

A methodology for the characterization of portable sensors for air quality measure with the goal of deployment in citizen science

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1	A methodology for the characterization of portable sensors for air
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3	
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16 Abstract

17 The field of small air quality sensors is of growing interest within the scientific community, 18 especially because this new technology is liable to improve air pollutant monitoring as well as be 19 used for personal exposure quantification. Amongst the myriad existing devices, the 20 performances are highly variable; this is why the sensors must be rigorously assessed before 21 deployment, according to the intended use. This study is included in the Polluscope project; its 22 purpose is to quantify personal exposure to air pollutants by using portable sensors. This paper 23 designs and applies a methodology for the evaluation of portable air quality sensors to eight 24 devices measuring PM, BC, NO₂ and O₃. The dedicated testing protocol includes static ambient 25 air measurements compared with reference instruments, controlled chamber and mobility tests, as 26 well as reproducibility evaluation. Three sensors (AE51, Cairclip and Canarin) were retained to 27 be used for the field campaigns. The reliability of their performances were robustly quantified by

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28	using several metrics. These three devices (for a total of 36 units) were deployed to be worn by
29	volunteers for a week. The results show the ability of sensors to discriminate between different
30	environments (i.e., cooking, commuting or in an office). This work demonstrates, first, the ability
31	of the three selected sensors to deliver data reliable enough to enable personal exposure
32	estimations, and second, the robustness of this testing methodology.

Keywords: Paris region; Black carbon; Nitrogen oxides; Particulate matter; Personal exposure,
Mobile measurements.

38 **1 INTRODUCTION**

Atmospheric pollution is a well-identified threat to health (WHO, 2003; IARC, 2013; Sante Publique France, 2016; European Environment Agency, 2017). WHO (2014) indicates that 94% of world population is exposed to levels of air pollution that are hazardous. This highlights that monitoring pollution accurately is very important to understand better the phenomenon and to suggest solutions for its mitigation.

44 So far, monitoring networks do not enable the precise measurement of personal exposure to 45 pollution, defined as the pollutant concentration inhaled by people over a period of time. The first 46 reason is the interpolation between stations. Even with a great number of stations, interpolation is 47 still required to quantify personal exposure, and that leads to errors. Second, the air sampling 48 height (set by the legislation between 1.5 and 3 meters above ground level) is often above the 49 average height of our respiratory system, which also induces a difference with the inhaled air. 50 Furthermore, monitoring stations maps and daily reports are only based on outdoor 51 measurements, but people spend most of their time indoors (Klepeis et al., 2001), where high 52 concentrations of air pollutants may exist (Adgate et al., 2004). This is why it is of primary 53 importance to measure also indoor concentrations and to take into account the different 54 environments where people live to quantify their personal exposure.

The solution to quantifying personal exposure could be portable air pollutant sensors. The field of small sensors is constantly improving (Borghi *et al.*, 2017) thanks to the progress of available technologies. This type of sensor presents two main advantages over classical measurements. First, the sensor units are small and thus easily worn all day long by people, which could enable robust 24/7 personal exposure measurements (including indoor air measurements). Second, some of these sensor units are relatively low cost, which allows large numbers of units to be purchasedand enables simultaneous monitoring of a large number of places.

Conversely, the main drawback of these devices is their questionable accuracy. In the field of small air quality sensors, several kinds of studies were published, such as sensor development (Hu *et al.*, 2016; Mead et al., 2013; Peng *et al.*, 2013), sensor assessment (Lin *et al.*, 2015; Burkart *et al.*, 2010; Sousan *et al.*, 2016), exploratory measures of personal exposure (Velasco *et al.*, 2016; Velasco and Tan, 2016; Hu *et al.*, 2014) and full-scale project involving large field campaigns (Mead *et al.*, 2013; Castell *et al.*, 2015, 2017; Schneider *et al.*, 2017; Hasenfratz *et al.*, 2015).

69 Although previous projects were interesting, some limitations invite researchers to keep on 70 improving the methods to quantify personal exposure. First, it is highlighted that most of the 71 small sensors suffer from lower precision than reference instruments, which shows the 72 importance of a robust assessment prior to launching field campaigns. The report by Lewis et al. 73 (2018) (not yet published during the experiments presented below) gave the state of the art of the 74 low-cost air quality sensors. However, not all studies pushed the sensor characterization as far as 75 it should be. Second, personal exposure is about measuring air quality as close as possible to the 76 inhaled air, which is why it is of primary importance to give the sensor units directly to people. 77 But some studies used sensor units on static measurements or attached to vehicles (Velasco and 78 Tan, 2016; Castell et al., 2017; Deville Cavellin et al., 2016; Duvall et al., 2016; Fishbain et al., 79 2017; Gao et al., 2015; Holstius et al., 2014), which is not as relevant as asking people to 80 personally carry the sensor units. Finally, personal exposure is relevant in health impact studies 81 for which it is interesting to have several measured pollutants. This last consideration points out 82 single pollutant measurement studies as a limitation.

⁸³ Île-de-France is the Paris region, it is the most densely populated in France but with great ⁸⁴ disparities between Paris centre and remote places. This region is also characterized as an ⁸⁵ important atmospheric pollutant emission area. More than one million inhabitants are exposed to ⁸⁶ nitrogen dioxide concentration exceeding the *limit value*¹ (annual mean of 40 µg m⁻³) and 85 % ⁸⁷ of the population is exposed to PM_{2.5} levels above the *long-term objective*¹ (annual mean of ⁸⁸ 10 µg m⁻³) (Airparif, 2017, 2018, 2019).

The project Polluscope funded by the French National Research Agency addresses precisely the previously mentioned issues by asking volunteers to carry portable geolocalised sensor units all day long during one week in order to quantify personal exposure to several pollutants. This project is characterized by multidisciplinary objectives (large field campaigns with many volunteers, a cloud platform for data processing, a big data analysis, an epidemiological study, a deep data processing, etc.), but the first step was to conduct a robust sensor selection and assessment.

