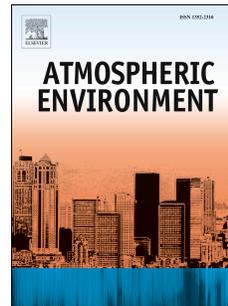


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Assessment of air quality microsensors *versus* reference methods: The EuNetAir Joint Exercise – Part II

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1 Assessment of Air Quality Microsensors Versus Reference Methods: the EuNetAir 2 Joint Exercise – part II

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21 Abstract

22 The EuNetAir Joint Exercise focused on the evaluation and assessment of environmental
23 gaseous, particulate matter (PM) and meteorological microsensors versus standard air quality
24 reference methods through an experimental urban air quality monitoring campaign. This work
25 presents the second part of the results, including evaluation of parameter dependencies,
26 measurement uncertainty of sensors and the use of machine learning approaches to improve the
27 abilities and limitations of sensors. The results confirm that the microsensor platforms,
28 supported by post processing and data modelling tools, have considerable potential in new
29 strategies for air quality control. In terms of pollutants, improved correlations were obtained
30 between sensors and reference methods through calibration with machine learning techniques
31 for CO ($r^2=0.13-0.83$), NO₂ ($r^2=0.24-0.93$), O₃ ($r^2=0.22-0.84$), PM₁₀ ($r^2=0.54-0.83$), PM_{2.5}
32 ($r^2=0.33-0.40$) and SO₂ ($r^2=0.49-0.84$). Additionally, the analysis performed suggests the
33 possibility of compliance with the data quality objectives (DQO) defined by the European Air
34 Quality Directive (2008/50/EC) for indicative measurements.

35
36 *Keywords:* Air quality monitoring; Reference methods; Low-cost microsensors; Experimental campaign;
37 Measurement uncertainty; Machine learning

38 1. Introduction

39 Air pollution is a very significant environmental and social issue. At the same time, it is a
40 complex problem posing multiple challenges in terms of management and mitigation of harmful
41 pollutants. Air pollutants have numerous impacts on health, ecosystems, built environment and
42 climate; they may be transported or formed over long distances, and they may affect large areas.
43 Air pollution continues to affect the health of Europeans, particularly in urban areas. It also has
44 considerable economic impacts; cutting lives short, increasing medical costs and reducing
45 productivity through working days lost across the economy (EEA, 2017; WHO, 2018).
46 According to The World Health Organization (WHO), in 2016, 91% of the world population

47 were living in places where the WHO air quality guidelines limits were not met. Additionally,
48 outdoor air pollution in both cities and rural areas was estimated to cause 4.2 million premature
49 deaths worldwide in 2016 (WHO, 2018).

50 Europe's most problematic pollutants in terms of health are PM, NO₂ and ground-level O₃.
51 In 2015, about 7 % of the EU-28 urban population was exposed to PM_{2.5} levels above the EU's
52 annual limit value. Considering the stricter WHO guidelines, approximately 82 % were exposed
53 to levels exceeding the limit values. Exposure to PM_{2.5} caused the premature death of
54 estimated 428 000 people in 41 European countries in 2014. Regarding NO₂, around 9 % of the
55 EU-28 urban population was exposed to levels above the EU's annual limit value and WHO
56 guidelines in 2015. Exposure to NO₂ caused the premature death of an estimated 78 000 people
57 in 41 European countries in 2014. For O₃ levels, 30 % of the EU-28 urban population was
58 exposed to concentrations above the EU's target value in 2015. Considering the stricter WHO
59 guidelines, approximately 95 % were exposed to levels exceeding the limit value. Exposure to
60 O₃ caused the premature death of an estimated 14400 people in Europe in 2014 (EEA, 2017).

61 A wide range of adverse effects of ambient air pollution on health has been well
62 documented in multiple studies (Pascal et al., 2013; Wu et al. 2016). By working to reduce air
63 pollution levels, countries can lower the burden of stroke, heart disease, lung cancer, and both
64 chronic and acute respiratory illness, with long-term benefits to the population (WHO, 2018).

