

Performance of low-cost monitors to assess household air pollution

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ABSTRACT

Exposure to household air pollution is a leading cause of morbidity and mortality globally. However, due to the lack of validated low-cost monitors with long-lasting batteries in indoor environments, most epidemiologic studies use self-reported data or short-term household air pollution assessments as proxies of long-term exposure. We evaluated the performance of three low-cost monitors measuring fine particulate matter (PM_{2.5}) and carbon monoxide (CO) in a wood-combustion experiment conducted in one household of Spain for 5 days (including the collocation of 2 units of HAPEX and 3 units of TZOA-R for PM_{2.5} and 3 units of EL-USB-CO for CO; a total of 40 unit-days). We used Spearman correlation (ρ) and Concordance Correlation Coefficient (CCC) to assess accuracy of low-cost monitors versus equivalent research-grade devices. We also conducted a field study in India for 1 week (including HAPEX in 3 households and EL-USB-CO in 4 households; a total of 49 unit-days). Correlation and agreement at 5-min were moderate-high for one unit of HAPEX ($\rho= 0.73$ / CCC= 0.59), for one unit of TZOA-R ($\rho= 0.89$ / CCC= 0.62) and for three units of EL-USB-CO ($\rho= 0.82-0.89$ / CCC= 0.66-0.91) in Spain, although the failure or malfunction rate among low-cost units was high in both settings (60% of unit-days in Spain and 43% in India). Low-cost monitors tested here are not yet ready to replace more established exposure assessment methods in long-term household air pollution epidemiologic studies. More field validation is needed to assess evolving sensors and monitors with application to health studies.

Keywords: Household air pollution; Particulate matter; Carbon monoxide; Air quality sensing; Low cost instrument

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HIGHLIGHTS

- We assessed PM_{2.5} and CO low-cost monitors in semi-controlled and field studies.
- Devices experienced high failure rate and susceptibility to environmental changes.
- Low-cost monitors tested may not be ready to replace more established methods.

1. INTRODUCTION

Inefficient combustion of solid fuels such as wood, animal dung, and coal are used by nearly half of the world's population (~3 billion people) for cooking, lighting and heating ¹.

Household air pollution from combustion of solid fuels was ranked the eighth leading risk factor for non-communicable diseases globally in 2015, accounting for an estimated 2.9 million deaths and 85.6 million disability-adjusted life years lost ². Household air pollution is particularly relevant in resource-limited regions where a substantial proportion of the population lacks access to clean household energy, such is the case for South Asia and sub-Saharan Africa, where household air pollution represents the fourth leading environmental risk factor ².

Fine particulate matter (particulate matter with aerodynamic diameter of 2.5 µm or less; PM_{2.5}) and carbon monoxide (CO) are commonly used as indicators of exposure to the mixture of particulate and gaseous products of incomplete combustion resulting from inefficient household fuel use ^{1,3}. Epidemiologic studies focused on quantifying the adverse health effects of chronic exposure to household air pollution would ideally assess participants' exposure to household air pollution over the long-term, perhaps over weeks to months rather than hours or days. However, many of the devices currently available are expensive and have high logistical barriers or suffer from technical constraints (e.g. low battery life, high cleaning frequency, filter exchange, etc.) that preclude continuous and long term monitoring of household air pollution in population-based studies, particularly in low- and middle-income countries where often electricity supply is lacking or unreliable ⁴⁻⁶. Thus, most household air

pollution studies have relied on self-reported data or short-term measurements taken in 24h^{7–16} or in two or three 24-h increments (i.e. 48h, 72h)^{17–21} as proxies for long-term exposure. Although short-term measurements can serve as useful indicators of long-term exposures when taken repeatedly in panel studies, longer term measurements may be preferable to reduce some forms of exposure misclassification and for the study of chronic disease risk. Recent rapid growth of low-cost, easy-to-use, battery-operated, portable air pollution sensors potentially offers important new opportunities for long-term sampling in population-based studies^{22–24}. These off-the-shelf sensors are commonly custom-built in aerosol monitors for different air pollutants and research purposes^{25–30}. Sensors' performance characteristics have been summarized elsewhere^{31,32}, although most of the emerging monitors have not yet been thoroughly characterized^{33,34}. Most of the existing field-validation studies have been done in outdoor environments^{26–28,35–38} such that their performance in real-world indoor environments remains hard to predict. Although remarkable efforts have been done to develop inexpensive and rugged household air pollution-based monitors⁴, validation of affordable measurement technology and approaches to accurately assess long-term household air pollution exposure remains a research priority⁶.

We therefore tested and benchmarked low-cost monitors with potential application for monitoring PM and CO related to household air pollution exposure, by characterizing monitors in terms of accuracy, within-device variability (or inter-variability), response to wide ranges of concentrations and environmental conditions, and ease of use. Specifically, our objective was to evaluate the performance of three monitors (HAPEX, TZOA-R, and EL-USB-CO) with potentially long-lasting batteries in two scenarios: i) a semi-controlled wood-combustion validation study using col-located equivalent benchmark monitors under different concentration and ventilation conditions, and ii) a 1-week field-based pilot study in households in southern India.

2. MATERIAL AND METHODS

2.1 Selection of low-cost monitors

Among all devices available in January 2016, we considered those that met the criteria required for long-term, unattended monitoring of household air pollution in epidemiologic studies: (i) low-cost (< US\$600), (ii) battery-operated with long battery life (> 48h), (iii) not filter-based, (iv) having wide measurement range, and (v) suitable for use across meteorological extremes. All information was extracted from manufacturer datasheets. Based on these considerations, we selected HAPEX (HAPEX Nano, Climate Solutions Consulting, VT, USA) and TZOA-R (Model RD02, MyTZOA, SFO, USA) for assessing PM_{2.5} and EL-USB-CO (Lascar Electronics Ltd., PA, USA) for assessing CO. A table summarising characteristics of all the devices used in the study is shown in Table 1. For more details of these devices and those excluded, see expanded version in Table S1.

2.2 Low-cost monitors

HAPEX Nano is a passive data-logger designed to monitor household air pollution exposure³⁹. HAPEX is built on the Sharp sensor GP2Y1010AU0F (Sharp GP for simplification, Sharp Corporation, Osaka, Japan), which is based on 90° light-scattering technology. Briefly, the Sharp's sensor diode emits a beam of infrared light that illuminates part of the stream of particles that enters the sensing chamber. The light is scattered by particles depending on their size, index of refraction, and light absorbing characteristics. The scattered light is then collected by a light-sensitive transistor (phototransistor), located at 90° of the source beam. The transistor converts light into a voltage, which is proportional to the concentration of particles. Although not size-selective, according to manufacturer HAPEX is most sensitive to PM with aerodynamic diameter between 1 and 3 μm. HAPEX has a rechargeable battery that is advertised to run for 2 years with 20 years of shelf life, although the sampling session cannot last more than 1.2 years in terms of memory capacity.

TZOA-R is a particle counter that simultaneously categorizes particles into three non-overlapping bins, labelled according to the manufacturer as: “ultrafine” (total particles less than 1 μm - PM_{10}), “fine” (total particles sized 1 μm up to 2.5 μm), and “coarse” (total particles sized 2.5 μm up to 10 μm)⁴⁰. However, the TZOA-R minimum detection particle size is 0.3 μm , above the established cut-point of < 0.1 μm for ultrafine particles. Like HAPEX, the aerosol in TZOA-R is analysed through light-scattering technology (with a 655 nm wavelength laser), although in the TZOA-R the air is drawn in by a tiny fan. TZOA-R has a rechargeable battery whose duration depends on the duty-cycle set from the 8GB microSD card (e.g. with a duty-cycle of 0.83%, i.e. 1 reading every 10 minutes, the manufacturer estimates 60 days of battery life). There is the option to switch the device off between readings to reduce the frequency of fan and laser functioning, which saves battery life and lengthens the shelf life of the device by 2 years.

