance analysis determines the relative importance of one factor among all the given factors (Azen and Budescu, 2003; Budescu, 1993). It has been recently applied to the ICHEAR dataset to determine the dominant factors influencing the overall human-generated OH reactivity (Zannoni et al., 2021). In this study, the dominance analysis is conducted with the Python library "dominance-analysis 1.1.7" (https://pypi.org/project/dominance-analysis/, last access: July 18, 2021). Seven potential influencing factors were input into the analysis to calculate their relative importance to the whole-body CO_2 and CH_4 emission rates. Their relative importance is measured in all possible submodels pair-wise, i.e. 2^7–1 = 127 sub-models in total. Adding one factor into any sub-model causes a change in incremental R^2, which is defined as the additional contribution of this factor. The relative importance of one factor is calculated by dividing the overall average incremental R^2 contribution of this factor by the R^2 of the complete model. The seven factors considered in this study are time of day (morning or afternoon experiments), average air temperature and absolute humidity under the steady-state condition in the chamber, experimental day (to represent inter-day variation), volunteer group, ozone (present or absent), exposed skin surface (long or short clothing).

3. Results

3.1. Inter-day variation of whole-body CO_2 and CH_4 emissions

Eight pairs of replicate whole-body experiments with two groups of young adults (A1, A2) and the group of seniors (S5) were available for this analysis. The diamonds in Fig. 1 represent the averages of two replicate experiments (marked as empty circles) performed under nearly identical conditions – target air temperature and humidity, ozone level, clothing type and time of the day (before or after lunch). These experiments confirm the high reproducibility of the results; the differences between the replicate whole-body CO_2 emission rates were negligible (standard deviations (SD) between 0.05 and 1.47 g/h/person, average SD being 2% of average of the mean replicate emission rates). Larger differences were observed for CH_4 emission rates (SD for replicate measurements between 0.25 and 0.95 mg/h/person, average SD being 23% of average of the mean replicate emission rates), indicating a more pronounced inter-day variation of human CH_4 emissions, which will be discussed in the next section.

3.2. Influencing factors for the whole-body CO_2 and CH_4 emissions

The average CO_2 emission rate across all of the whole-body experiments was 28.7 ± 2.1 g/h/person. For CH_4 it was 2.17 ± 1.14 mg/h/person. Fig. 2 shows the relative importance (%) of seven potential influencing factors for the whole-body CO_2 and CH_4 emission rates (from ICHEAR) determined in the dominance analysis. Time of day (morning or afternoon), air temperature and absolute humidity were the top three relative important factors for whole-body CO_2 emission rate, suggesting an intra-day variation and a possible temperature and humidity dependency of whole-body CO_2 emissions. These factors were followed by the experimental day (indicating inter-day variation) and volunteer group (representing inter-group variation). The whole-body CH_4 emission rate was largely dominated by the volunteer group, indicating that the number of CH_4 producers in each group and individual differences in CH_4 emission have a large impact on the estimated whole-body CH_4 emission rates. Lower relative importance of temperature, absolute humidity and experimental day suggest their weaker but possible impact on whole-body CH_4 emission rates. Intra-day variation of whole-body CH_4 emission was negligible. Consistent with Sakamoto et al. (2022), both ozone and exposed skin surface (long or short clothing) were relatively unimportant for whole-body CO_2 and CH_4 emissions, which can be explained by the facts that CO_2 and CH_4 are unreactive with ozone and their dermal emissions contribute very little to their whole-body emissions (see Section 3.4).

3.3. Effect of temperature, humidity and volunteer group on the whole-body CO_2 and CH_4 emissions

Fig. 3 shows an Arrhenius plot illustrating the effect of air temperature in the chamber on the whole-body CO_2 and CH_4 emissions. The differences between volunteer groups are also indicated. There was a clear correlation between the whole-body CO_2 emission rates and temperature across all data as well as within the volunteer groups. For detailed discussion of the relationship between CO_2 emission rates, temperature and volunteer group, see Sakamoto et al. (2022). No clear relationship between temperature and whole-body CH_4 emissions was observed. The weak relationship was presumably driven by the differences between volunteer groups, as the emission rates did not change with temperature within groups. The whole-body CH_4 emission rates of the senior group (S5) were higher than that of young adults (>60%). CH_4 did not reach steady-state during the experiments with a group of teenagers (T4) and the results are therefore not shown in Fig. 3.

Based on exhaled CH_4 emission rates obtained in a companion study (see SM) and assuming 5% whole-body emission being attributed to dermal emissions (CH_4 from flatus was ignored, see Section 3.4), we estimated the average and range of whole-body CH_4 emission rates (mg/h/person) for a group of four adults: (1) 4 non-CH_4 producers:1.73 (0.42, 6.89); (2) 3 non-CH_4 producers and 1 CH_4 producer: 5.54 (3.27, 10.89); (3) 2 non-CH_4 producers and 2 CH_4 producers: 9.34 (6.11, 13.89); (4) 1 non-CH_4 producer and 3 CH_4 producers: 13.15 (8.95, 17.38); (5) 4 CH_4 producers: 16.96 (11.80, 20.88). The measured average whole-body CH_4 emission rate for group A1 was 1.23 (0.87–1.75) mg/h/person, for group A2 it was 2.24 (1.04–3.27) mg/h/person, for group A3 it was 1.67 mg/h/person (2 measurements: 1.64 and 1.69), and for group S5 it was 4.37 (3.91–4.65) mg/h/person. Thus, groups A1, A2 and A3 likely consisted of four non-CH_4 producers and group S5 may have included one CH_4 producer.