65 However, there is significant inequality in exposure to air pollution and related health risks:
66 air pollution combines with other aspects of the social and physical environment to create a
67 disproportionate disease burden in less affluent parts of society. The WHO guidelines address
68 all regions of the world and provide uniform targets for air quality that would protect the vast
69 majority of individuals from the adverse effects on health. More than 80% of the population in
70 the WHO European Region (including the European Union, EU) lives in cities with levels of
71 PM exceeding WHO Air Quality Guidelines. Since even at relatively low concentrations the
72 burden of air pollution on health is significant, effective management of air quality that aims to
73 achieve WHO Air Quality Guidelines levels is necessary to reduce these risks to a minimum.
74 Exposure to air pollutants is largely beyond the control of individuals, requiring action by public
75 authorities at the national, regional and international levels. A multisector approach, engaging
76 transport, housing, energy production and industry is needed to develop and effectively
77 implement long-term policies that reduce the risks of air pollution to health (WHO, 2013).

78 The evaluation of the status of air quality (AQ) is based on ambient air measurements, in
79 conjunction with data on anthropogenic emissions and their trends. Holistic solutions must be
80 found that involve technological development, and structural and behavioural changes. Air
81 quality policies have delivered, and continue to deliver, many improvements. However,
82 substantial challenges remain and considerable impacts on human health and on the
83 environment persist (EEA, 2017).

84 The increasing availability of low cost sensors employing various monitoring principles
85 creates the need to identify which are the most appropriate to be further refined and calibrated.
86 For this purpose, it is necessary to estimate the overall performance of a large number of
87 collocated sensors (Kotsev et al., 2016). This not only calls for the application of standard time
88 series analysis and comparison methods, but also the incorporation of overall measurement
89 profile and behaviour, so that a sensor network may generate reliable AQ data. Calibration
90 methods have been based on sensor intercomparison, as well as on overall sensor network
91 calibration (Jiao et al., 2016) and self-calibration (Fishbain and Moreno-Centeno, 2016). Recent
92 studies indicate that machine learning technologies may significantly improve the performance
93 of air quality sensor nodes reducing the impact of cross-sensitivity issues (Spinelle et al., 2015;
94 De Vito et al., 2018). Most of them however, have been carried out on single systems,
95 developed by the same company or research institutions, limiting the understanding and
96 potential about their general applicability.

97 In the first part of this work, the overall results of an intercomparison of AQ microsensors
98 with reference methods during an AQ monitoring campaign in Aveiro, Portugal, were presented
99 (Borrego et al., 2016). The overall performance of the diverse sensors in terms of their statistical
100 metrics and measurement profile indicated significant differences in the results. In terms of
101 pollutants, the following results were observed: O₃ (r²: 0.12-0.77), CO (r²: 0.53-0.87) and NO₂
102 (r²: 0.02-0.89) with some promising results, but equally sensors showing no correlation with the
103 reference method. For PM (r²: 0.07-0.36) and SO₂ (r²: 0.09-0.20) the results showed a poor
104 performance with low correlation coefficients between the reference and microsensor
105 measurements.

106 The purpose of this study is to present the second part of the results of the intercomparison
107 campaign in Aveiro for two weeks in October 2014, complementing the analysis performed by
108 Borrego et al. (2016). More specifically, it is intended to (a) understand parameter
109 dependencies, (b) measurement uncertainty of sensors, (c) the use of machine learning
110 approaches to improve the abilities and limitations of sensors, contributing to their calibration
111 and further development.

112 The paper is organized into the following sections: Section 2 gives a description of the
113 experimental campaign and methodology; Section 3 presents the results obtained with the
114 different data analysis strategies; finally, Section 4 provides the conclusions.

115 2. Experimental Design

116 2.1. Characterization of the study site

117 In this exercise, the AQ microsensor systems were installed side-by-side on the IDAD Air
118 Quality Mobile Laboratory (LabQAr), supplied with standard equipment and reference
119 analysers for CO (Infrared photometry), NO_x (Chemiluminescence), O₃ (Ultraviolet
120 photometry), SO₂ (Ultraviolet fluorescence), particulate matter PM₁₀ / PM_{2.5} (Beta-ray
121 absorption), and meteorological variables (Vaisala WXT520). During the exercise, LabQAr was
122 parked on Avenue Santa Joana, near the Cathedral of Aveiro, in an urban traffic location in
123 Aveiro city centre. The sensors were mainly installed between 2.5 and 3 m above ground on the
124 roof of the mobile laboratory, with the reference meteorological measurements at ~ 5 m on a
125 telescopic mast (Fig. 1).