EL-USB-CO is a USB data-logger that uses an electrochemical sensor (the NAP-505, Nemoto Sensor Engineering Company Ltd., Tokyo, Japan) to measure CO. Briefly, gas enters the internal cell of the sensor via a capillary by diffusion, removing unwanted gases with a charcoal based filter. CO is then oxidised after reacting with an electrode. The reactions generate an electrical current proportional to CO concentration. EL-USB-CO has a non-rechargeable internal battery (1/2AA 3.6V) that is advertised to last up to 3 months under specific conditions (i.e. 10min logging rate, 25°C temperature, with audible warnings disabled, and readings below 10 ppm) and with 4 years of shelf life⁴¹ that can be reduced or extended according to the CO levels to which it is exposed (e.g. the lower the levels, the longer it lasts).

The TZOA-R and EL-USB-CO devices were commercially-available and purchased new for this study. We obtained the HAPEX units directly from the manufacturer at no cost for the purposes of testing. Each device (except TZOA-R) was factory-calibrated by the

manufacturer. HAPEX calibration process has been described in detail elsewhere⁴². Briefly, it consists in two steps: i) slope adjustment to reduce inter-device variability down to about $\pm 5\%$ (in a smoke chamber), and ii) zero offset adjustment (in a clean air chamber). All units of EL-USB-CO were calibrated at 250 ppm with a flow rate of 0.5 L/min. We used duplicate or triplicate low-cost monitors to study within-device precision.

2.3 Wood-combustion experiment in Spain

Sampling was conducted during five days in a non-smoking private single-family house in the municipality of Terrassa (Spain) during February-March 2016. The three low-cost monitors (HAPEX, TZOA-R, and EL-USB-CO) were collocated with benchmark devices 1 m from an indoor fireplace and 0.6 m above the ground. The fireplace was the sole source of heat in the house. We used newspaper to start the fire; the fire was maintained for 12h using dried hardwood logs from Holm oak tree. We weighed the wood fuel prior to starting the fire each day. We controlled air ventilation conditions during fire hours each experiment day according to a pre-specified protocol by opening or closing the living room window (120 x 140-cm), such that improved ventilation hours (21 / 120h) covered different fire phases (ignition, steady, extinction) (Figure S1).

We used the DustTrak DRX (Model 8534, TSI Inc., MN, USA) and the Q-Trak (Model 7575, TSI Inc.) as the PM_{2.5} and CO benchmark monitors, respectively. Although they are not regulatory-grade monitors, their portability and affordability have made them widely used in prior air quality research studies^{4,25,27,28,43-46}. We also included a PM_{2.5} sampling pump (BGI4004, BGI/Mesa Labs Inc., Waltham, MA, USA) provided with a cyclone sampler (GK2.05SH (KTL), BGI/Mesa Labs Inc.) and 37-mm and 2- μ m pore size Teflon filters (Zefon International Inc., Ocala, FL, USA). Five gravimetric samples were obtained each experiment day; filters were changed every 4h during the daytime and after 8h during the

night time (Figure S1) in order to obtain a larger number of measurements (n= 25). All pollutant concentrations measured in the experiment are shown in Table S2.

2.4 Field-based study in India

Field sampling was conducted in 4 convenience-sampled households located in 4 villages outside of Hyderabad (southern India) during March-April 2016 (see Figure S2 for the map of villages). Participants were part of the CHAI project (Cardiovascular Health effects of Air pollution in Telangana, India) ⁴⁷. Based on our experience in the wood-combustion experiment in Spain, we tested only the HAPEX and EL-USB-CO devices in the Indian households. In each household we placed one HAPEX and one EL-USB-CO unit in the main living area at least at 1 m above the ground for 1 week. The first 24h, devices were collocated with a PM_{2.5} sampling pump (Universal 224-PCXR8, SKC Inc., Eighty Four, PA, USA) provided with a Triplex cyclone (SCC1.062, BGI/Mesa Labs Inc.) and 37-mm Pallflex[®] Emfab[™] membrane filters. Ambient temperature and relative humidity were measured using LabJack (Model Digit-TLH, LabJack, Lakewood, CO, USA) in a CHAI background fixed station.

Baseline and post-monitoring questionnaires were available for all households. Post-monitoring questions related to the first 24h of sampling and included type of cooking and lighting fuels used during monitoring, quantity of the cooking fuels used (weighed with a balance pre- and post- sampling), cooking characteristics (e.g. type and location of the stove used, number of windows opened, cooking time), household characteristics (e.g. location, size, kitchen type) and the use of other sources contributing to indoor air pollution (e.g. smoking, incense, mosquito coil).

2.5 Data post-processing

Since light-scattering readings are dependent on the site-specific characteristics of the aerosol sampled (e.g. size), we corrected all DustTrak DRX raw data using the linear function obtained from our 25 co-located gravimetric measurements. This correction is also relevant for the low-cost PM_{2.5} monitors, which are not direct-mass reading: HAPEX is unitless and TZOA-R is a particle counter. For HAPEX and TZOA-R we obtained the correction factor as follows:

Correction Factor= [average PM from gravimetric] (in $\mu\text{g}/\text{m}^3$) / [average PM from light-scattering] (raw unit)

Since gravimetric methods are cumbersome and expensive, for long-term sampling ideally one would do a collocation with the low-cost device for the shortest time possible to obtain an on-site correction factor. To simulate this scenario, we obtained the correction factor using the first 24h for HAPEX (in both settings) and for the last 24h for TZOA-R (in the experiment; due to TZOA-R's failure the first 3 days as is further explained in subsection 3.1). All HAPEX and TZOA-R raw data were then multiplied by its respective 24-h correction factor to get mass concentration and to account for the characteristics of the aerosol sampled. As a sensitivity analysis, we further evaluated the potential additional benefit (if any) of using longer gravimetric collocations calculating correction factors from 1 to 5 days. Since light-scattering devices tend to underestimate PM at low relative humidity (RH) (< 40%) and overestimate PM at high RH (> 60%)⁴⁸, we adjusted all light-scattering PM data (i.e. DustTrak, HAPEX, and TZOA-R) for humidity effects using a standard adjustment equation⁴⁹. All PM_{2.5} data shown is filter- and humidity-corrected.

We defined monitor failure when the monitor did not provide data (i.e. missing values or repeated zeros). Failure rate was calculated dividing the failed unit-days (e.g. 2 units \times 5 days = 10 unit-days) by the total unit-days. We judged a monitor to be unreliable when values and

time-series patterns were considered implausible given the sampling conditions or contextual information.

Scripts for reading the raw output files from the low-cost monitors are provided in Supplementary Material. Raw PM_{2.5} and CO data from all devices used in the wood-combustion experiment is available for download at <http://hdl.handle.net/10230/28201>.

2.6 Statistical analysis

We used Spearman correlation coefficients and concordance correlation coefficients (CCC) to evaluate the correlation and agreement respectively between PM_{2.5} and CO levels from each low-cost monitor and the corresponding benchmark monitor in the experiment. CCC, also known as Lin's coefficient, is in practice similar to the intraclass correlation coefficient (ICC)⁵⁰. CCC takes into account how far observations are from the best-fit line and how far the best-fit line is from the perfect agreement line (i.e. 45-degree line) when the results from one device are plotted against the other⁵⁰. Bland-Altman plots were also produced. We also calculated these metrics to assess correlation and agreement among multiple units of the same low-cost devices (within-device variability) and to compare the PM₁ and PM₁₀ fractions given by TZOA-R and DustTrak DRX.