Polluscope is also defined by the diversity of the studied pollutants as this project will monitor PM (PM_{10} , $PM_{2.5}$ and PM_1), black carbon (BC) and nitrogen dioxide (NO_2). The choice of the monitored pollutants was made according to their impact on health as well as the exceedances experienced in Île-de-France for each pollutant. Indeed, Airparif, the French agency for air quality monitoring, states that some pollutants (PM_{10} , $PM_{2.5}$, NO_2 , O_3 , benzene) still exceed the limits. As these pollutants (as well as BC, which is not regulated) have a positive deleterious

¹ Definitions of *limit value* and *long-term objective* are deeply detailed in Directive 2008/50/EC of the European

Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe

102	impact on health (Schwartz et al., 1996; WHO, 2012, 2003; Liu and Peng, 2018; IARC, 2013;
103	Niranjan and Thakur, 2017; Janssen et al., 2011) they are of great interest for the project.
104	Within Polluscope, the purposes of this paper consist of:
105	•Selecting the sensors in accordance with the project expectations (measurement
106	performance, size, price, etc.); and
107	•Assessing the selected sensors to determine precisely their ability to be used in the

2 METHODS 109

110 2.1 Sensor selection protocol

project.

111 2.1.1 Pre-selection

112 Among the main expected sensors specifications (listed in Table 1, measurement ranges were set in accordance with the maximum measured hourly levels in Île-de-France by Airparif from 113 1990 to 2017 (O₃: 170 ppb, NO₂: 270 ppb and PM₁₀: 660 μ g m⁻³). BC hourly mean values up to 114 115 tens of micrograms per cubic meter were measured close to major traffic axes. As one of the 116 objectives of this work is to monitor the exposure in different environments (inside, outside, in a 117 car, etc.) throughout the day, a fine time resolution (below 5 min) was needed. Furthermore, some 118 practical requirements were added due to the need to have sensors able to measure all day long, 119 whatever the season, and which are easy to carry every day of the week. This is why a long 120 enough battery life was necessary, as well as being light weight and having a large working 121 temperature range. In addition to these fundamental specifications, the sensors had to present a 122 detection limit, a precision and an accuracy in accordance with the project goals. Moreover, close attention had to be paid to other possible issues, such as interference from other pollutants, long-term drift, lack of data storage capacity, etc.

Tens of sensors measuring the pollutants of interest existed, but very few matched our expectations. First, the sensor units must be commercially available; nevertheless, beta versions of sensor units were considered in order to give a chance to almost commercialized units. Furthermore, the considered units had to be built for mobile measurements.

According to these specifications, a pre-selection was made, mostly based on a bibliographic
survey. About fifty articles were studied; they are listed in Table S1 in the appendix.

At the end of the pre-selection stage, eight sensors fulfilling or mostly fulfilling the requirements were chosen to be tested. Unfortunately, some devices gave aberrant values, this is why no VOC results are presented in this work even if it was a pollutant of interest.

134 **2.1.2 Testing strategy**

135 The first step consisted of testing the sensors in static measurements, for up to 15 days, in 136 ambient air, close to fixed reference instruments. These measurements took place at Site 137 instrumental de recherche par télédétection atmospherique (SIRTA²) which is an atmospheric observatory belonging to the research insfrastructure ACTRIS³. For particulate matter, the 138 139 reference instruments were Fidas (Palas), TEOM 1405F for PM₁₀ (Thermo Scientific), TEOM 140 1400 for PM_{2.5} and PM₁ (Thermo Electron) and Aethalometer AE33-7 for BC (Magee Scientific). 141 For gas monitoring, the reference instruments were T200UP for nitric oxides and T400 for ozone 142 (Teledyne). The time step was 1 minute for every instrument, except for TEOM, which was 15

² http://sirta.ipsl.fr

³ https://www.actris.eu/

143 minutes. The reference instruments' inlets were situated on the roof of the building 144 (approximately 4 meters above ground level). Sensor units were fastened under a steel shelter 145 (next to the reference instruments building) in order to be protected from rain; they sampled the 146 air at around 2.5 meters above ground level. The units were powered all day long. This first step 147 consisting of static measurements compared with reference instruments aimed to be a first 148 assessment of sensor accuracy. For this kind of test, sensors are usually assessed compared with 149 reference instruments by using root mean squared error (RMSE) and Pearson correlation R^2 . 150 However, these two indicators do not totally assess the capability of the sensor units to be used in 151 field campaigns. First, RMSE is too sensitive to brief discrepancies between the reference 152 instrument and the sensor. For instance, if two time series are very similar most of the time, but a 153 sudden and significant difference occurs, the RMSE could be large. The Pearson correlation is a 154 good indicator, but only for the linear domain. However, for extreme values, sensors could 155 measure outside their linear domain. Therefore, other correlation coefficients dealing with non-156 linear domains are needed. This is why a tool developed by Fishbain et al. (2017) that 157 quantitatively assesses the sensors was used. The algorithm is called SET for sensor evaluation 158 toolbox, and as it is well described in the paper, only a brief explanation is given here. The SET 159 adds six parameters to the two above-mentioned (RMSE and Pearson correlation): the Kendall 160 and Spearman correlation (assessing the non-linear correlation), the presence (quantifying the 161 data loss), the source analysis (not used in the present work because not relevant in this context), 162 the match score (a metric assessing the ranking order similarities) and the lower frequencies 163 energy (LFE, estimating the sensor ability to properly capture the signal variability). Then, an 164 integrated performance index (IPI) is calculated from the eight parameters; this index goes from 0 165 to 1 and the higher the IPI, the better is the sensor.

166 The second step consisted of mobility/reproducibility tests. These tests were done to assess the 167 ability of the sensors to measure not only in static positions but also in motion. To assess the 168 reproducibility of the measurements, a group of three persons were equipped, each with one unit 169 of the selected sensors. The measurements lasted one working day and took place by following a 170 route previously set, as done in scripted exposure studies (Jarjour et al., 2013). The itinerary was 171 chosen to pass through different environments (public bus, tramway, metro, office and restaurant) 172 in downtown and suburbs of Paris. The route also went close to Airparif monitoring stations 173 (www.airparif.fr), so as to get some "reference points" throughout the day. This itinerary is 174 plotted in Fig. 1.

175 2.2 Selected sensors assessment

176 Once the selection tests were over and the sensors were selected, it was of primary importance 177 to conduct a robust assessment of these instruments to be used in the field campaigns. The 178 different steps of this assessment are described below.

179 2.2

2.2.1 Reproducibility tests

Fifteen sensor units were purchased for the "real" campaigns, all of them had to measure in the same way, which is why reproducibility, accuracy and precision had to be properly checked. From June 26th to July 2nd 2018, all fifteen units of each selected sensor were tested simultaneously in static measurements co-located with reference instruments just like during the selection step.