Results from two pairs of experiments with low and high humidity provide insights into the effect of absolute humidity (Table 1). In pair 1 (Table 1), the temperature was high, the volunteers wore long clothing and there was ozone in the chamber in the afternoon, but not in the morning. On day 1 with low humidity, the whole-body CO_2 emission rates were

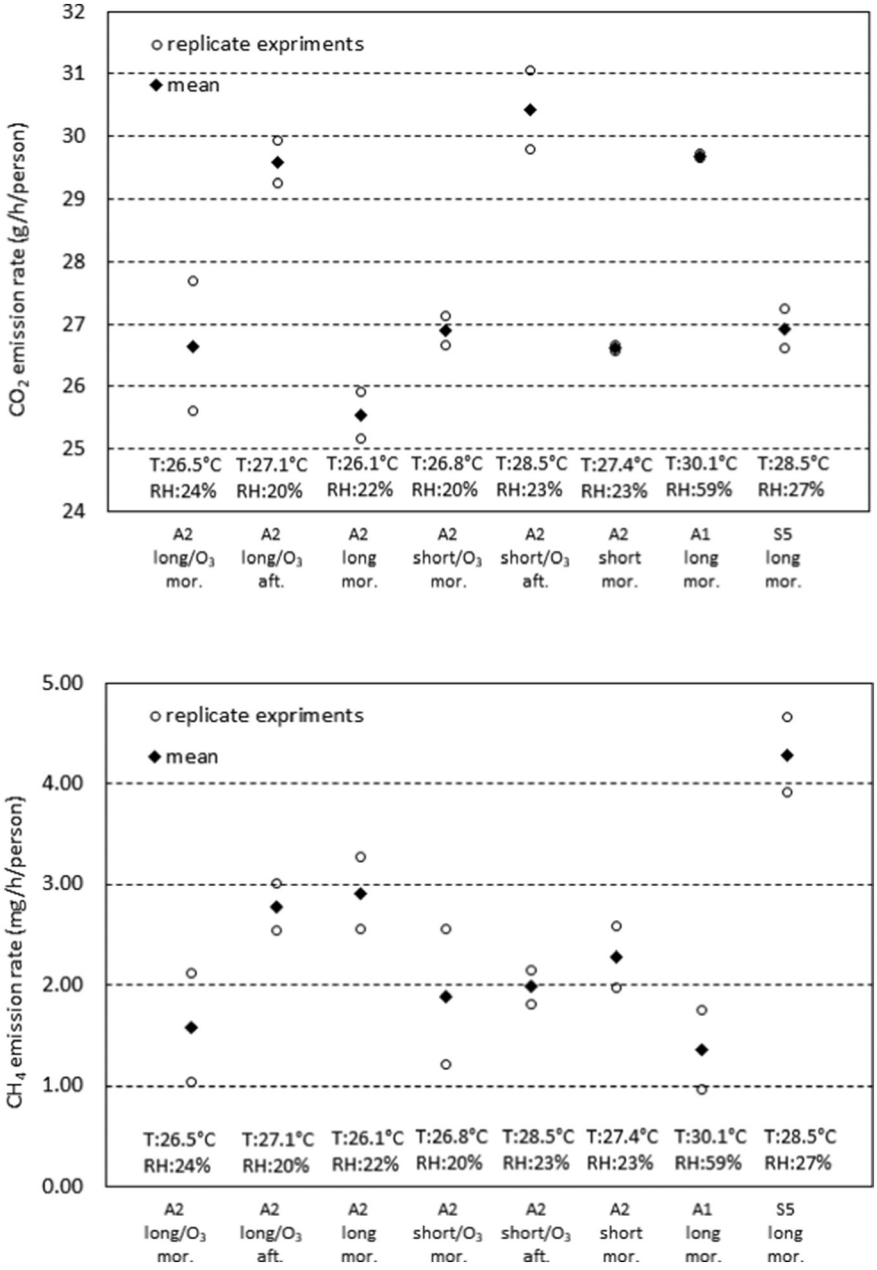

Fig. 1. Results of two replicate experiments (empty circles, n = 1 for each replicate) and their mean (solid diamonds, average values of the two replicate experiments) for whole-body CO_2 and CH_4 emission rates under different indoor environmental conditions (mor: morning, aft: afternoon; long: long clothing, short: short clothing; O_3: ozone present in the chamber). The difference between steady-state air temperatures was within 1 °C for replicate experiments. The average steady-state temperature and relative humidity for the two replicate experiments are shown.