We fitted linear regression models between the low-cost (dependent) and its equivalent benchmark device (independent) values allowing for different intercepts and slopes under each condition: “Non-fire”, “Fire, window opened”, and “Fire, window closed”. As our primary performance metric, we chose the adjusted coefficient of determination (or R²). We also calculated the low-cost monitor partial contribution to R²⁵¹. We considered both logarithmic and squared root transformations.

All analyses were conducted with R (version 3.3.1, the R Foundation for Statistical Computing, <https://www.r-project.org/>) using “corrplot” and “epiR” packages. Additional details about the methods, such as the dimensions of the sampling sites and the filter weight process is in Supporting Information.

3. RESULTS

3.1 Failure rate of low-cost monitors: wood-combustion experiment

One of the two HAPEX units and two of the three TZOA-R units failed early in the experiment. Data from the first three sampling days of the remaining TZOA-R unit contained repeated zeros. Thus, PM_{2.5} data was available from only one HAPEX unit (with 7,232min of data) and one TZOA-R unit (with 2,315min of data) (see time-series in Figure S3). The unsuccessful HAPEX unit gave repeated missing values, which was determined to be part of a defective batch according to manufacturer. One of the unsuccessful TZOA-R devices had a broken fan; the cause of failure of the other TZOA-R unit is unknown. On the other hand, all the three EL-USB-CO units took CO measurements during all the experiment days. CO levels from all the three units, however, were disproportionately higher (an average of 48 times higher) the last two days than the first days of operation (Figure S4). This could be because the last two days the fire was more intense, and higher intensity increases temperature, lowers RH, and limits the oxygen entrance into the flame zone, which creates high concentrations of CO⁵². As a sensitivity analysis, we therefore created five regression models, one per each sampling day, adding temperature as an explanatory variable. Temperature had a positive and significant effect on EL-USB-CO measurements, but the magnitude of the effect was ≈ 20 times larger the last two days (3.5 and 3.1 ppm increase for each °C) than the first days (0.02-0.153), indicating that the effect of temperature was not constant across days. We therefore judged the monitor to have been unreliable and performed the analysis omitting the last two days (i.e. $\sim 4,390$ min).

3.2 Wood-combustion experiment characteristics

Wood consumption averaged (min-max range) 50.7 (42.5 – 70.0) kg · d⁻¹. According to the EL-USB-2-LCD data-logger, temperature and RH reached during fire hours ranged from 19 to 55.5°C and from 10 to 48%, respectively (Table S3). There were no high-humidity episodes; the maximum RH reached if taking into account non-fire hours was 53.5%. Correlation (ρ) and agreement (CCC) between the EL-USB-2-LCD data-logger (low-cost) and the Q-Trak (benchmark) was high for both temperature ($\rho= 0.95$ / CCC= 0.80) and RH ($\rho= 0.96$ / CCC= 0.89). There was a high linear correlation between the DustTrak DRX and gravimetric (BGI pump) techniques (Figure S5).

3.3 Low-cost versus benchmark devices: wood-combustion experiment

Correlation and concordance matrices of all the PM_{2.5} and CO devices used are shown in Figure 1. Bland-Altman plots are shown in Supplementary Material (Figure S6, S7, S8).

When compared to BGI, using 4h (daytime) or 8h (night time) averaging times, correlation (ρ) and agreement (CCC) of HAPEX ($\rho= 0.65$ / CCC= 0.58) and TZOA-R ($\rho= 0.69$ / CCC= 0.66) were each moderate. When compared to DustTrak DRX, 5-min correlations were found to be higher for both low-cost monitors ($\rho= 0.73$ / CCC= 0.59 for HAPEX; $\rho= 0.89$ / CCC = 0.62 for TZOA-R), while agreement remained moderate. The three col-located units of EL-USB-CO showed moderate-high correlation and agreement when compared to Q-Trak ($\rho= 0.82$ -0.89 / CCC= 0.66-0.91) and between units of the EL-USB-CO ($\rho= 0.80$ -0.93 / CCC= 0.41-0.84).

When the averaging time was increased to 1-h, the correlation between DustTrak DRX and HAPEX was lower than the 5-min averaging time, although the agreement was slightly higher ($\rho= 0.68$ / CCC= 0.66). For TZOA-R, the 1-h averaging time did not materially change the correlation, but substantially improved the agreement with the DustTrak DRX ($\rho= 0.91$ / CCC= 0.81). Longer averaging time did not materially change the correlation or agreement

between the EL-USB-CO with its benchmark instrument (Q-Trak), but did result in improved within-device correlation and agreement ($\rho = 0.82-0.94$ / $CCC = 0.51-0.86$). The correlation and agreement when comparing the PM_1 fraction of TZOA-R with DustTrak DRX were moderate-high ($\rho = 0.90$ / $CCC = 0.64$), whereas they were very poor when considering the PM_{10} fraction ($\rho = 0.46$ / $CCC = 0.21$). When applying gravimetric correction to HAPEX using gravimetric data from all the sampling days (rather than only using the first 24h of col-located data), 5-min agreement between HAPEX and DustTrak DRX improved modestly from $CCC = 0.59$ (correction based on 1 day of gravimetric data) to $CCC = 0.66$ (correction based on 5 days).

Scatter plots showing pollutant levels stratified by fire and room ventilation conditions and by low-cost devices are shown in Figures 2, 3 and 4. Since the performance of the low-cost monitors varied according to each of the three conditions, we included the 3-category variable as an interaction term in all models. When only analyzing the last two days of data to allow comparison between HAPEX and TZOA-R, model R^2 values were 74% for HAPEX and 85% for TZOA-R. The partial R^2 for the effect of the device (Table S3) was 43% for HAPEX and 76% for TZOA-R, indicating that HAPEX was highly influenced by the fire and ventilation conditions. HAPEX performance was poor during fire and window opened hours (Figure 2, central panel), substantially overestimating DustTrak DRX values. TZOA-R values were consistently lower than DustTrak DRX in all conditions, especially during non-fire conditions (Figure 3, left panel). When analyzing only the first three days of data for unit 1 of EL-USB-CO (Figure 4), R^2 was 82% and the partial contribution to R^2 for the device was 71%. All units of EL-USB-CO (see Figure S9 and S10 for the other two units) clearly underestimated the Q-Trak values during non-fire hours (left panels). Regarding the other conditions, there was not a clear pattern since performance depended on the condition and the device unit under consideration. Logarithmic and squared root transformations for all pollutants did not improve the model fit.

3.4 Field-based study characteristics

All households monitored were non-smoking single houses located in residential areas with the kitchen in a separated room from the living area. All households used liquefied petroleum gas (LPG) and electricity as the only sources of cooking and lighting, respectively.

Although not used the first day of sampling, three households reported periodic use (2-10 times/month) of a secondary outdoor stove using either coal (household 2) or biomass (households 3 and 4) fuel.

During the first 24h of sampling, the total cooking time per household varied from 90 to 190min consuming between 175-425 g of LPG; half of the households had a window open near the LPG stove. One household (household 3) reported the use of incense (9h duration) and an oil lamp (45min) in the living area during the first 24h of sampling. $PM_{2.5}$ and CO concentration, temperature and RH ranges were overlapping between the semi-controlled sampling in Spain and the real-world sampling in India (Table 2). However, the influence of RH on $PM_{2.5}$ values was more evident in India than in Spain.