185 **2.2.2 Controlled chamber tests**

The second step's goal was to assess the sensors' sensitivity to humidity, reactivity and interferences to pollutant level changes as well as the sensors' signal to noise ratio under constant conditions. These tests were conducted in a controlled chamber where the humidity is controlled

189 (from approximately 40 % to 80 % relative humidity, the entire cycle lasted one hour, two cycles 190 were conducted) as well as NO₂ concentration (from 0 to 200 ppb). These are the typical orders 191 of magnitude the sensors will have to deal with due to environmental changes during the coming 192 campaigns. NO₂ variation is of course useful to characterize sensors measuring NO₂, but this test 193 was also relevant for other gas sensors, as some of these devices can suffer from NO₂ 194 interference.

195 **2.2.3 Feasibility campaign**

Finally, a feasibility campaign was performed, which consisted of a rehearsal or a practice for the coming "real" campaigns. In other words, the selected sensor units were worn by collaborators all day long for a week, in the exact same conditions that would occur during the real campaigns. The goal here was to check for any issues of any kind (battery life, inlet sampling, data flow, data processing, etc.) This exercise also allowed us to validate the final campaign protocol.

202 **2.2.4 Final sensor selection**

The authors have decided to anonymize the non-selected sensors in order not to discredit devices that did not give good results during our experiments but which may have been improved since then. The final selected sensors are presented in detail in this work: the AE51, the Cairclip and the Canarin; below called, respectively, sensor A, sensor C and sensor F (technical details about measurement principle are shown in appendix A).

208 **3 RESULTS**

Eight sensors were tested during the selection tests. The main specifications are listed inTable 2.

211 **3.1 Static ambient air tests**

212 For static ambient air tests the results of six of them are presented (the sensors G and F showed 213 aberrant values). An example of the time series is given for the sensors A, B and C in Fig. 2 and 214 gives a preliminary assessment of the sensors' reliability. As shown in Fig. 2, the results from 215 several days of continuous measurements of sensors versus reference instruments are 216 heterogeneous among the different sensors and thus difficult to assess. The first time series 217 exhibits a BC sensor (black line), which gave results very close to the reference instrument (grey 218 line). For this sensor, the results were satisfying: the two lines are almost always overlapping. 219 This first basic tool (studying the time series) identified sensor A as being in agreement with the 220 project expectations.

However, the results were not always that unambiguous, and some sensors gave medium results like the nitrogen dioxide sensors presented in the second time series (Fig. 2). For these two devices, it is difficult to assess the performance of the sensors by only using the time series. Furthermore, the difference with the reference instrument and the correlation are not the only characteristics to focus on, but also the medium term shifting, the lack of data, dynamics, etc. are important. This is why the SET tool (presented in the methods section) is relevant.

In Table 3, the integrated performance index (IPI) and the other SET results are presented for 227 228 the six sensors used in this work. The measurements time bases and the dates of the considered 229 period of time are also given here. BC measurements were done only by sensor A. The satisfying 230 performance of this sensor demonstrated with the time series is corroborated by the SET 231 evaluation, with a very good IPI of 0.91, which is due to the high results for every single 232 parameter. Ozone was only measured by sensor B. For this pollutant, the IPI is mediocre with a 233 value of 0.46. This is explained by the non-negligible data loss: the presence is 0.75, which 234 means that one value out of four is missing. Furthermore, the RMSE is high (15 ppb) compared 235 with the mean value of 8 ppb, and even the match score (0.3) is poor. Three sensors measured

236 NO₂. The best one was sensor B, with an IPI of 0.76 and a RMSE of 5 ppb. Sensor C gave poor 237 results and even aberrant values highlighted by very low correlation coefficients (below 0.15). 238 Sensor D has a fair correlation coefficient (higher than 0.5) but suffers from a large RMSE 239 (37 ppb), poor match score (0.24) and quite significant data loss (presence of 67 %). Particulate 240 matter was measured by three sensors: sensor B measured PM₁₀ and the sensors E and F 241 measured PM₁₀, PM_{2.5} and PM₁. Sensor F gave the best results for all the PM sizes, with an IPI of 242 0.64, 0.80 and 0.78 for PM₁₀, PM_{2.5} and PM₁, respectively. The others gave a much lower IPI. 243 The major advantage of sensor F is the data availability, which does not suffer from data loss. Its 244 match score is acceptable for the three PM sizes (always larger than 0.6) although this parameter 245 is lower than 0.43 for the others. Even if its RMSE is large, sensor F gives a relevant 246 approximation of the PM concentration.

247 This first static ambient test led us to rule out sensor E, which gave aberrant values for PM₁₀, as well as sensor D because of its non-satisfactory results. Sensor B was kept despite the 248 249 mediocre results for O₃ (IPI of 0.46) and PM₁₀ (IPI of 0.40), thanks to its multi-pollutant measuring ability and because the producing firm should improve the sensor before the next 250 251 testing step. Sensor C gave here poor results, but the authors were aware that good results had 252 been obtained with this device, and it was suggested that these unsatisfactory results could be due 253 to an out-of-date electrochemical sensing cell or an inappropriate storage, which would lower the 254 sensor performance. This is why new units of this sensor were purchased for the following testing 255 steps.

256 **3.2 Mobility/reproducibility test**

This test involved the following sensors: sensor B, sensor C and sensor F. The BC sensor A was not involved in this mobility tests because studies (Ezani *et al.*, 2018; Lin *et al.*, 2017) have already demonstrated its ability to perform mobile measurements. Fig. 3 and Fig. 4 (and Fig. S1 260 in Appendix) show, for the three units of each sensor, the entire time series for every sensor of 261 each pollutant and the comparison with the Airparif stations when the route goes by these stations. 262 Fig. 3 refers to the NO₂ sensors. The whole-day time series shows that sensors C present a better 263 reproducibility between the units than sensors B. Secondly, the sensors C show a better dynamic 264 response whereas sensors B present averaged values. Moreover, this figure demonstrates the 265 sensitivity of the sensors C to monitor the environmental changes. Three specific environments 266 are pointed out in the time series: "Opéra" is an Airparif monitoring station classified as traffic 267 influenced, "Restaurant" refers to the lunch break, which took place in a cafeteria and "Bus" 268 stands for bus travel. Sensor C was able to identify different levels associated with different 269 environments: NO₂ was high (around 50 ppb) during the time spent close to the Opéra traffic 270 location, low (about 10 ppb) in the restaurant (indoors) and presented strong variations during bus 271 travel. Furthermore, these sensors quickly detected the environment changes. On the contrary, 272 sensors B were very slow to monitor pollutant level variations, and were unable to properly 273 discriminate environmental changes. Below the main time series, graphs allow us to estimate the 274 accuracy of the sensors against Airparif stations, which can be considered as reference 275 measurements. Except for the "Paris centre" station, the three sensors C were in the right range; 276 the variations were also well monitored, especially for the "Célestins" and "Opéra" stations. The 277 sensors B never showed clear variation, and a significant difference existed between the three 278 units (up to tens of ppb for the "RN2" station). Fig. 4 shows the mobility test results for PM_{10} 279 sensors B and F. The time series depict poor consistency between the units, especially for sensor B, for which a gap of up to $50 \ \mu g \ m^{-3}$ was observed. The comparison with the Airparif stations 280 281 shows that sensors B gave poor results: for the stations "Paris centre" and "Bobigny", the sensor 282 values were significantly different from the reference; for the "Opéra" station, results were fair (a shift of about 10 μ g m⁻³ appears); and for the "RN2" station, the difference between the three 283 units was substantial (about 30 µg m⁻³). Results were better for the sensors F: the sensors were 284