30.2 g/h/person (morning, ~32.5 °C, RH 32%, absolute humidity 11.1 g/m^3) and 30.5 g/h/person (afternoon, ~31.8 °C, RH 30%, absolute humidity 10.1 g/m^3). On day 2 with higher humidity, the whole-body CO_2 emission rates were 31.1 (morning, ~32.6 °C, RH 62%, absolute humidity 21.7 g/m^3) and 32.1 g/h/person (afternoon, ~32.3 °C, 63%, absolute humidity 21.7 g/m^3). In pair 2 (Table 1), the temperature was moderate, the volunteers wore long clothing and there was no ozone in the chamber. On day 3 with low humidity, the whole-body CO_2 emission rate was 28.5 g/h/person (morning, ~29.3 °C, RH 33%, absolute humidity 9.7 g/m^3). On day 4 with high humidity, the whole-body CO_2 emission rate was 29.7 g/h/person (morning, ~29.4 °C, RH 62%, absolute humidity 18.3 g/m^3). A replicate experiment to the latter (day 5) exhibited identical results, a CO_2 emission rate of 29.7 g/h/person (~30.9 °C, RH 56%, absolute humidity 17.9 g/m^3). The differences between low and high humidity conditions were small and within the differences between replicate conditions (Section 3.1; SD between 0.64 and 1.13 g/h/person, average SD being 2.8% of average of the mean emission rates for low and high humidity). Additionally, the 3–5% increase in the whole-body CO_2 emission rate under higher humidity is within the 0–8% difference between replicate experiments. This is in line with the fact that we did not find a clear relationship between CO_2 emission rates and relative humidity (Sakamoto et al., 2022).

The corresponding whole-body CH_4 emission rates were 1.15 (morning) and 1.21 mg/h/person (afternoon) under low humidity (day 1, Table 1), and 0.87 and 1.00 mg/h/person, respectively, under higher humidity (day 2) in the first pair of experiments. They were 1.26 mg/h/person under low humidity (day 3) and 0.97 mg/h/person (day 4) under high humidity in the second pair of experiments. The replicate experiment (day 5) to the latter exhibited an 80% higher emission rate of 1.75 mg/h/person. For CH_4, the differences between low and high humidity conditions were larger, but within the differences between replicate conditions

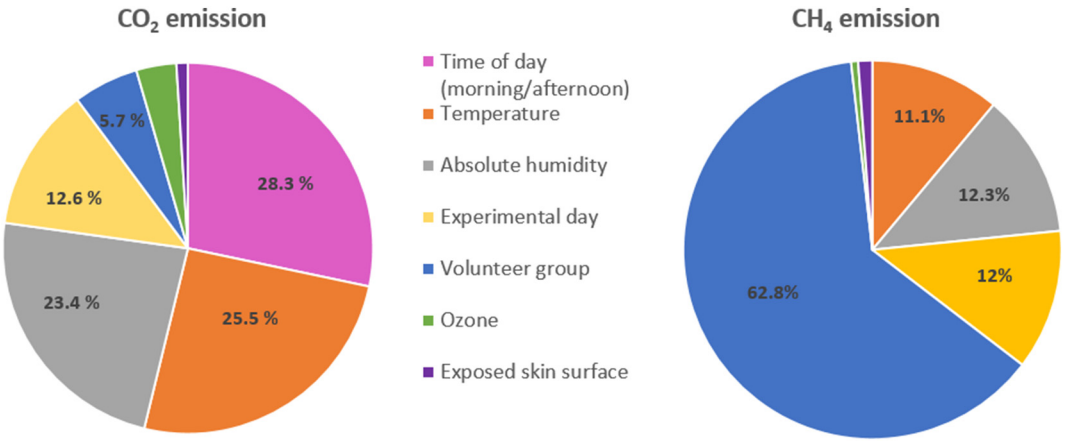

Fig. 2. Relative importance of seven factors for the whole-body CO_2 and CH_4 emission rates.

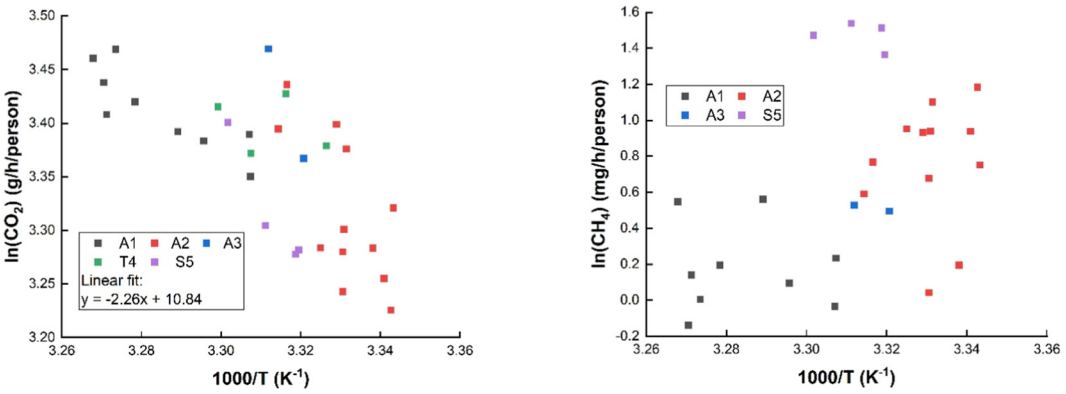

Fig. 3. Correlation of the natural logarithm of the whole-body CO_2 or CH_4 emission rates versus (1000/T). Left figure adapted from Sakamoto et al. (2022).