Temporal variability of $PM_{2.5}$ and CO over 1 week is shown in Figure 5 (using 6,010 filter- and humidity-corrected 5-min of $PM_{2.5}$ data, and 8,317 5-min of CO). $PM_{2.5}$ and CO peaks did not coincide and correlation based on 5-min average data between pollutants was weak for the three households with simultaneous data: 0.05, -0.04, and -0.17. Although none of the monitors failed in India, we considered EL-USB-CO to be unreliable in three of the households (1, 2, and 4) because there was a noticeable baseline shift in the EL-USB-CO time-series (i.e. baseline values were not constant over time) that was accompanied by an unexpected decreasing pattern of CO. None of these households reported any type of indoor burning activity the first 24h. We observed increases in $PM_{2.5}$ in the living area corresponding to self-reported cooking activities during the first 24h, but no corresponding increase in CO.

4. DISCUSSION

All three low-cost monitors evaluated in the semi-controlled experiment showed very high (> 40%) failure or malfunction rates, with TZOA-R reaching the highest: 86% of unit-days.

Under more heterogeneous sampling conditions in India, HAPEX and EL-USB-CO devices showed no device failures, but unreliable CO patterns, suggesting EL-USB-CO malfunctioning. Successful low-cost units showed moderate to high correlation and agreement when compared side-by-side with more robust benchmark monitors. However, the performance of HAPEX (based on the Sharp GP sensor) and EL-USB-CO (based on the NAP-505 sensor) was influenced by the conditions created in the experiment (e.g. increased natural ventilation).

All devices were previously unused, therefore failures were unlikely due to instrument fatigue. Since the PM monitors tested were prototypes (alpha version for HAPEX and beta version for TZOA-R), their high failure rate may have resulted from early stage design problems (e.g. TZOA-R had a firmware bug in the internal Real Time Clock, resulting in poor time stamp mapping). However, this is not the case of the EL-USB-CO, which is an established device that has been previously used in household air pollution studies as a personal monitor^{53,54} and as an indoor monitor^{16,55,43,56,57}. Generally it has been used to measure CO for periods of 24h or less, although three studies have attempted to use it for longer periods^{43,54,56}. None of these studies showed unexpected patterns of CO, although Ochieng *et al*⁵⁶ also suggested EL-USB-CO malfunctioning after obtaining 48-h average CO concentrations of 0 ppm in two households. The expected pattern, shown in Chen *et al*¹⁶ and Tumwesige *et al*⁵⁷ and also described in Quinn *et al*⁵⁴, is a flat line with CO background levels close to 0 indicating the absence of any combustion source, with some peaks indicating a prompt burning activity, similar to what we obtained for household 3 in Figure 5. For other households, we observed a week-long baseline shift with an implausible decreasing CO pattern not attributable to self-reported burning activities and not correlated with simultaneous

PM_{2.5} concentrations. The high temperatures and the low oxygen and humidity conditions achieved in the last two monitoring days in the experiment could have caused the instability problems found in EL-USB-CO, which typically occur in electrochemical-based monitors³². However, authors cannot ensure to what extent this situation accelerated sensor aging and lead to the malfunctioning later in India. But taking into account the days that the monitor failed and the time (i.e. after starting a new monitoring session), malfunction was likely due to zero drift, which accentuates the need to zeroing the device before each deployment.

Although the EL-USB-CO has been widely used to assess CO levels in different settings^{16,43,53–58}, there are limited published reports evaluating the performance of the EL-USB-CO monitor versus benchmark monitors under semi-controlled or real-world conditions.

However, we highlight the work in Ghana of Piedrahita *et al*⁵⁸ who identified failure and malfunction of the EL-USB-CO in 25% of their sampling days and subsequently conducted laboratory tests to further investigate its performance. They concluded that frequent calibration of the EL-USB-CO is needed, since using raw data (as we did) can produce an average exposure error of 19% (versus 1.9% doing multiple calibrations each sampling day and 1.2% doing pre- and post-calibrations)⁵⁸.

The peer-reviewed literature on low-cost monitor validation studies in real-world conditions is scarce^{33,34}. Some agencies, university departments and research groups have made public their low-cost monitors performance evaluations^{59–61}. Although some of these evaluations are performed for weeks or use true reference monitors such as Federal Equivalent Methods (FEM), they are usually done in laboratory chambers^{25,44,61–66} or in fixed outdoor locations^{26–28,37,38,44,65}, not reflecting a wide range of realistic indoor conditions, such as those reached in our study. However, the field-based study done by Semple *et al*⁶⁷ is notable as they co-located a low-cost particle monitor, the Dylos, against a TSI SidePak under real-life indoor smoking conditions in 17 Scottish homes during approximately 24h. Although Dylos was found to be a useful low-cost monitor for indoor environments, it was excluded from our

analysis for having a short battery life (~6h; Table S1). None of these evaluations include the monitors tested in this study although the sensor inside the HAPEX (the Sharp GP) has been evaluated^{31,35,45,63,64} and as part of other monitors (e.g. Foobot, TSI AirAssure)^{4,35,36,59,60,62,66}. Manufacturers of HAPEX and TZOA-R tested their monitors under laboratory conditions before making them commercially available. Both low-cost monitors were respectively collocated inside a test chamber with an Indoor Air Pollution Meter (Aprovecho Research Center, OR, USA) and a DustTrak DRX (Model 8533) similar to the one used in this study. Both R-squared reported were very high: 0.99 for HAPEX⁴² and 0.89-0.92 for TZOA-R⁴⁰. In contrast, our study found lower R-squared for HAPEX ($R^2 = 0.74$) and TZOA-R ($R^2 = 0.85$) both at 5-min averaging time, and only a moderate agreement when compared to a gravimetric method (CCC= 0.58 for HAPEX; CCC= 0.66 for TZOA-R) at 4h/8h averaging times, suggesting that the performance of low-cost monitors can vary considerably outside a controlled testing laboratory, as has been previously pointed out^{38,64,65}. Furthermore, these manufacturer's tests were performed over relatively short time periods, generally less than 12h, and in the case of HAPEX less than 1h. Due to the logistical constraints of continuously maintaining the fire in the wood-combustion experiment, we did not evaluate the performance of the low-cost monitors against benchmark devices over long time periods relevant for household air pollution research (e.g. ≥ 1 week). However, our data provide insights on the performance of these devices with ≥ 2 continuous days of sampling, a considerable improvement above the duration typically used in laboratory testing.

The performance of the low-cost devices varied across concentration ranges and ventilation conditions created in the experiment. The HAPEX device was particularly sensitive to the fire and ventilation conditions, with large overestimates during hours with increased ventilation. This may be because HAPEX is a passive device that operates on the principle of diffusion, such that higher rates of natural ventilation may have affected its performance. This finding is not surprising as increasing ventilation transforms the relatively homogenous aerosol mix into

a relatively heterogeneous mixture of indoor and ambient atmospheres. This is consistent with previous work done by Olivares *et al*³⁵, who evaluated the performance of the ODIN monitor, which is also based on the Sharp GP, in an outdoor environment influenced by woodsmoke. When compared to a TEOM-FDMS, ODIN tended to slightly overestimate PM_{2.5} values as wind speed increased. As other PM sensors⁶³, Sharp sensor performs worst at low concentrations, which are often created at higher wind speeds³⁵. TZOA-R generally underestimated PM_{2.5} concentrations, but its performance was less sensitive to changes in conditions resulting from the fire and increased ventilation. The EL-USB-CO also tended to underestimate CO levels, being particularly relevant during hours with no fire. This is probably because of the differences in accuracy and resolution between EL-USB-CO (± 7 ppm accuracy; 0.5 ppm resolution) and Q-Trak (± 3 ppm; 0.1 ppm), which may be more important when CO values are closer to 0. Although overall we found moderate-high within-device correlation for the three EL-USB-CO units, each device had different sensitivity to conditions created in the experiment. Similarly, Ochieng *et al*⁵⁶ found high within-device variability (24%) when collocating 12 EL-USB-CO units in Kenya for 48-h three times.