126
127 **Fig. 1.** Set-up of the AQ mobile station and micro-sensors during the 1st EuNetAir campaign.

128 2.2. Comparison of technical requirements of the sensor nodes

129 Aside from the performance of the sensor nodes with regard to comparability with
130 reference instruments, discussed in Borrego et al. (2016), the selection of a specific sensor node
131 should also consider a number of technical parameters as well as data quality and uncertainty.

132 These include; size, power, connectivity requirements, and number of pollutants monitored
 133 (Table 1).

134 **Table 1.** Technical requirements and features of the sensor nodes evaluated during the Aveiro intercomparison campaign.

Sensor node	AQMesh	SNAQ	Prototype Module	NanoEnvi	ECN airbox	EveryAware SensorBox	OdorCheckerOutdoor	AUTH-ISAG	Prototype Module (with IAQcore)	Air-sensor box
Operated by	IDAEA-CSIC + AQMesh	Cambridge Univ.	UCL/CCS	NILU + Envira	ECN	VITO	3S	AUTH	Siemens AG	ENEA
Parameters measured	NO, NO ₂ , CO, O ₃	NO, NO ₂ , CO, CO ₂ , O ₃ , VOC, PM10	T, RH	CO, NO ₂ , O ₃ , T, RH	PM2.5, PM10, NO ₂	NOx, CO, O ₃ , VOC, gasoline/diesel exhaust fumes, T, RH	T, RH, VOC	T, RH, p, NO ₂ , O ₃	CO/VOC (nonspecific but calibrated to CO)	CO, NO ₂ , O ₃ , SO ₂ , PM10, T, RH
Time resolution (and ability to modify it, Y/N)	15 min (Y)	20 sec (Y)	1 sec (Y)	5 min; Yes (5, 10, 15, 30 and 60 min)	10 min (Y)	1 s (N)	3 min (Y)	5 min (Y)	10 s (Y)	15 min (Y)
Able to operate while connected to power (Y/N)	No; no power required	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
May operate on battery (Y/N, and duration if Yes)	Yes; <24 months	Yes; >1 month	Yes (external batterypack)	Yes; 2 days	Yes	Yes (external batterypack)	No	Yes, <10 hours, increased with extended battery	Yes (<5h, powered by connected notebook)	No
Able to store data internally (Y/N, max duration with default time resolution)	Yes	Yes	No	Yes; >1 year	Yes; one month	Yes; 6 months	Yes, > 2 months	Yes, > 1 year	No	Yes; > 1 year
Online data transmission possible (Y/N)	Yes	Yes	Yes	Yes	Yes	Yes (with smartphone and 'Airprobe' app)	Yes, modem under development	Yes	No	Yes
If online data transmission possible, how? Wifi, SIM card, etc.	SIM card	SIM card	Zigbee optional	SIM card, Ethernet, Zigbee	SIM card	Bluetooth communication with smartphone	SIM w/ GPRS/UMTS	SIM card, Xbee wireless module	---	SIM card, Wi-Fi, Ethernet
Able to be deployed outdoors (waterproof) (Y/N)	Yes	Yes	Yes	Yes	Yes	No	Yes	Yes	No	Yes
Frequency of maintenance required (inlet cleaning, etc.)	No maintenance	No maintenance	No maintenance	Biannual calibration	Yearly	No maintenance	~ 2 months	No maintenance	No maintenance	Yearly
Other features	Standard sun roof	Meteo variables measured as well	Highly modular platform	Solar battery available	Other parameters also possible	Open source hardware and software	Modular setup: other sensors attachable (EC, wind, ...)	Open source + commercial hardware and software	Commercial hardware (IAQcore)	Modular platform with other optional gas sensors

135

136 Among the sensor nodes deployed in this work, the number of parameters monitored
 137 ranged between one and seven, also covering meteorological variables. Time resolution ranged
 138 from 1-second up to 15-minute averages, suggesting different potential applications. For
 139 example, the 1-second time resolution of the EveryAware sensor box is well suited to personal
 140 exposure, while the 15-minute averages produced by the AQMesh and ENEA nodes would be
 141 more representative of ambient pollutant concentrations. However, this does not preclude the
 142 different sensor nodes being deployed for the same application, as was the case in this exercise.