(Section 3.1; SD between 0.15 and 0.21 mg/h/person, average SD being 17% of average of the mean emission rates for low and high humidity). The 21–32% higher whole-body CH_4 emission rate under low humidity is within the 18–110% difference between replicate experiments. These results indicate that the effect of humidity on CH_4 emission rates is negligible and smaller than the rather substantial inter-day variations (Polag and Keppler, 2018).

3.4. Dermal vs. exhaled CO_2 and CH_4 emissions

Dermal-only (2 experimental days) and breath-only (2 experimental days) emission measurements were conducted along with whole-body measurements (1 experimental day) with one group of four young adults (group A3). For CH_4, the dermal-only emission rate on May 2 (morning, ~31 °C, ~70% RH, short clothing) was 0.09 mg/h/person, on May 7 it was 0.05 mg/h/person in the morning (~29 °C, ~28% RH, short clothing) and 0.13 mg/h/person in the afternoon (~30 °C, ~28% RH, short clothing). The breath-only emission rate on May 3 (morning, ~32 °C, ~60% RH, short clothing) was 1.65 mg/h/person, on May 6 it was 1.65 mg/h/person in the morning (~26 °C, ~28% RH, long clothing) and 1.86 mg/h/person in the afternoon (~26 °C, ~31% RH, long clothing). The differences between conditions may have been caused by differences in the subjects' thermal environment, time of the day (before or after lunch) and inter- and intra-day variability. The whole-body (dermal and breath) emission rates on May 8 were 1.64 mg/h/person in the morning (~29 °C, ~28% RH, long clothing) and 1.69 mg/h/person in the afternoon (~29 °C, ~28% RH, long clothing). The average estimated exhaled CH_4 emission rate was thus about 19 (max. range 13–37) times higher than the average dermal emission rate. CH_4 is primarily emitted via breath. The slightly higher breath-only emission rate observed in the afternoon of May 6 compared with the whole-body experiment under similar conditions was likely caused by the inter-day variation of human CH_4 emissions. It appears that the generally significant contribution of flatus to human CH_4 emissions was negligible in the current experiments.

Table 1
Effect of humidity on whole-body CO_2 and CH_4 emission rates.

	Day	Time of day	Relative humidity (%)	Absolute humidity (g/m^3)	Temperature (°C)	Whole-body CO_2 emission rate (g/h/person)	Whole-body CH_4 emission rate (mg/h/person)
Pair 1	day 1	Morning	Low (32)	11.1	32.5	30.2	1.15
	day 1	Afternoon	Low (30)	10.1	31.8	30.5	1.21
	day 2	Morning	High (62)	21.7	32.6	31.1	0.87
	day 2	Afternoon	High (63)	21.7	32.3	32.1	1.00
Pair 2	day 3	Morning	Low (33)	9.7	29.3	28.5	1.26
	day 4	Morning	High (62)	18.3	29.4	29.7	0.97
	day 5	Morning	High (56)	17.9	30.9	29.7	1.75

For CO_2, the dermal-only emission rates were 1.34 g/h/person (May 2, morning), 0.81 g/h/person (May 7, morning) and 1.16 g/h/person (May 7, afternoon). The breath-only emission rates were 25.5 g/h/person (May 3, morning), 28.2 g/h/person (May 6, morning) and 29.9 g/h/person (May 6, afternoon). The corresponding whole-body emission rates were 29.0 g/h/person (May 8, morning) and 32.1 g/h/person (May 8, afternoon). The average estimated exhaled CO_2 emission rate was therefore about 25 (max. Range 19–37) times higher than the average dermal emission rate. The average dermal emission rates ($n = 3$) of CO_2 and CH_4 constituted a minor contribution (3.5% and 5.5%, respectively) to the average whole-body emission rates across all experiments ($n = 27$). Dermal emission can thus be considered of minor importance, given that it is similar to or lower than the variability of replicate whole-body emission rates. For comparison, the average breath-only emission rates (n = 3) were 96.5% (CO_2) and 80% (CH_4) of the average whole-body emission rates across all experiments (n = 27). The value for CO_2 matches well the remaining contribution from dermal emissions (3.5%) and reflects the airtightness of the breathing system in our chamber. The value for CH_4 does not add up to 100% with the contribution from dermal emission and this may reflect the larger variability in CH_4 emission rates, the relatively few breath-only and dermal-only experiments and the lack of quantification of CH_4 emission from flatus.

4. Discussion

The measured whole-body CO_2 emission rates from this study match well with earlier studies. A detailed comparison to the literature can be found in Sakamoto et al. (2022). The average measured whole-body CH_4 emission rate (2.17 mg/h/person) was somewhat higher than the global average emission of 1.54 mg/h/person estimated by a human CH_4 emission study (Polag and Keppler, 2019). Significantly higher emission rates were obtained for the senior group, suggesting that this group consisted of more CH_4 producers than the younger groups. Indeed, earlier studies suggest that the fraction of CH_4 producers within a population increases with age (Keppler et al., 2016; Polag et al., 2014).