The suitability of emerging low-cost air quality monitors for long-term household air pollution assessment depends not only on measurement performance, but also reliability, ease of use for researchers, and impact on participant burden. All of the devices tested in this study have a lightweight, miniaturized, and silent design that make them well suited for personal and area monitoring. However, there were remarkable differences between the devices measuring PM in terms of ease of use. Whereas HAPEX software provides an interface that we found to be user-friendly and intuitive, TZOA-R lacked such an interface (data need to be copy-pasted from the microSD card, which is more prone to errors during data management) and we also found that the TZOA-R operating manual was not clear regarding the pre-sampling settings (e.g. distinguishing between “sample_period” and “sample_time”). On the other hand, none of the PM monitors are direct-mass reading, which means they need to be

collocated with cumbersome gravimetric devices in order to obtain mass concentration measurements. Although this has been pointed out as a main concern for light-scattering monitors³³, we found that one day of collocated monitoring is adequate for gravimetric correction: adding an additional four days of data improved the agreement between HAPEX and DustTrak DRX by approximately 12%. Additionally, it is well-known that sensors' performance depends on the type of aerosol being sampled (e.g. the Sharp GP sensor is sensitive to organic and smaller particles)⁶³, and this is particularly relevant in household air pollution-related research where correction factors are influenced by fuel and combustion characteristics, which differ across households and seasons⁴. One advantage of TZOA-R is that it includes temperature and humidity sensors that are used as input in an automated adjustment step, thus separate temperature and RH correction would not be necessary. TZOA-R also performed quite well for particles <1 μm in diameter, making it a potentially useful candidate in studies focusing on exposures to smaller size fractions and adverse health effects, which has been recognized a major challenge for low-cost sensing²³.

This is one of the few published studies characterizing the performance of low-cost monitors with potential application for long-term household air pollution research in conditions commonly encountered in low- and middle-income countries. Other strengths of our study include evaluation of multiple monitors, over the course of multiple days and across a range of air pollution levels and environmental conditions such as natural ventilation and temperature stress. While the conditions in the wood-combustion experiment provide unique insights into the performance of these devices in a more realistic environment than a laboratory setting, they are not likely representative of some conditions typically observed in household air pollution studies conducted in low-resource settings, especially in environments with high RH. Nonetheless, the pollutant concentrations, temperature, and RH conditions encountered in our field study in southern India were not dissimilar from those in the semi-

controlled experiment, and these additional data provide an indication of the device performance under more realistic conditions over one week.

Limitations of the study also should be considered. This study was not intended to conduct a systematic and comprehensive evaluation of all monitors available in the market. Rather we chose monitors that we thought potentially useful for our future studies on the basis of potential for long-term, unattended sampling. Although the benchmark monitors used are expensive research-grade monitors, they are not true “reference” monitors. We did a custom calibration with a col-located gravimetric sampler to correct DustTrak DRX values, but we did not calibrate the Q-Trak monitor as we did not have a reference method to measure CO. The small sample size and convenience sampling in the pilot study in India included only households using relatively clean household energy for cooking, which might not be comparable to households relying primarily on biomass fuel for cooking. Another limitation of our study is that we were not able to study the within-device variability in all low-cost devices because of the high rate of device failure. Although we corrected our PM data for meteorological outdoor data, we had no indoor temperature and RH data to more accurately correct the PM concentration in the Indian households. This also impeded us to further explore the role of changing indoor temperatures to the CO pattern observed, although if temperature had had a big impact, CO patterns would have been cyclic and diurnal throughout the week rather than consistently decreasing. This is an important limitation given that both light-scattering and electrochemical sensors have temperature and RH dependence. Manufacturers should provide calibration equations or tables with correction constants to help users to better deal with meteorological changes. Future field studies should include additional temperature and RH monitors or multi-sensor monitors like TZOA-R to correct for indoor meteorological conditions.

Low-cost technology for measuring air quality is a quickly evolving field and device models are often updated in quick succession; more advanced sensors/monitors will continue to change the landscape of air quality monitoring⁶⁸. Since our research was completed, HAPEX and TZOA-R have launched newer versions. According to the manufacturer, HAPEX version 3.0 incorporates improvements in the user interface and the algorithms that reduce noise, reaching 100% working rates. Similarly, TZOA-R has launched the RD03, which adds a Volatile Organic Compound (VOC) sensor, longer lifetime and better sensitivity and accuracy, among other improvements. Other potentially-useful devices that should be included in upcoming evaluations for monitoring household air pollution is the PATS+⁴, successor of the UCB-PATS and also built on the Sharp GP sensor, but not available at the time of our study (Table S1).

5. CONCLUSIONS

Advances in battery technology have improved the feasibility of continuous monitoring over periods greater than 24h. But in the light of our semi-controlled validation and field-based measurements, none of the low-cost monitors we tested seem ready to replace more established household air pollution measurement devices. The next generation sensors and/or monitors may deliver more material benefits in long-term monitoring of air pollution exposure. However, real-world or in-field validation studies of these devices are essential before they are deployed in household air pollution-related research. Further evaluations should include several weeks of continuous sampling to test stability and durability, multiple units of each device, and cover a range of concentrations, temperatures, and humidity levels.

DECLARATION OF INTEREST

None declared.

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Table 1 Summary of device characteristics.

| Device (Model/version) | Measure(s) | Raw output | Sampling site | Number of units used | Range¹ | Type | Approximate battery life¹ | Maximum Operating Temperature (°C)¹ | Cost per unit (US Dollars) |
|---|---|-----------------------|----------------------|---------------------------------|--|-------------|---|---|---|
| DustTrak DRX (Model 8534, hand-held) | PM ₁ , PM _{2.5} , PM ₄ , PM ₁₀ | Mass concentration | Spain | 1 | 0.001 to 150 mg/m ³ (PM _{2.5}) | Benchmark | 6 hours | +50 | 7,900 |
| BGI/Mesa Labs pump (Model BGI4004) - discontinued | PM _{2.5} , absorbance ² | Non applicable | Spain | 1 | Non applicable | Benchmark | 24 hours | +50 | >1,000 |
| SKC pump (Model Universal PCXR8) | PM _{2.5} , absorbance ² | Non applicable | India | 1 | Non applicable | Benchmark | 12 hours (with extended times with intermittent sampling) | +40 | >1,000 |
| TZOA-R (version RD02) | PM ₁ , PM _{2.5} , PM ₁₀ , T, RH | Particle counts | Spain | 3 | Not reported | Testing | 60 days (10min logging rate) | Not reported | 400 |
| HAPEX Nano | PM _{2.5} | Unit less | Spain / India | 2 | 5 µg/m ³ to 150 | Testing | 2 years | Not reported | 95 |

| | | | | | | | | | |
|---|--------------------------------|--------------------|---------------|---|----------------------------|---------------|-------------------------------------|-----|-------|
| (version 1.0) | | | | | mg/m ³ | | | | |
| Q-Trak (Model 7575) | CO ₂ , CO, T, RH | Concentration | Spain | 1 | 0 to 500 ppm (CO) | Benchmark | 6 hours | +45 | 3,100 |
| EL-USB-CO (Model EL-USB-CO) | CO | Concentration | Spain / India | 3 | 3 to 1,000 ppm | Testing | 3 months (10min logging rate) | +40 | 125 |
| EL-USB-2-LCD (Model EL-USB-2-LCD) | T, RH | T and RH levels | Spain | 1 | -35 to +80°C, 0 to 100% | Complementary | 3 months | +80 | 95 |
| LabJack (Model Digit- TLH)- discontinued | T, RH | T and RH levels | India | 1 | -35 to +85°C, 5 to 95% | | 3.3 years | +85 | 36 |