almost always in the range of the reference station, with an inter-sensor difference not larger than 20 μ g m⁻³. Nevertheless, sensors F results were poor for the "Bobigny" station, for which the difference with the reference went up to 30 μ g m⁻³. Three units of each sensor (B, C and F) were tested during this mobility steps. Sensors C and F presented the best results.

289 **3.3 Selected sensors**

The BC sensor A gave satisfying results and was therefore selected for the next step. The sensors C gave suitable results during the mobility tests, which confirms the hypothesis that a deteriorated unit was used during the static measurement tests. This sensor gave better results than sensor B for NO₂. The PM sensor F gave more accurate results than sensor B. For this reason and because of the poor results for NO₂ and O₃, sensor B was excluded.

The retained sensors were sensors A (BC), C (NO₂) and F (PM_{10} , $PM_{2.5}$ and PM_1). The next section deals with the assessment of their capabilities. Several units of each of the three sensors were purchased: 15 units for the sensors C and F, and 6 units for the sensor A (due to its high cost).

299 **3.4 Reproducibility tests in static measurements**

300 An overview of the results is presented in Fig. 5. From the top to the bottom, are presented the 301 BC, NO₂ and PM_{2.5} measurements. The reference instrument is plotted in black and the sensors in 302 coloured lines. Generally, the sensors closely followed the reference instrument trend even if 303 some discrepancies were observed. The BC sensors A were very accurate, despite some noisy 304 periods. On the whole, the NO₂ sensors C overestimated the concentration, this is certainly due to 305 the low NO₂ ambient concentration compared with the limit of detection (20 ppb according to the 306 manufacturer's specifications). This behaviour was already observed by Duvall et al. (2016). 307 Even if these sensors were still able to monitor the global variability in these conditions, another 308 measurement campaign was conducted. The objective was to submit the sensors to NO₂ ambient

309 levels higher than the devices' detection limit. From August 28th to September 4th, fifteen NO₂ 310 sensors C were used to measure conditions at the Airparif station close to a major road (Paris ring 311 road). The results overview is presented in Appendix (Fig. S2), the rest of this work is based on 312 the results from this experiment. Concerning the PM sensors F, they were both quite close to the 313 reference and very reproducible to one another, except for one unit plotted in light green, which 314 presented erratic values. For the PM_{2.5}, the mean RMSE is $6 \mu g m^{-3}$, which is fairly low 315 compared with the measured concentration in mobility in Paris and its suburbs (often higher than 316 $40 \,\mu g \, m^{-3}$).

The SET results for the BC sensors are presented in details in Table S2 in appendix. The IPI is high (around 0.8) for all the sensor units. These sensors did not suffer from data loss at all (the presence parameter is always almost 100 %). The reproducibility between the units can be quantified by the measuring range, defined as the average of the difference across the units between the maximum and the minimum for each measurement date. For these BC sensor units, the measurement range is 616 ng m⁻³, which is not negligible but below the mean concentration value.

The SET results for the fifteen NO_2 sensor units (Table S3 in appendix) are homogeneous, the IPI spans from 0.75 to 0.79. The mean measured concentration was above 40 ppb, which is higher than the limit of detection. This leads to good correlation coefficients (above 0.76 for the mean Pearson coefficient) and reasonable RMSE compared with the ambient levels (mean RMSE is 14 ppb). Moreover, data loss was very uncommon as shown by the presence parameter.

The PM SET results are presented in Tables S4, S5 and S6 in appendix for the three PM sizes. Regarding PM_{2.5} sensor units, the IPI is above 0.68, except for the sensor unit F7, which was defective (plotted in light blue in Fig. 5). The presence is higher than 0.72 (except for F6 at 0.62): the sensor units F data loss was low. The correlation coefficients were never below 0.6, which shows good agreement with the reference. The mean measuring ranges for the three classes of PM are $16 \mu g m^{-3}$, $19 \mu g m^{-3}$ and $20 \mu g m^{-3}$ for PM₁, PM_{2.5} and PM₁₀, respectively. These measuring ranges are not negligible but the mean RMSE values are lower: $4 \mu g m^{-3}$, $6 \mu g m^{-3}$ and $14 \mu g m^{-3}$ for PM₁, PM_{2.5} and PM₁₀, respectively. This means that a discrepancy existed between the units, but the general agreement with the true value is acceptable.

338 **3.5 Chamber controlled test**

Fig. 6 shows the results of this test. The top chart represents the controlled parameters
monitored: humidity and NO₂ concentration. The three others are the sensors results.

341 The sensor F values were always zero: it was not affected at all by humidity changes (at least 342 in the absence of PM). The BC sensor A was clearly affected by humidity variations as it showed 343 BC concentration variations at the same time as humidity ones. When the humidity was decreasing, a positive artefact of up to 250 ng m⁻³ was observed. Conversely, a negative artefact 344 of 150 ng m^{-3} was reached when the humidity decreased to 40 $\%\,$ RH. The mean value and the 345 346 standard deviation of this sensor over the constant humidity period were respectively 15 ng m⁻³ and 14 ng m⁻³, which is low compared with ambient levels. Two NO₂ sensor units C were 347 348 measuring in the chamber. They presented a sensitivity to humidity changes with the same 349 pattern as the BC sensor. The positive artefacts went up to 66 ppb and the negative artefact was 350 constrained at zero (there is no negative value). We have also to note that the two sensor units 351 gave results close to one another, with a RMSE of 7 ppb. Over constant humidity and a NO_2 352 concentration of 25 ppb, their mean values were 21 ppb and 26 ppb with a standard deviation 353 around 1 ppb.