The whole-body emission rates of CO_2 and CH_4 were overwhelmingly dominated by breath. Dermal emissions as well as the differences between breath emissions and whole-body emissions were smaller than the variability in breath and whole-body emissions across these experiments. The absolute contributions of breath and dermal emissions to the whole-body CO_2 and CH_4 emission rates should be interpreted with caution, considering the numerous factors influencing both, the accuracy of the measurements and the small number of experiments with isolated breath or dermal emissions. The exhaled emission rates of CH_4 were comparable with the ones obtained in a companion study of breath emissions directly measured using the same instrumentation in Porto, Portugal (1.7 vs. 1.6 mg/h/person in ICHEAR and Porto, respectively; see SM). The difference is negligible especially considering that CH_4 producers were found to exhale about three times more than these average values, while non-CH_4 producers were found to emit about one fourth of these average emission rates. These average emission rates reflect both individual differences and proportions of CH_4 producers in the two studies. One-third of the volunteers in the Porto breath study were identified as CH_4 producers. This corresponds to the global average of 38% estimated from numerous studies summarized by Polag and Keppler (2019). The results of the Porto study, however, should be interpreted with caution, because tidal volume was not measured directly in the study, and our instrument had a low resolution (2–3 s); it was not fast enough to continuously measure each breath profile.

More importantly, even though both studies were performed with Caucasian volunteers of similar age, the very different experimental designs and different personal and environmental conditions (e.g. diet, metabolic rate, body mass, room temperature, health conditions) make direct comparisons of the two studies difficult. For example, long-term dietary patterns influence the human gut microbiome (David et al., 2014; Wu et al., 2011), and thus possibly CH_4 production. Even vaccinations and short-term dietary changes can change humans from high to low CH_4 emitters and vice versa (Polag and Keppler, 2018). We did not observe statistically significant differences in the exhaled CH_4 emissions between females and males, which is consistent with the findings of Stönner et al. (2018) and Levitt et al. (2006). It should be noted that the breath-only emission rates in the ICHEAR study are not expected to be influenced by losses (e.g. absorption) in the 22 mm diameter vinyl Teleflex medical tubes (< 2 m long) connecting the facemasks with the adjacent chamber (Bekö et al., 2020; Li et al., 2020).

A broad range of CO_2 emission rates from skin has been reported. Frame et al. (1972) summarized earlier results, which range from 11×10^{-5} to 370×10^{-5} ml/cm^2/min for different body parts, with most values being under 35 ml/cm^2/min (except for axillae and forehead). The study also measured a CO_2 emission rate of 3.4×10^{-5} ml/cm^2/min for an arm and forearm, 4.6×10^{-5} ml/cm^2/min for the hand only, and 1.8×10^{-5} ml/cm^2/min for the forearm only. Emission rates increased after exercise and after wetting the skin. Ernstene and Volk (1932a) reported a range for cutaneous CO_2 emission from arm for 38 subjects aged 15 to 75 between 9.7 and 28.2 ml/cm^2/min. Evans and Rutter (1986) found an emission rate of 6.75×10^{-5} ml/cm^2/min for newborns (on abdomen) and 14.4×10^{-5} ml/cm^2/min for adults (forearm), although lower values were found by Cunico et al. (1977) (2.31 vs 2.18×10^{-5} ml/cm^2/min for adults and newborns, respectively). In a study by Carlson et al. (1992), hands continuously emitted under laboratory conditions 1.0–1.8 ml/h CO_2. Considering a total body surface area of 1.8 m^2 and that the hands constitute about 5% of the total body surface area (US EPA, 2011), this corresponds to an emission rate of $1.85–3.33 \times 10^{-5}$ ml/cm^2/min. The literature indicates large individual differences, but no relationship of dermal emissions with age and sex. Our average dermal emission rate of CO_2 (1.1 g/h/person) corresponds to 57.2×10^{-5} ml/cm^2/min (assuming a body surface area of 1.8 m^2 and density of CO_2 at 25 °C of 1.78 kg/m^3). The average rate of dermal CO_2 emission was about 3.5% of the average total CO_2 emission rate, which is slightly higher than the 1–3% reported in the literature (Shaw et al., 1929; Ernstene and Volk, 1932a; Fitzgerald, 1957; Frame et al., 1972). This is reasonable given that our study is the only one we could identify, which measured the dermal emission of CO_2 during whole-body chamber exposure. Earlier studies were performed on unclothed body parts, while our subjects were partially clothed. Moreover, dermal emissions have been shown to be affected by the behavior of the volunteers (movement, talking, laughing; Carlson et al., 1992) and by temperature, humidity and skin health, as these can impact skin permeability and cutaneous blood flow (Shaw and Messer, 1930; Ernstene and Volk, 1932a, 1932b; Frame et al., 1972). Finally, the breathing arrangement in the ICHEAR study was confirmed to be airtight; we do not expect meaningful contribution of exhaled air leakage to the measured dermal emissions.

Dermal emission of CH_4 has received much less attention in previous studies. Nose et al. (2005) measured CH_4 emissions from the hands of 10 subjects aged 21 to 59 years. The average emission rate was 5 ± 2.1 pg/cm^2/min (range 2.3–7.8). Assuming a body surface area of 1.8 m^2, our average dermal emission rate of CH_4 (0.09 mg/h/person) corresponds to 83.3 pg/cm^2/min, one order of magnitude higher. It may suggest that other parts of the human body than the arms may emit significantly more CH_4. However, numerous other factors outlined above, including the fundamentally different experimental approach may explain the difference. The dermal emission of CH_4 warrants further investigation.