143 One of the goals for widespread use of sensor technologies for air quality monitoring is to
 144 maximise spatial data coverage (Castell et al., 2013; Schneider et al., 2017), by deploying dense

145 networks of sensors, e.g. across urban areas. However, this is not feasible if the nodes present
146 limitations with regard to connectivity or power requirements. As shown in the Table 1 most of
147 the nodes (7 out of 8) are able to operate on battery, minimising the need to provide mains
148 power at the measurement locations. Also, all of them are able to transmit data, either directly to
149 “cloud” platforms or through apps on mobile phones. Only one of the nodes requires Wi-Fi
150 access, which is a potentially limiting factor for large scale deployment across urban areas.
151 Finally, 7 out of the 8 nodes may be deployed outdoors (only one of them requiring additional
152 protection to be built), being resistant to weather conditions. The latter requirement would be
153 irrelevant when dealing with indoor air quality monitoring.

154 For outdoor air quality monitoring, the frequency of maintenance is a relevant parameter.
155 Whereas most of the nodes require no or limited (yearly) maintenance, one node requires
156 maintenance or calibration every 2-6 months. In terms of operation and data availability,
157 frequent maintenance may be an issue for sensor node selection.

158 *2.3. Calibration strategies*

159 Some of the installed AQ microsensors output results in concentration units while others
160 provide voltage or frequency data. Therefore, a pre-processing of raw data was necessary to
161 proceed to concentration units. All sensor nodes (with the exception of the Siemens node) have
162 hence been pre-calibrated. Each team was responsible for their own unit conversion, including
163 different calibrations and conversion strategies, depending on the sensors used. Additional
164 information is presented in Supplementary Material (Table S1).

165 The gas sensors used for the ATh-ISAG node were off-the-shelf metal-oxide sensors that
166 did not undergo a specialized calibration procedure by their manufacturer. In this case it was
167 decided to apply a simple signal correction procedure in order to calibrate the readings received,
168 assuming a linear calibration approach as suggested by Balzano and Nowak (2008).

169 The NanoEnvi node did not undergo a specialized calibration procedure by their
170 manufacturer. The data was not post-processed to correct for temperature and humidity effects
171 or cross-interference with other gases. For the analysis, only the negative concentration values
172 were removed. Negative values were only registered for the NO₂ sensors and represented about
173 20% of the total data.

174 The two ECN-Airbox were calibrated before the Aveiro campaign carrying out co-located
175 measurements with reference equipment at an official monitoring station in Amsterdam
176 (NL49014 GGD Vondelpark). The ECN sensors were developed to minimize cross
177 contamination and meteorology interference. For the NO₂ sensor this was established by
178 introduction of a differential measurement technique enforced by a pre-processing step prior to
179 the sensor by switching frequently to zero ambient airflow (NO₂ removed). Moreover, the
180 sample flow was stabilized by a patented RH delaying cartridge.

181 The AQMesh v 4.0 pods used in this experiment reported NO, NO₂, CO and O₃
182 concentrations which are the result of a two-stage process, following the AQMesh standard
183 operating procedure (SOP). In the first stage, the AQMesh algorithm (a fixed mathematical
184 formula which does not use machine learning) is applied to the raw data in counts, which are
185 converted to ambient concentration units, along with compensation for various environmental
186 effects upon the sensors, providing precision of measurement. The AQMesh SOP requires that
187 pods are deployed 2 weeks prior to the actual measurements for stabilization and application of
188 scaling using reference data. Given that the logistics did not allow doing so in this field
189 campaign, the calibration via scaling was done in this study using the reference data from the
190 trial period.