The second test conducted in the controlled chamber was the NO_2 variation (Fig. S3 in Appendix). This experiment consisted of a succession of one-hour steps of 0 ppb, 50 ppb,

100 ppb, 200 ppb, 0 ppb and a final longer 50 ppb stage over several hours. Inside the chamber,
the relative humidity was set at 60 % and there is no particulate matter.

The ability of the sensors to monitor quick concentration changes was demonstrated here as the two sensor units reacted at the same time as the reference instrument. The sensor units were able to monitor increases (up to 200 ppb) and decreases down to 0 ppb. However, a gap can be observed between the sensor units and the reference, the associated RMSE is 11 ppb and 15 ppb. For the final longer step at 50 ppb, the RMSE stood at 9 ppb and 2 ppb.

To conclude for the controlled chamber tests, although sensor F presented no artefacts due to humidity changes (with a zero concentration of PM), the sensors A and C were sensitive to humidity. During the following campaigns, this will have to be taken into account, a correction or an invalidation protocol may be needed.

367 **3.6 Feasibility campaign**

The feasibility campaign was conducted with fifteen volunteers from Monday 18th to Friday 22nd June 2018. One sensor unit C and one sensor unit F were given to each volunteer and six sensor units A were shared between the participants. This campaign was conceived as a proof of the Polluscope concept, thus, only a limited analysis of the results was done.

Globally, the campaign was a success: all the sensor units were worn for the whole week. The data availability (the time resolution was one minute) is 66 % for sensors A and 69 % for sensors F, which can be considered as a satisfying result. The data loss was due to minor problems and routine maintenance (filter change, turning on and off, powering, etc.). However, the data availability for the sensors C only reaches 41 %. This was caused by storage memory erasure when the sensor ran out of power. The coming campaigns protocols will prevent this issue. Generally, the data availability was slightly lower than during the previous tests, this was due to

the campaign environment and the fact that the sensor units were operated by volunteers withoutexpert skills.

381 Fig. 7 shows the results of the three sensor units worn for the whole week by a volunteer. Four 382 kinds of environments are pointed out: "indoor" for the time spent inside, "polluted indoor" for emitting activities conducted indoors (cafeteria, smoking or cooking, for instance), "commuting" 383 384 journeys (whatever the travel mode) and "outdoor" for the time spent outside any building. The 385 indoor environment is the more frequent environment, nevertheless, the spikes were usually 386 observed during commuting or in polluted indoor environments. The major BC and NO₂ peaks 387 occurred most frequently during commuting trips. Inversely, the highest particulate values were 388 measured during "polluted indoor" episodes. An example of contrasted environments 389 (commuting, indoors and tobacco smoke in indoor environment) measurements is presented in 390 Appendix, Fig. S4. During the campaign, artefacts due to quick environmental change (studied in 391 the controlled chamber in Section 3.5) were observed; this is more detailed in Appendix, Fig. S5).

This feasibility campaign demonstrated the capability of the Polluscope protocol to conduct a campaign lasting a whole week with volunteers. The results from the sensors enable us to discriminate several emitting activities; a preliminary estimation of the personal exposure is thus available.

396 4 DISCUSSION

The first stages of Polluscope (the selection and assessment of the sensors) have been conducted. The AE51 (BC), the Cairclip (NO₂) and the Canarin (PM_{10} , $PM_{2.5}$ and PM_1) have been selected and assessed. The feasibility campaign demonstrated that these three sensors are reliable enough to be used for full-scale campaigns involving volunteers from the general public. Their ability to discriminate different environments (commuting trips, polluted or clean indoor environment, etc.) has been proven. 403 For the static measurement assessments, we used the SET algorithm designed by Fishbain et al. 404 (2017), available as an open source resource⁴. In their article, they presented results from 25 405 AQMesh NO₂ sensor units that had taken measurements for about three months in static positions. 406 The mean related IPI is 0.58, this is very close to our mean Cairclip IPI (from the reproducibility 407 test) of 0.54; even if the Cairclip is designed for mobile measurements whereas the AQMesh 408 system is designed for static monitoring (i.e. expecting to have a better performance than a 409 portable device). Knowing the successful deployment of the AQMesh sensor units, this result 410 demonstrates the reliability of the Cairclip. Fishbain et al. (2017) also used the SET with PM 411 sensors (DC1700 Dylos and GeoTech), the mean resulting IPI is 0.63. The PM_{2.5} Canarin sensor 412 used in our study gave significantly better results with a mean IPI of 0.73.

The SET algorithm was also used by Broday and the Citi-Sense Project Collaborators (2017). They presented unpublished results from about three months of ambient air measurements of six PM₁₀ sensor units located in Ostrava, Czech Republic. The mean IPI is 0.72, which is very close to the result from our study (0.73).

Due to its recent release, SET algorithm results have not been published in other articles yet. To the best of our knowledge, our study is the first to apply the SET evaluation to the AE51 and the Cairclip sensors. However, these two devices have been largely used and several results have been published, some of the more relevant for our study are discussed below.

421 4.1 Static comparison

Lin *et al.* (2017) compared AE51 with reference instruments and found a good mean correlation of 0.77. Viana *et al.* (2015) conducted a study involving six AE51 and a reference

⁴ fishbain.net.technion.ac.il

424 station in static measurements, the correlation coefficient was above 0.75. In our study, the mean 425 correlation coefficient was 0.80. This higher agreement may be due to higher inlet flow 426 $(150 \text{ mL min}^{-1} \text{ compared with } 100 \text{ mL min}^{-1} \text{ in the Viana$ *et al.* $} (2015) study) or coarser time$ 427 resolution (1 min in our study and 1 sec in the Lin*et al.*(2017) study).

428 Several other studies have pointed to the good results of this black carbon sensor in agreement 429 with our results (Cai *et al.*, 2014; Gillespie *et al.*, 2017; Velasco and Tan, 2016).

430 The recent low-cost sensors review by the World Metrological Organization (Lewis et al., 431 2018) described several performance evaluation programs as the work supported by the United States Environment Protection Agency (EPA)⁵; they described an air sensor toolbox where the 432 433 main performances of tens of sensors were gathered and compared. For the Cairclip NO₂ sensor, 434 the EPA and Jiao et al. (2016) state that a correlation coefficient between 0.42 and 0.76 was 435 obtained with reference instruments. Our mean Pearson correlation was 0.76, which is in the high 436 part of the EPA range. The additional information given by the SET algorithm in our study is the 437 good match score of 0.64 and the absence of data loss (the presence parameter reaches almost 438 100%). Another example is the study conducted by Spinelle et al. (2015). They found a 439 correlation of up to 0.75 for the Cairclip sensor units. This result is both in the EPA range and 440 close to our result of 0.76.