As the whole-body emission rates above were derived from Caucasian volunteers in Denmark, we estimate the total metabolic CO_2 and CH_4 emissions for the Danish Caucasian population using a simple calculation based on the obtained emission rates. We assumed a population of 5.1 million (Caucasians in Denmark; The World Factbook at cia.gov) emitting CO_2 at a whole-body CO_2 emission rate of 28.7 g/h/person (range 25.16–32.11). The whole-body CH_4 emission rate for non-CH_4 producers was assumed to be 1.31 mg/h/person (range 0.87–1.75) based on groups A1 and A3 in the ICHEAR study. A ten times higher rate was used for CH_4 producers. The ratio between non-CH_4 producers and CH_4 producers was set to be 2:1. Lower breath emissions during sleep-time and higher during other than sedentary activity levels were not considered (Fan et al., 2021). The CO_2 emission from the Danish Caucasian population was estimated to be

1.29 Tg/year (range 1.12–1.43), and the CH_4 emission was estimated to be 0.23 Gg/year (range 0.15–0.30).

Applying the same per-person emission rates to the global population, the estimated global CO_2 and CH_4 emissions from human metabolism would be 2.00 Gt/year for CO_2 and 332 Gg/year for CH_4. Despite the limited representativeness of this estimate caused by the specific experimental requirements and conditions, the latter is similar to the global human CH_4 emission of 344 Gg/year estimated by Polag and Keppler (2019), which considered the factors affecting the number of CH_4 producers (ethnicity, age, sex). Additional underlying factors and changes in their magnitude or distribution over time would need to be considered for a more accurate estimate of global human metabolic emissions. These include indoor and outdoor environmental parameters, health conditions (e.g. gastrointestinal diseases strongly related to CH_4 emissions), dietary differences, physical status, diurnal changes in emissions (e.g. lower breath emissions during sleep) and population structure (age, sex, ethnicity, geographical residence, socioeconomics).

Although human emissions contribute little to current global CH_4 emissions, these emissions and their changes in the future may be important at a local scale, such as in megacities, where natural and agricultural sources are minimal. On the other hand, the net effect of human metabolic carbon emissions on the atmospheric CO_2 budget is negligible. In a fast-acting loop, CO_2 is transferred from the atmosphere first through photosynthesis into plant matter, then to human diet (via plants directly or animals fed with plants) and eventually returned to the atmosphere as CO_2. In contrast to land-use change and fossil fuel use, which discharge carbon with extensive storage time, the effect of the annual photosynthesis/respiration cycle occurring between the atmosphere and the terrestrial biosphere (of which humans are a small part) has little net effect on the longer-term accumulation of atmospheric CO_2. However, the impact on the spatial distribution of carbon dioxide uptake and release across regions and continents may be significant (West et al., 2009).

5. Conclusions

We quantified the exhaled, dermal and whole-body CO_2 and CH_4 emission rates in a controlled experimental study. In the whole-body chamber experiments, CO_2 emissions were relatively consistent across similar experimental conditions, while CH_4 emissions exhibited larger variation. CO_2 emissions were strongly influenced by air temperature. CH_4 emissions reflected large individual differences. Dermal emissions of CO_2 and CH_4 were relatively small compared with exhaled emissions; they contributed ~3.5% and ~ 5.5% to the whole-body emission rates, respectively. In direct exhaled breath measurements, one-third of the volunteers were identified as CH_4 producers. The exhaled CH_4 emission rate of CH_4 producers was ten times higher than that of non-CH_4 producers. No difference in exhaled CH_4 emissions was found between males and females.

Author contributions

J.W., G.B., P.W., and P.N. developed the studies. M.L. measured and analyzed CO_2 and CH_4 data. M.L. and J.W. developed the idea of this manuscript. M.L., G.B., and J.W. wrote the manuscript. N.Z. calculated OH concentrations. G.B., N.Z., G.P., N·C, C.M., P.V., P.W., P.N., N.W., and L.E. helped with experimental setup. M.C. and G.B. provided information on volunteers. All authors revised the manuscript.

Declaration of competing interest

Authors declare that they have no competing interests.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.155241.