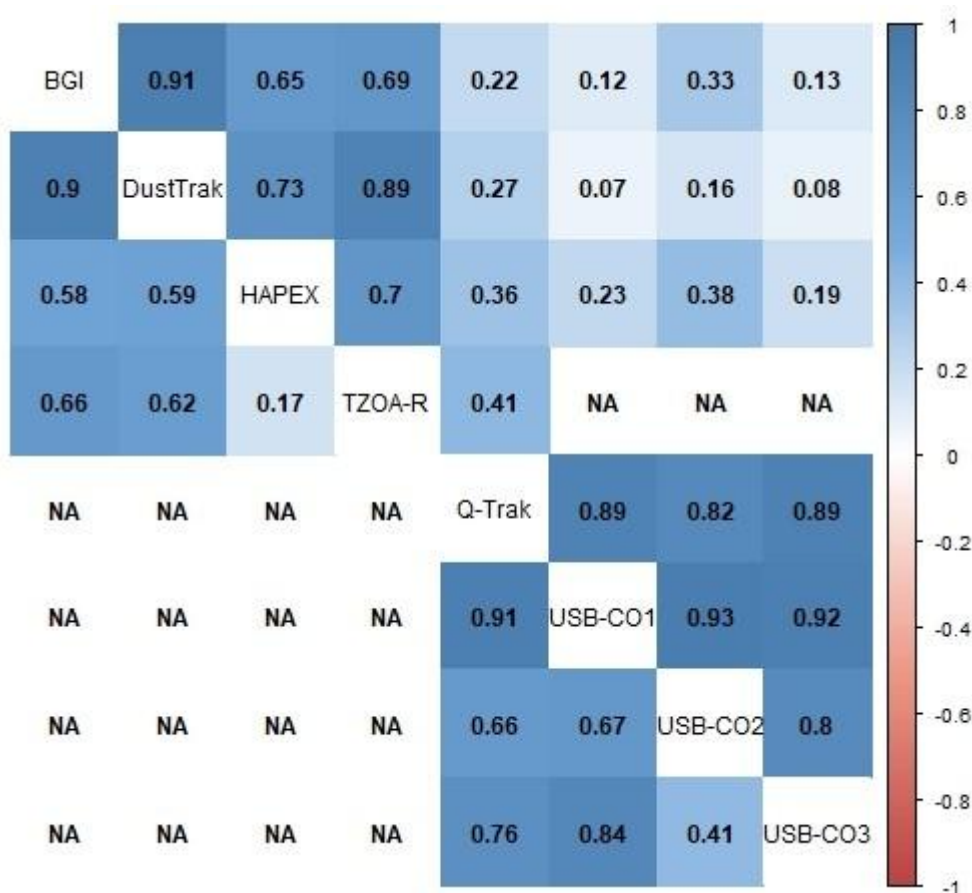
PM₁: particles less than 1 µm; PM_{2.5}: particles less than 2.5 µm; PM₄: particles less than 4 µm; PM₁₀: particles less than 10 µm; T: Temperature; RH: Relative Humidity; CO₂:

Carbon Dioxide; CO: Carbon Monoxide.

¹ *According to operating manufacturer manuals or datasheets. Battery life varies according to the settings specified.*

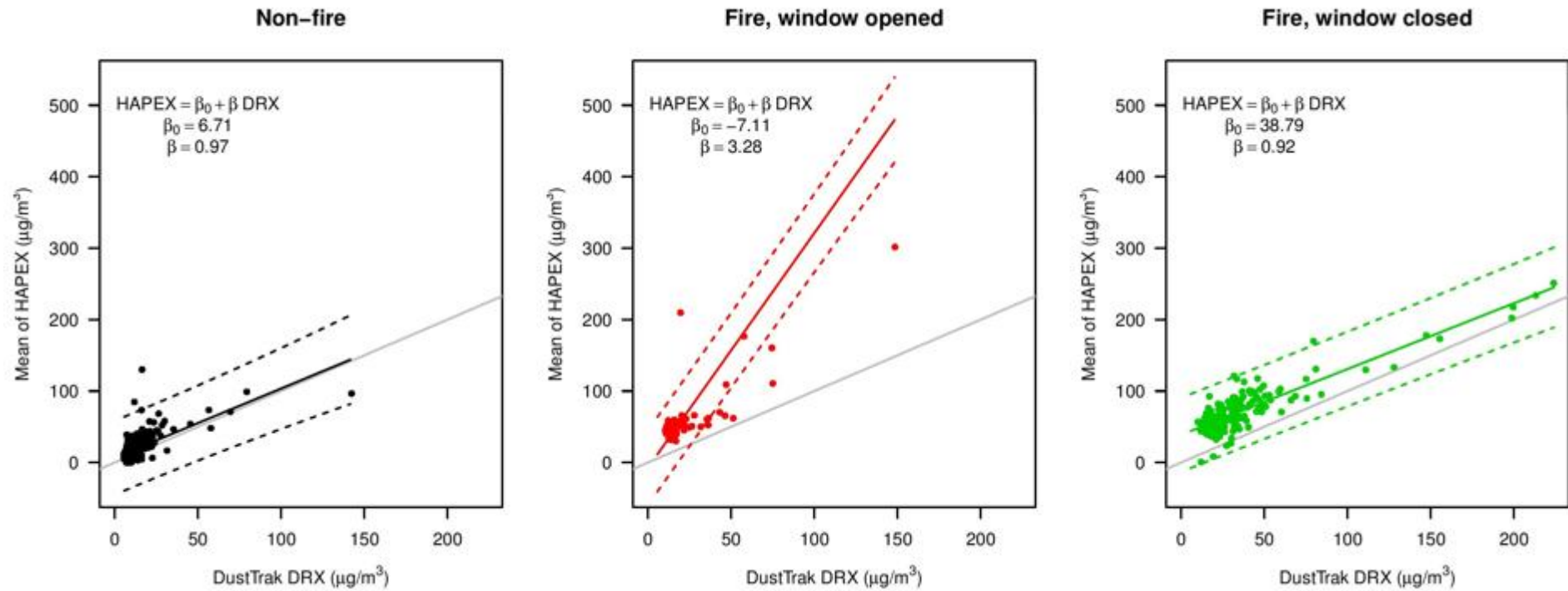
² *Absorbance was also measured both in Spain and India, but data are not shown.*

Figure 1 Correlation (above the diagonal) and concordance (below the diagonal) matrices between the low-cost devices and the benchmark monitors used in the experiment in Spain.