Due to its new release, only a few research works including Canarin have been conducted. For instance, Tse *et al.*, 2018B presented a project based on static measurements from four Canarin units. Some tests were conducted in Bologna, Italia, and PM_{10} maps have been produced. These works were preliminary and the most accomplished article about Canarin sensor is certainly the one conducted by Tse *et al.*, 2018A. They deployed nine Canarin units in a library inducted at the

⁵https://www.epa.gov/air-sensor-toolbox

446 UNESCO world heritage list. The sensors were measuring 24/7 for months (from Summer 2017 447 to Spring 2018), which enabled to show that a clear diurnal pattern occurred with higher levels 448 during night time. On a longer period of time it was the winter season which experienced more 449 pollution. This protocol also permitted to quantify that 56 % of the time, the PM_{2.5} air pollution level was low according to the EPA standards (below 12 µg m⁻³). The coming improvements 450 451 announced in these three articles suggested a wider use of the Canarin in a near future. Overall, 452 these works underlined the promising capabilities of this sensor. The present paper confirmed this 453 first evaluation and went a step further (larger amount of units, mobile measurements, etc.) to 454 prove the ability to use the Canarin to equip volunteers for the personal exposure quantification.

455 **4.2 Mobile measurements**

It is usually more difficult to robustly assess sensor accuracy in mobile measurements as the reference instruments are unlikely to be usable in motion. A metric that can be used is the agreement between several units of portable sensors (previously assessed – or not – in static measurements versus a reference instrument). This provides information on the reproducibility and thus on the reliability of the mobile device. Another possibility is to compare the sensor measurements with static stations considered as a reference if the mobility route goes close to this kind of monitoring site.

The Ezani *et al.* (2018) study was based on mobile measurements performed with two AE51 units. There were no reference instruments but the correlation between the two units was good: 0.92. Lin *et al.* (2017) performed mobile AE51 measurements. Comparison was possible thanks to 17 transient immobile periods (of less than one hour) nearby reference stations. The AE51 unit showed an interquartile range agreement with the reference instrument of 82 %. High-resolution mapping is possible with the AE51, as in the study of den Bossche *et al.* (2015) where sensor units were mounted on bikes in Antwerp, Belgium. A 50-meter resolution was obtained with an uncertainty of 25 %. Pant *et al.* (2017) performed a study aiming at quantifying personal
exposure to BC in New Delhi, India. The AE51 were given to volunteers and environments
(commuting, cooking, etc.) were distinguished.

473 Few studies have been published on the Cairclip sensor being used in mobile measurements, 474 especially compared with the abundant literature related to the AE51. The recent work by 475 Chambers et al. (2018) found no consistent relationship between NO₂ concentrations and health 476 parameters. The authors state that the Cairclip was able to appropriately monitor personal 477 exposure and a clear diurnal cycle was observed but no more validation data was provided. The 478 study by Reid (2015) is based on the qualification of Cairclip sensors. Mobile measurements 479 were conducted with two Cairclip units in different environments: public transport, outdoor and 480 indoor. The sensors monitored interesting variability, especially close to traffic.

481 Lastly, Aguiari *et al.*, 2018 introduced a possible use of the Canarin by attaching them to bikes.

482 **4.3 Methodology discussion**

483 Our work has revealed that the three selected sensors are appropriate for personal exposure 484 assessment. Beyond that first result, the Polluscope selection and assessment methodology was 485 also an outcome of this study.

486 It is now well known, even in the emerging field of small air quality sensors that a complete 487 sensor assessment is of primary importance to obtain reliable data. Some studies were only based 488 on laboratory experiments (Manikonda et al., 2016; Ng et al., 2018), but in-the-field calibration 489 was identified as necessary to properly assess the sensors' capabilities (Castell et al., 2017; 490 Schneider et al., 2017) as the results can be substantially different from laboratory-controlled 491 environments. For instance, during our tests, the Cairclip showed better results during the 492 controlled chamber tests. The ambient air tests were very useful to reveal that the Cairclip had 493 difficulties in measuring low ambient concentrations.

As seen in the section concerning the reproducibility tests, non-negligible differences were observed between units of the same sensor. This highlights the importance of testing several units at a time. In this study, we conducted tests with 6 AE51, 15 Cairclip and 15 Canarin. For some of the previously published studies, the small number of tested units was a limitation, for example, Lin *et al.* (2017) (two AE51 and only one for the mobility tests), Ezani *et al.* (2018) (two AE51), Duvall *et al.* (2016) (two Cairclip).

500 **4.4 Conclusions**

501 No remote sensor is perfect, and the three selected ones are the result of compromises and each 502 have strengths and weaknesses. The AE51 is accurate, its IPI (above 0.8) is higher than all other 503 sensors. This BC sensor is also reliable and easy to use with very little data loss. But it is 504 sensitive to humidity, which leads to some artefacts when quick environmental changes occur, 505 and its high price is also a weakness because fewer units can be purchased. The Cairclip is very 506 light and thus easy to carry all day long, but the storage memory is erased if the sensor unit runs 507 out of battery, thus it is consequently more demanding for the operator. Moreover, even if these 508 sensors demonstrated their ability to perform reliable measurements in mobile measurement (see related section), the detection limit (20 ppb) is not appropriate for low NO₂ levels. The Canarin is 509 510 able to send data via Wi-Fi and has a high storage capacity (several weeks of measurements), 511 which is useful when the data sending is not possible. Its robustness in mobility is also an 512 important advantage: the Canarin was the sensor that presented the highest data availability 513 during the feasibility campaign (69%). This sensor showed satisfying results for the PM_{2.5} 514 measurements (IPI of 0.7) but substantially lower for PM₁₀ (IPI of 0.4). Finally, the weight of the 515 sensor unit (it is quite heavy) is a drawback.

Even if the three selected sensors have some weaknesses, their ability to be used in mobile measurements has been demonstrated. For the coming campaigns, attention will be given to their drawbacks.

519 To conclude, the Polluscope project is one of the few studies that has conducted an in-depth 520 sensor assessment including the most important following steps:

- Several kinds of tests were performed. Ambient air static measurement tests against
 reference instruments was the first assessment of the sensors in real atmosphere with
 natural meteorological parameters variability (temperature, humidity, wind speed and
 direction, etc.) The laboratory tests were of primary importance to quantify the sensors;
 responses to rapid atmospheric changes (i.e., humidity, pollutants levels). Mobile
 measurements were necessary as the project goal is to use the sensors for personal
 exposure, to be worn by volunteers.
- •A large number of units of each sensor was tested to quantify the reproducibility and to
 eliminate problems arising from a single deficient unit.
- •A robust multi-metric static measurement assessment with the SET algorithm was
 conducted in order to be as rigorous as possible in the assessment.