References

Alberts, W.M., 1994. Indoor air pollution: NO, NO2, CO, and CO2. J. Allergy Clin. Immunol. 94, 289–295.
Atkinson, R., Baulch, D., Cox, R., Hampson Jr., R., Kerr, J., Rossi, M., et al., 1997. Evaluated kinetic, photochemical and heterogeneous data for atmospheric chemistry: supplement V. IUPAC Subcommittee on gas kinetic data evaluation for atmospheric chemistry. J. Phys. Chem. Ref. Data 26, 521–1011.
Azen, R., Budescu, D.V., 2003. The dominance analysis approach for comparing predictors in multiple regression. Psychol. Methods 8, 129.
Azuma, K., Kagi, N., Yanagi, U., Osawa, H., 2018. Effects of low-level inhalation exposure to carbon dioxide in indoor environments: a short review on human health and psychomotor performance. Environ. Int. 121, 51–56.
Basseri, R.J., Basseri, B., Pimentel, M., Chong, K., Youdim, A., Low, K., et al., 2012. Intestinal methane production in obese individuals is associated with a higher body mass index. Gastroenterol. Hepatol. 8, 22.
Bekö, G., Wargocki, P., Wang, N., Li, M., Weschler, C.J., Morrison, G., et al., 2020. The indoor chemical human emissions and reactivity project (ICHEAR): overview of experimental methodology and preliminary results. Indoor Air 30, 1213–1228.
Bond Jr., J.H., Engel, R.R., Levitt, M.D., 1971. Factors influencing pulmonary methane excretion in man. An indirect method of studying the in situ metabolism of the methane-producing colonic bacteria. J. Exp. Med. 133, 572–588.
Budescu, D.V., 1993. Dominance analysis: a new approach to the problem of relative importance of predictors in multiple regression. Psychol. Bull. 114, 542.
Carlson, D.A., Schreck, C.E., Brenner, R.J., 1992. Carbon dioxide released from human skin: effect of temperature and insect repellents. J. Med. Entomol. 29 (2), 165–170.
Cummings, J., 1983. Fermentation in the human large intestine: evidence and implications for health. Lancet 321, 1206–1209.
Cunico, R.L., Maibach, H.I., Khan, H., Bloom, E., 1977. Skin barrier properties in the newborn: transepidermal water loss and carbon dioxide emission rates. Biol. Neonate 32, 177–182.
David, L.A., Maurice, C.F., Carmody, R.N., Gootenberg, D.B., Button, J.E., Wolfe, B.E., et al., 2014. Diet rapidly and reproducibly alters the human gut microbiome. Nature 505, 559–563.
de Lacy Costello, B.P., Ledochowski, M., Ratcliffe, N.M., 2013. The importance of methane breath testing: a review. J. Breath Res. 7, 024001.
Dryahina, K., Smith, D., Spanel, P., 2010. Quantification of methane in humid air and exhaled breath using selected ion flow tube mass spectrometry. Rapid Commun. Mass Spectrom. 24, 1296–1304.
Ernstene, A.C., Volk, M.C., 1932a. Cutaneous respiration in man. IV. The rate of carbon dioxide elimination and oxygen absorption in normal subjects. J. Clin. Invest. 11, 363–376.
Ernstene, A.C., Volk, M.C., 1932b. Cutaneous respiration in man. V. The rate of carbon dioxide elimination and oxygen absorption in subjects with diseases of the skin. J. Clin. Invest. 11, 377–382.
Evans, N.J., Rutter, N., 1986. Percutaneous respiration in the newborn infant. J. Pediatr. 108 (2), 282–286.
Fan, X., Sakamoto, M., Shao, H., Kuga, K., Ito, K., Lan, L., et al., 2021. Emission rate of carbon dioxide while sleeping. Indoor Air 31, 2142–2157.
Fernandes, J., Wang, A., Su, W., Rozenbloom, S.R., Taibi, A., Comelli, E.M., et al., 2013. Age, dietary fiber, breath methane, and fecal short chain fatty acids are interrelated in archaea-positive humans. J. Nutr. 143, 1269–1275.
Fitzgerald, L.E., 1957. Cutaneous respiration in man. Physiol. Rev. 37, 325–336.
Frame, G.W., Strauss, W.G., Maibach, H.I., 1972. Carbon dioxide emission of the human arm and hand. J. Investig. Dermatol. 59, 155–158.
Furnari, M., Savarino, E., Bruzzone, L., Moscatelli, A., Gemignani, L., Gianini, E.G., et al., 2012. Reassessment of the role of methane production between irritable bowel syndrome and functional constipation. J. Gastrointest. Liver Dis. 21.
IPCC, 2014. Mitigation of climate change. Contribution of Working Group III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, p. 1454.
Keppler, F., Schiller, A., Ehehalt, R., Greule, M., Hartmann, J., Polag, D., 2016. Stable isotope and high precision concentration measurements confirm that all humans produce and exhale methane. J. Breath Res. 10, 016003.
Kuga, K., Ito, K., Wargocki, P., 2021. The effects of warmth and CO2 concentration, with and without bioeffluents, on the emission of CO2 by occupants and physiological responses. Indoor Air 31, 2176–2187.
Kunkel, D., Basseri, R.J., Makhani, M.D., Chong, K., Chang, C., Pimentel, M., 2011. Methane on breath testing is associated with constipation: a systematic review and meta-analysis. Dig. Dis. Sci. 56, 1612–1618.
Levitt, M.D., Furne, J.K., Kuskowski, M., Ruddy, J., 2006. Stability of human methanogenic Flora over 35 years and a review of insights obtained from breath methane measurements. Clin. Gastroenterol. Hepatol. 4, 123–129.

Li, M., Weschler, C.J., Bekö, G., Wargocki, P., Lucic, G., Williams, J., 2020. Human ammonia emission rates under various indoor environmental conditions. Environ. Sci. Technol. 54, 5419–5428.

Mathur, R., Amichai, M., Chua, K., Mirocha, J., Barlow, G., Pimentel, M., 2013. Methane and hydrogen positivity on breath test is associated with greater body mass index and body fat. J. Clin. Endocrinol. Metab. 98, E698–E702.