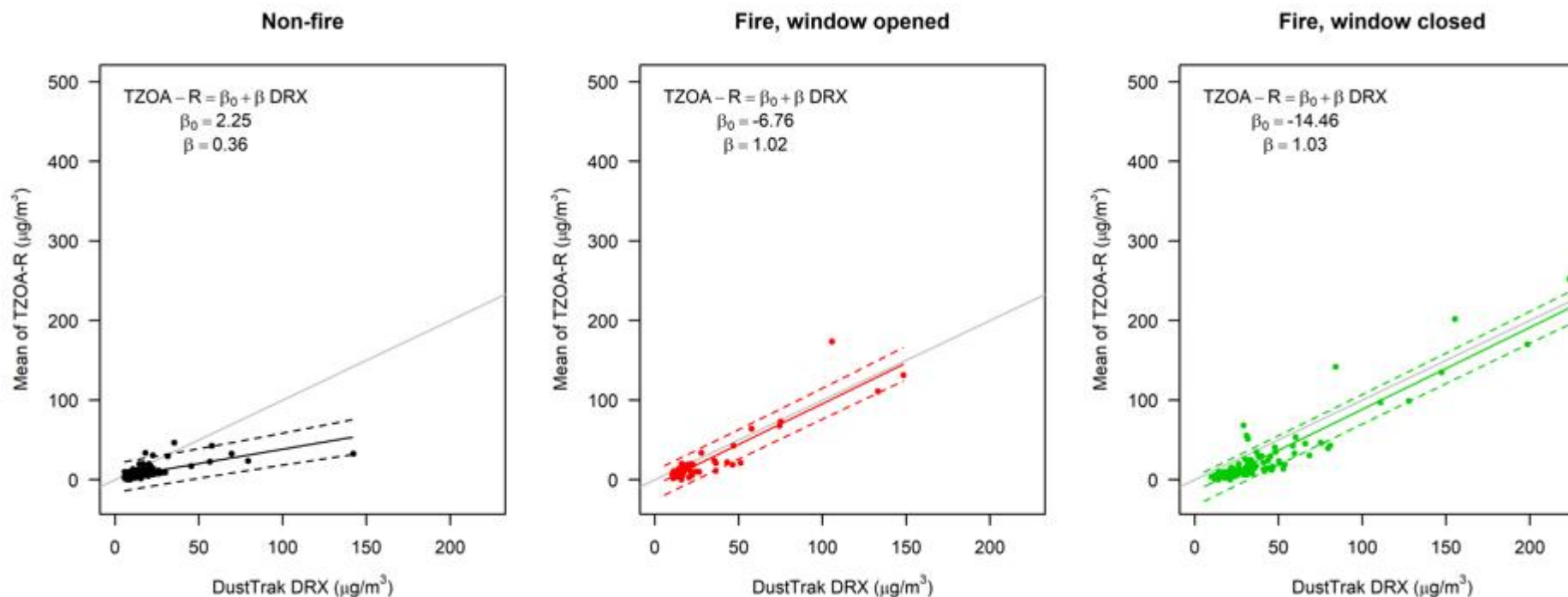
BGI and DustTrak DRX: benchmark $PM_{2.5}$ monitors; HAPEX and TZOA-R: low-cost $PM_{2.5}$ monitors; Q-Trak: benchmark CO monitor; (EL-)USB-CO: low-cost CO monitor. The number at the end of each (EL-)USB-CO indicates a different unit of the same device. To obtain both matrices, $PM_{2.5}$ data from the 5 sampling days was used for all monitors except for TZOA-R, for which only data from the two last sampling days was obtained. To obtain both matrices, CO data from the last two sampling days was omitted due to monitor malfunctioning. The “NA” in the case of TZOA-R correlations means that no simultaneous and valid data are available for both devices. Note that correlation and agreement for BGI pairs were averaged over 4h for daytime / 8h for night time. All other data in Figure are based on 5min averages. DustTrak DRX, HAPEX, and TZOA-R data presented here are adjusted gravimetrically and by relative humidity. Colour legend bar at the right indicates the magnitude of the correlation and agreement from -1 to 1; lighter cells indicate proximity to 0.

Figure 2 Scatter plots of 5-min $PM_{2.5}$ levels from HAPEX (low-cost) versus DustTrak DRX (benchmark) during the experiment in Spain stratified by fire and room ventilation conditions.



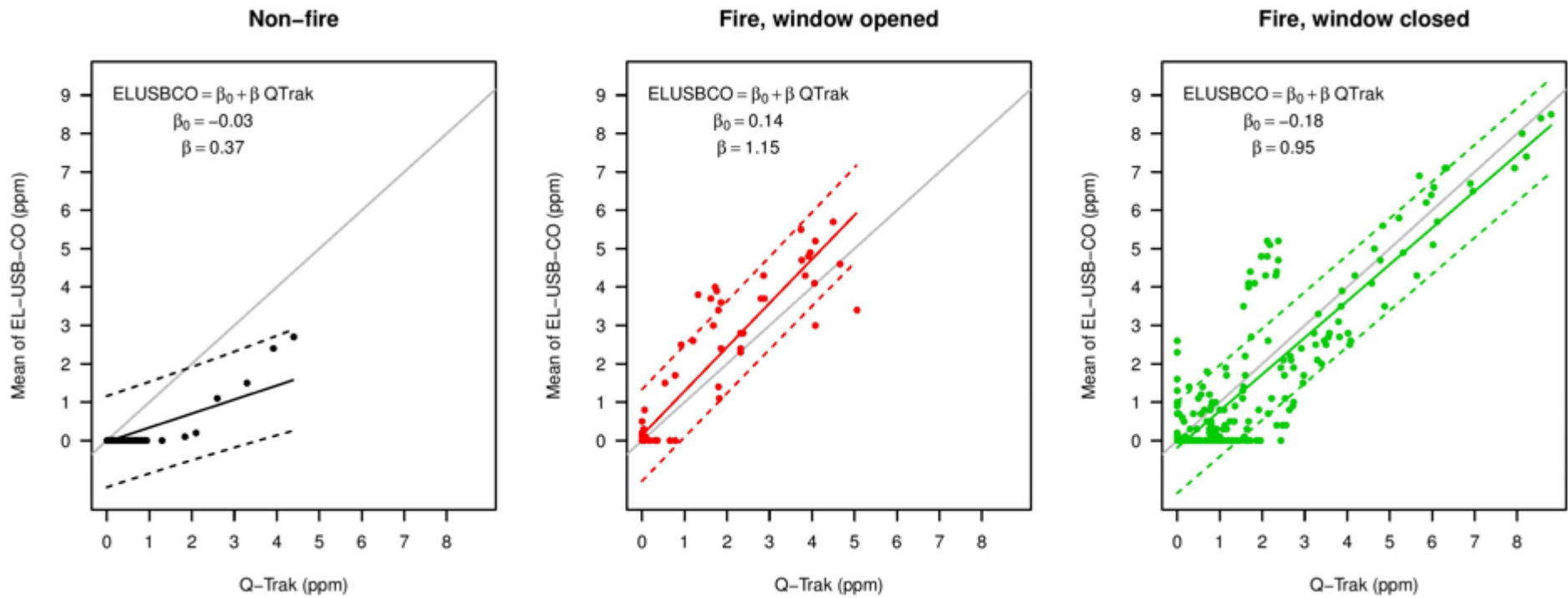
$PM_{2.5}$: particles less than $2.5 \mu\text{m}$ (in $\mu\text{g}/\text{m}^3$). Plots include only the last two days to allow comparison with TZOA-R. Solid lines correspond to the fitted mean concentration of HAPEX. Dashed lines correspond to the 95% confidence interval for the prediction. Grey lines represent the ideal ($\text{HAPEX} = \text{DustTrak DRX}$). The fitted linear model showed an $R^2 = 0.74$. All data presented here are adjusted gravimetrically and by relative humidity.

Figure 3 Scatter plots of 5-min $PM_{2.5}$ levels from TZOA-R (low-cost) versus DustTrak DRX (benchmark) during the experiment in Spain stratified by fire and room ventilation conditions.



$PM_{2.5}$: particles less than $2.5 \mu m$ (in $\mu g/m^3$). Plots include only the last two days. Solid lines correspond to the fitted mean concentration of HAPEX / TZOA-R. Dashed lines correspond to the 95% confidence interval for the prediction. Grey lines represent the ideal ($TZOA-R = DustTrak DRX$). The fitted linear model showed an $R^2 = 0.85$. All data presented here are adjusted gravimetrically and by relative humidity.