The next stage of the Polluscope project will be the full-scale campaigns involving fifteen volunteers each week during the six weeks per studied season. These campaigns will take place over two years and will involve 160 people. Even if the size of the project is already consistent, a valuable perspective would be to recruit more volunteers over a larger area in order to increase the representativeness of the study.

537

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Table titles

- **Table 1.** Tested sensor specifications.
- **Table 2.** Main expected specifications of the sensors.

4	Table 3. SET results for every sensor, the units are "ppb" for gases, ng m ⁻³ for BC and μ g m ⁻³ for
5	PM. Pollutant sensor: measured pollutant and sensor's name; M: mean concentration; Match:
6	match score; RMSE: root mean squared error; r: Pearson correlation coefficient; t: Kendall
7	correlation coefficient; S: Spearman correlation coefficient; Pres: presence parameter; LFE: low
8	frequencies energy parameter; IPI: SET integrated performance index; SensTB: sensor time base:
9	RefTB: reference time base; Start time: measurement's beginning; End time: measurement's
10	ending.

Table 1. Tested sensor specifications.

	Expected specifications
	$O_3 = [0;250] ppb$
Measurement range	$NO_2 = [0;500] ppb$
Weasurement range	$BC = [0;50000] \text{ ng m}^{-3}$
	PM_{10} and $PM_{2.5} = [0;1000] \ \mu g \ m^{-3}$
	VOC = depending on the sensor specificity ability
Time step	below 5 min
Battery life	12 hours as a minimum
Temperature range	[-10;40] °C
Weight (total to be worn)	below 2 kg
Detection limit, precision and ac	curacy To be specified by the manufacturer

Sensor	Price (€)	Weight (g)	Measured pollutant
А	5000-10000	200-400	BC
В	2000-5000	600-800	PM_{10} , NO_2 , O_3 , VOC
С	500-1000	0-100	NO_2
D	1000-2000	400–600	NO_2
E	2000-5000	600-800	PM ₁₀ , PM _{2.5} , PM ₁
F	500-1000	600-800	$PM_{10}, PM_{2.5}, PM_1$
G	1000-2000	100-200	VOC
Н	2000-5000	400–600	CO, NO, NO ₂ , O ₃ , PM ₁₀

Table 2. Main expected specifications of the sensors.

Table 3. SET results for every sensor, the units are "ppb" for gases, ng m⁻³ for BC and μ g m⁻³ for PM. Pollutant sensor: measured pollutant and sensor's name; *M*: mean concentration; Match: match score; RMSE: root mean squared error; *r*: Pearson correlation coefficient; *t*: Kendall correlation coefficient; *S*: Spearman correlation coefficient; Pres: presence parameter; LFE: low frequencies energy parameter; IPI: SET integrated performance index; SensTB: sensor time base: RefTB: reference time base; Start time: measurement's beginning; End time: measurement's ending.

Pollutant sensor	М	Match	RMSE	r	t	S	Pres	LFE	IPI	SensTB (min)	RefTB (min)	Start time (TU)	End time (TU)
BC A	1077	0.88	268	0.98	0.85	0.96	0.97	1.00	0.91	5	1	23/01/2017 07:55	06/02/2017 15:05
O ₃ B	8	0.30	15	0.70	0.60	0.80	0.75	1.00	0.46	1	1	23/01/2017 07:43	03/02/2017 08:44
NO ₂ B	11	0.46	5	0.94	0.72	0.89	0.78	1.00	0.76	1	1	23/01/2017 07:43	03/02/2017 08:44
NO ₂ C	20	0.35	13	0.04	0.08	0.12	1.00	1.00	0.42	1	1	20/01/2017 14:00	27/01/2017 10:00
NO ₂ D	47	0.24	37	0.54	0.54	0.73	0.67	1.00	0.56	1	1	23/01/2017 13:56	01/02/2017 10:09
$PM_{10} B$	104	0.43	112	0.18	0.20	0.26	0.81	0.99	0.40	1	15	23/01/2017 07:45	03/02/2017 08:45
$PM_{10} E$	535	0.37	1819	0.06	0.38	0.52	0.69	0.89	0.07	1	15	23/01/2017 08:30	03/02/2017 16:00
$PM_{10} F$	21	0.63	16	0.84	0.33	0.46	1.00	0.99	0.64	1.5	15	26/01/2017 12:30	06/02/2017 15:15
PM _{2.5} E	136	0.43	185	0.45	0.52	0.72	0.69	0.99	0.49	1	15	23/01/2017 08:30	03/02/2017 16:00
PM _{2.5} F	18	0.76	10	0.91	0.66	0.82	1.00	0.99	0.80	1.5	15	26/01/2017 12:30	06/02/2017 15:15
$PM_1 E$	43	0.43	33	0.76	0.62	0.82	0.69	1.00	0.65	1	15	23/01/2017 08:30	03/02/2017 16:00
$PM_1 F$	13	0.77	8	0.88	0.66	0.82	1.00	0.99	0.78	1.5	15	26/01/2017 12:30	06/02/2017 15:15

Figure Captions

2	Fig. 1. Routes for the mobility tests (plotted in red and blue). Sources: actualitix, and
3	OpenStreetMap (modified).
4	Fig. 2. BC and NO ₂ time series in static measurements.
5	Fig. 3. Mobility tests for NO ₂ sensors. The three sensors C are plotted in shades of green, the
6	three sensors B in shades of orange, the reference Airparif stations are plotted in black.
7	Fig. 4. Mobility tests for PM_{10} sensors. The three sensors F are plotted in shades of blue, the three
8	sensors B in shades of orange, the reference Airparif stations are plotted in black.
9	Fig. 5. Reproducibility tests for sensors A, C and F with reference instruments.
10	Fig. 6. Chamber controlled tests for BC sensor A, PM sensor F and NO ₂ sensor C.
11	Fig. 7. Mobility test results.