Mello, C.S., Tahan, S., LCF, Melli, do Carmo Rodrigues, M.S., de Mello, R.M.P., ICA, Scaletsky, et al., 2012. Methane production and small intestinal bacterial overgrowth in children living in a slum. World J. Gastroenterol. 18, 5932.

Mochalski, P., Unterkofler, K., Teschl, G., Amann, A., 2015. Potential of volatile organic compounds as markers of entrapped humans for use in urban search-and-rescue operations. Trends Analyt. Chem. 68, 88–106.

Nose, K., Nunome, Y., Kondo, T., Araki, S., Tsuda, T., 2005. Identification of gas emanated from human skin: methane, ethylene, and ethane. Anal. Sci. 21, 625–628.

Ozato, N., Saito, S., Yamaguchi, T., Katashima, M., Tokuda, I., Sawada, K., et al., 2020. Association between breath methane concentration and visceral fat area: a population-based cross-sectional study. J. Breath Res. 14, 026008.

Persily, A.K., 1997. Evaluating building IAQ and ventilation with indoor carbon dioxide. ASHRAE Trans. 103 (2), 193–204.

Polag, D., Keppler, F., 2018. Long-term monitoring of breath methane. Sci. Total Environ. 624, 69–77.

Polag, D., Keppler, F., 2019. Global methane emissions from the human body: past, present and future. Atmos. Environ. 214, 116823.

Polag, D., Leiss, O., Keppler, F., 2014. Age dependent breath methane in the German population. Sci. Total Environ. 481, 582–587.

Qi, M., Li, X., Weschler, L., Sundell, J., 2014. CO2 generation rate in Chinese people. Indoor Air 24, 559–566.

Roccarina, D., Lauritano, E.C., Gabrielli, M., Franceschi, F., Ojetti, V., Gasbarrini, A., 2010. The role of methane in intestinal diseases. Am. J. Gastroenterol. 105, 1250–1256.

Sahakian, A.B., Jee, S.R., Pimentel, M., 2010. Methane and the gastrointestinal tract. Dig. Dis. Sci. 55, 2135–2143.

Sakamoto, M., Li, M., Kuga, K., Ito, K., Bekö, G., Williams, J., et al., 2022. CO2 emission rates from sedentary subjects under controlled laboratory conditions. Build. Environ. 108735.

Segal, I., Walker, A., Lord, S., Cummings, J., 1988. Breath methane and large bowel cancer risk in contrasting African populations. Gut 29, 608–613.

Shaw, L.A., Messer, A.C., 1930. Cutaneous respiration in man. II. The effect of temperature and of relative humidity upon the rate of carbon dioxide elimination and oxygen absorption. AmerJ. Physiol. 95, 13–19.

Shaw, L.A., Messer, A.C., Weiss, S., 1929. Cutaneous respiration in man. I. Factors affecting the rate of carbon dioxide elimination and oxygen absorption. Am.J. Physiol. 90, 107–118.

Stönner, C., Edtbauer, A., Williams, J., 2018. Real-world volatile organic compound emission rates from seated adults and children for use in indoor air studies. Indoor Air 28, 164–172.

Szabo, A., Ruzsanyi, V., Unterkofler, K., Mohacsi, A., Tuboly, E., Boros, M., et al., 2015. Exhaled methane concentration profiles during exercise on an ergometer. J. Breath Res. 9, 016009.

Szabó, A., Unterkofler, K., Mochalski, P., Jandacka, M., Ruzsanyi, V., Szabó, G., et al., 2016. Modeling of breath methane concentration profiles during exercise on an ergometer. J. Breath Res. 10, 017105.

Triantafyllou, K., Chang, C., Pimentel, M., 2014. Methanogens, methane and gastrointestinal motility. J. Neurogastroenterol. Motil. 20, 31–40.

United Nations, 2019. World Population Prospects, p. 2019.

US EPA, 2011. Exposure Factors Handbook. 2011 edn. US Environmental Protection Agency, Washington, DC, USA.

West, T.O., Marland, G., Singh, N., Bhaduri, B.L., Roddy, A.B., 2009. The human carbon budget: an estimate of the spatial distribution of metabolic carbon consumption and release in the United States. Biogeochemistry 94, 29–41.

Wu, G.D., Chen, J., Hoffmann, C., Bittinger, K., Chen, Y.-Y., Keilbaugh, S.A., et al., 2011. Linking long-term dietary patterns with gut microbial enterotypes. Science 334, 105–108.

Yang, L., Wang, X., Li, M., Zhou, X., Liu, S., Zhang, H., et al., 2020. Carbon dioxide generation rates of different age and gender under various activity levels. Build. Environ. 186, 107317.

Zannoni, N., Li, M., Wang, N., Ernle, L., Bekö, G., Wargocki, P., et al., 2021. Effect of ozone, clothing, temperature, and humidity on the total OH reactivity emitted from humans. Environ. Sci. Technol. 55, 13614–13624.

Zannoni, N., Lakey, P.S.J., Won, Y., Shiraiwa, M., Rim, D., Weschler, C.J., et al., 2022. The human oxidation field. Science, in review.