Figure 4 Scatter plots of 5-min CO levels from unit 1 of EL-USB-CO (low-cost) versus Q-Trak (benchmark) during the experiment in Spain stratified by fire and room ventilation conditions.



CO: carbon monoxide (in ppm). Plots include only the first three days of sampling. Solid lines correspond to the fitted mean concentration of EL-USB-CO. Dashed lines correspond to the 95% confidence interval for the prediction. Grey lines represent the ideal ($EL-USB-CO = Q-Trak$). The fitted linear model showed an $R^2 = 0.82$.

Table 2 Comparison of PM_{2.5}, CO, temperature, and relative humidity between sampling sites (Spain vs India).

| | PM _{2.5} (in µg/m ³), by HAPEX | | | CO (in ppm), by EL-USB-CO | | | Temperature (in °C) | | | Relative Humidity (in %) | | |
|---|---|--------|-----------------|---------------------------|--------|------------|---------------------|--------|--------------|--------------------------|--------|-------------|
| | Mean ± SD | Median | Min to max | Mean ± SD | Median | Min to max | Mean ± SD | Median | Min to max | Mean ± SD | Median | Min to max |
| Spain (5-day for PM _{2.5} ; 3-day for CO) | 41.4 ± 46.7 | 34.9 | -10.0 to 729.5 | 0.5 ± 1.4 | 0.0 | 0 to 8.5 | 29.4 ± 8.0 | 28.5 | 19 to 55.5 | 31.6 ± 9.6 | 32.0 | 10 to 53.5 |
| India household 1 (1-week) | 29.0 ± 13.2 | 27.8 | 5.9 to 144.0 | 4.0 ± 2.7 | 3.5 | 0 to 41 | 32.2 ± 6.7 | 31.9 | 20.4 to 44.8 | 37.4 ± 25.7 | 32.8 | 0 to 91.7 |
| India household 2 (1-week) | 53.2 ± 222.8 | 36.2 | 11.7 to 4 378.0 | 2.8 ± 1.6 | 2.5 | 0 to 10 | | | | | | |
| India household 3 (1-week) | 37.9 ± 15.2 | 37.2 | 14.6 to 540.9 | 0.3 ± 2.6 | 0.0 | 0 to 40.5 | 34.3 ± 6.1 | 33.4 | 23.6 to 46.9 | 37.4 ± 22.2 | 33.6 | 3.9 to 85.9 |
| India household 4 (1-week) | - | - | - | 2.3 ± 2.0 | 2.0 | 0 to 10 | | | | | | |

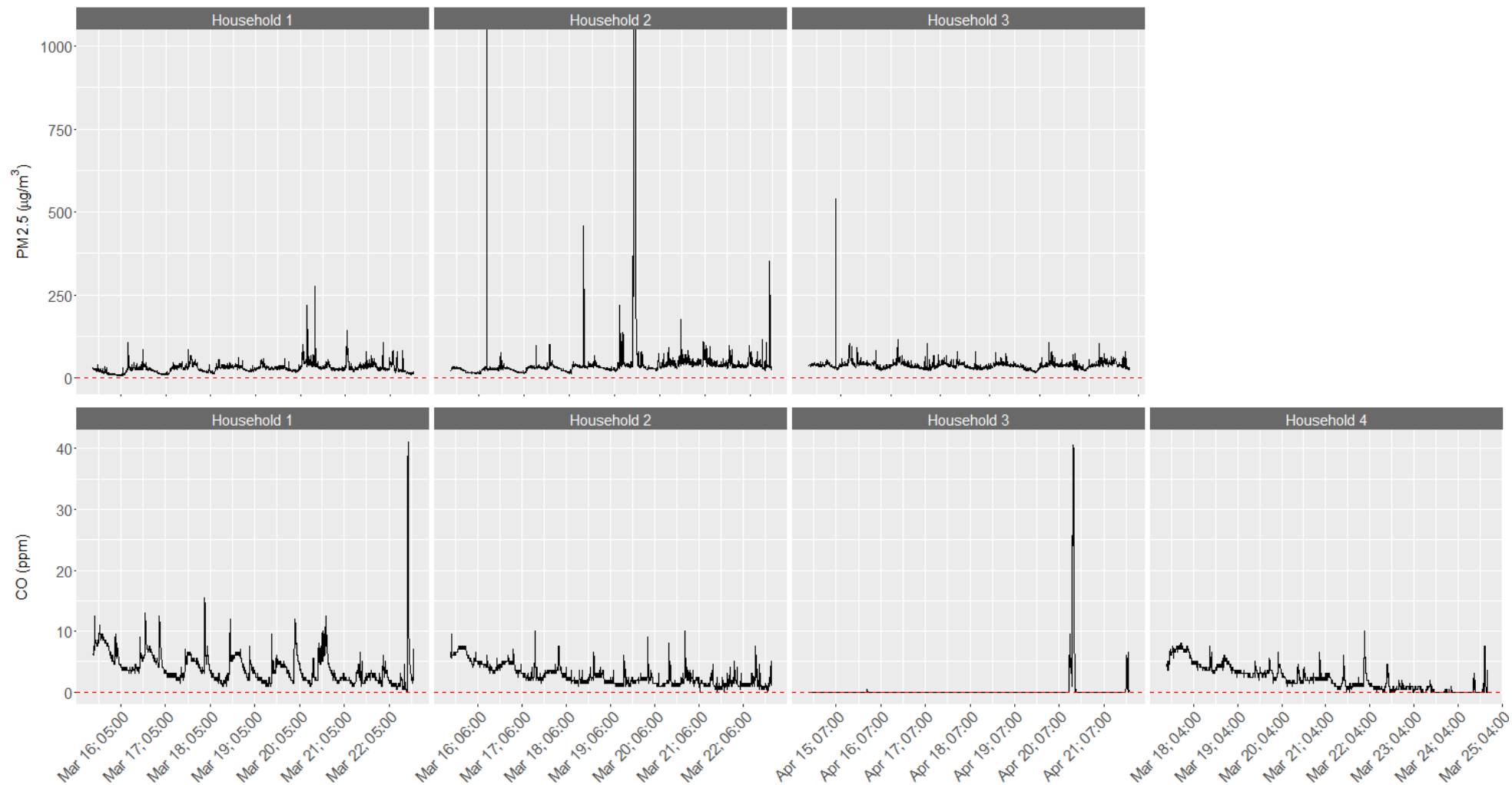
2 *PM_{2.5}: particles less than 2.5 µm; CO: Carbon Monoxide. PM_{2.5} was not measured in household 4 due to lack of device availability. PM_{2.5} data presented here were adjusted*

3 *gravimetrically and by relative humidity. Note that CO levels in Spain are shown for one of the three units used; levels for the other units for the same 3 days were similar (all medians*

- 1 were 0, means ranged from 0.3 to 1.3 ppm and maximum levels from 7.2 to 10.3 ppm). Temperature and Relative Humidity were measured indoors in the case of Spain and outdoors
- 2 in the case of India. Temperature and Humidity levels were the same for household 1 and 2 because the sampling was conducted the same week; the same for households 3 and 4.

1

Figure 5 1-week PM_{2.5} (measured by HAPEX) and simultaneous CO (measured by EL-USB-CO) levels (5min averaged) in households from southern India.



2

1 *PM_{2.5}: particles less than 2.5 μm; CO: Carbon Monoxide. Dashed red line is showing the 0 level. PM_{2.5} values were adjusted gravimetrically and by relative humidity. Only household*

2 *1 and 3 were measured with the same EL-USB-CO unit.*

3

4