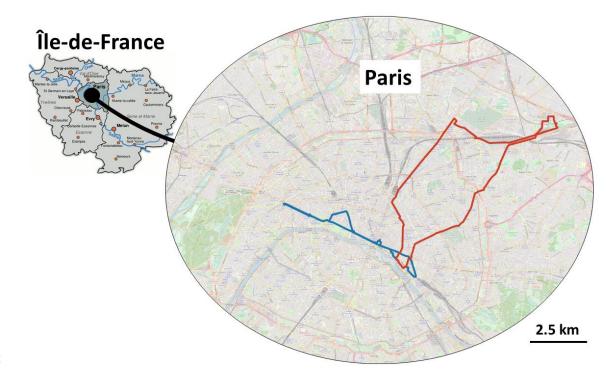
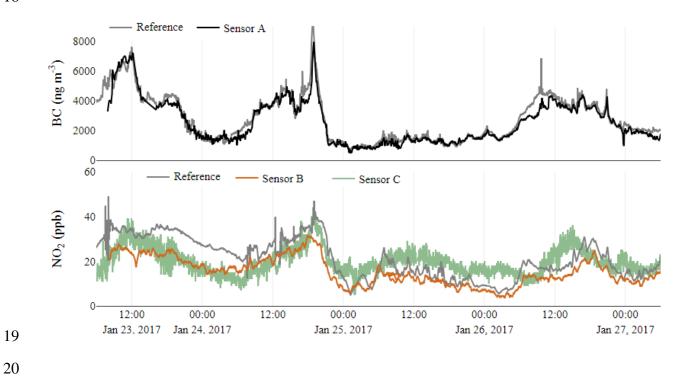


Fig. 1.





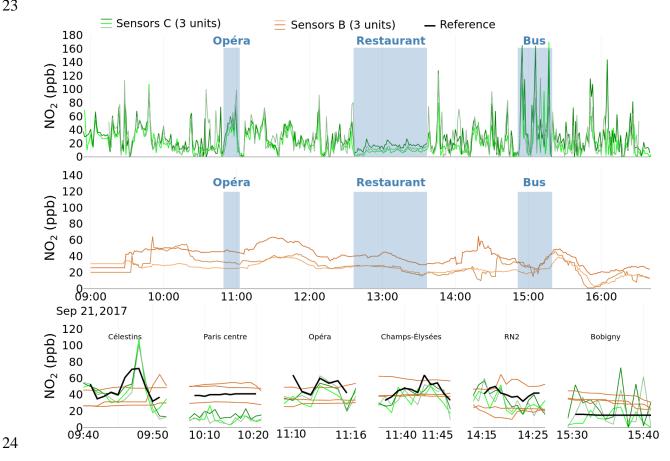


Fig. 3.

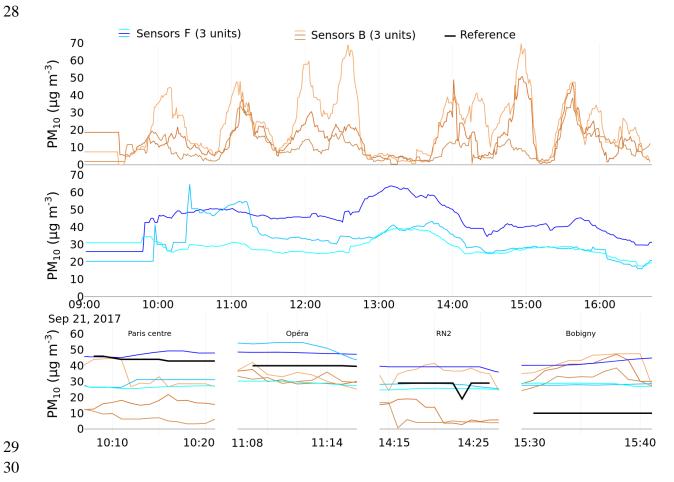




Fig. 4.

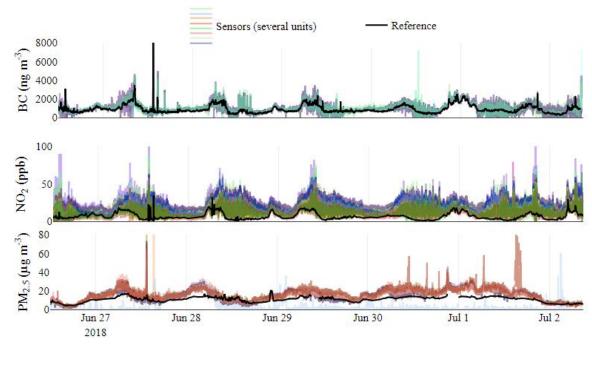
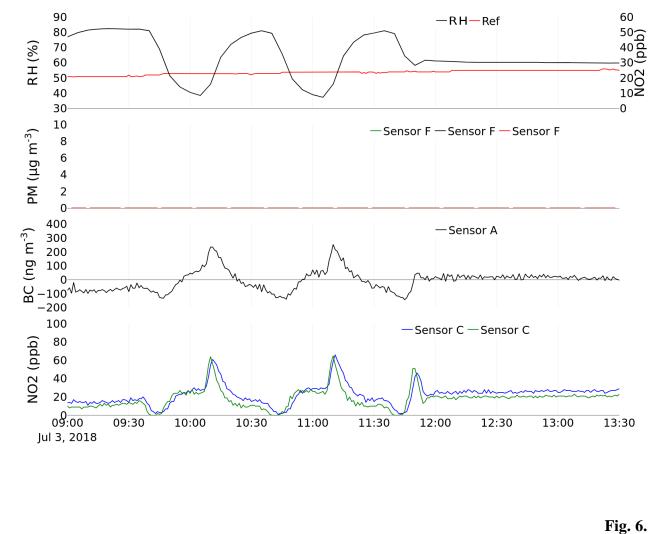


Fig.5.







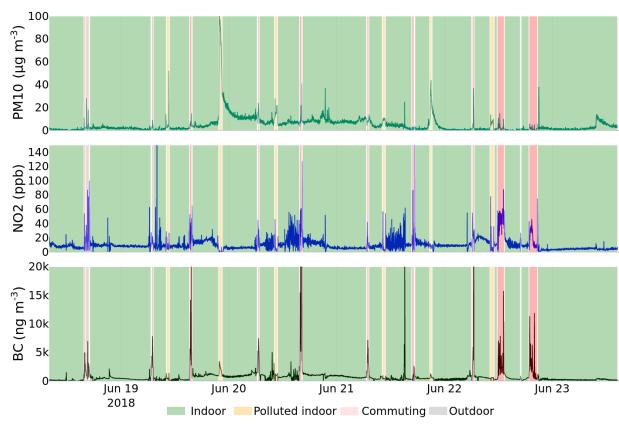


Fig. 7.