191 The EveryAware SensorBox (EA SB) is a portable, low-cost measurement device, which
192 allows measurements of the personal exposure to traffic pollution. The EA SB combines a

193 number of low-cost electrochemical and metal oxide sensors to measure concentrations of CO,
194 NO₂, gasoline exhaust and diesel exhaust. Furthermore, additional sensors have been added to
195 allow correction for meteorological influences (T and RH sensor) and for cross sensitivities (O₃
196 and VOC sensor).

197 The ENEA-Air-Sensor Box used in the Aveiro campaign consisted of commercial low-cost
198 electrochemical sensors for gas (NO₂, O₃, CO, SO₂) detection and commercial cost-effective
199 optical particle counter (OPC) for particulate matter (PM10) detection, including miniaturized
200 sensors for meteorology parameters (T and RH). Before the Aveiro campaign, ENEA calibrated
201 the prototype Airbox by co-located measurements with reference analysers in an official air
202 quality monitoring station at JRC, located in Ispra, Italy.

203 During the Aveiro Intercomparison Exercise, two SNAQ (Sensor Networks for Air Quality)
204 boxes were deployed (hereafter referred to as CAM). Both units utilised Alphasense
205 electrochemical cell (ECC, model B4/BH) for species NO, NO₂, O₃, CO, SO₂ and Total VOC.
206 Measurements of CO₂ (SenseAir K30) and particulate matter (University of Hertfordshire
207 CAIR) were also undertaken. The CAM boxes employ Gill WindSonic 2-D sonic anemometers
208 to assist in source attribution. Before deployment, each sensor box was fitted with new
209 Alphasense electrochemical cells. Throughout the campaign and subsequent data analysis, only
210 the raw signal data from the sensors was used, correcting for temperature and humidity effects
211 as per Popoola et al., (2016). In addition to the factory calibration of the ECC sensors, a second
212 calibration was employed based on a comparison of both CAM sensor boxes with reference
213 instruments of the Department of Chemistry in Cambridge.

214 *2.4. Data analysis and quality control*

215 *2.4.1 Meteorological measurements*

216 One of the most important benefits of AQ monitoring networks is the ability to pinpoint
217 pollution sources, and account for regional (> 50 km), meso-scale (500 m to 50 km) and micro-
218 scale (< 500 m) influences. This requires suitable meteorological data to support the
219 measurements. Due to the influence of urban topography and traffic at the micro-scale it is
220 beneficial to have meteorological measurements in the same place as the AQ sensors (Popoola
221 et al., 2013). During the Aveiro campaign, principal meteorological variables were measured by
222 the IDAD LabQAr van using a Vaisala WXT 520 weather station (Borrego et al., 2016). The
223 CAM_10 and CAM_11 boxes measured wind speed, wind direction, temperature and humidity,
224 allowing comparison of meteorological variables and source apportionment.

225 *2.4.2 Measurement uncertainty*

226 The European Air Quality Directive (EU, 2008) defines the Data Quality Objective (DQO)
227 that monitoring methods need to comply with to be used as indicative measurement for
228 regulatory purposes. The DQO is a measure of the acceptable uncertainty for indicative
229 measurements. According to the Directive, allowed uncertainties are 50% for PM10 and PM2.5,
230 30% for O₃ and 25% for CO, NO_x, NO₂ and SO₂.

231 To assess the performance of each sensor and of the sensor platform as a whole, the
232 measurement of uncertainty has been calculated following the methodology described in JCGM
233 (2008) and Spinelle et al. (2015). The relative expanded uncertainty was estimated using
234 Equation 1, where x_i indicates the reference measurement, y_i the candidate method (sensor), b_0
235 and b_1 are the slope and intercept of the orthogonal regression, respectively, RSS is the sum of
236 squares of the residuals (Equation 2), and u is the uncertainty of the reference instrument.
237 Further details on the calculation of the expanded uncertainty can be found in the Guide for the
238 demonstration of equivalence (EC WG, 2010).

$$239 \quad U_r(y_i) = \frac{2\left(\frac{RSS}{(n-2)} - u^2(x_i) + [b_0 + (b_1 - 1)x_i]^2\right)^{1/2}}{y_i} \quad (1)$$

$$240 \quad RSS = \sum(y_i - b_0 - b_1 x_i)^2 \quad (2)$$

241 *2.4.3 Multidimensional data visualization*

242 In order to investigate the behaviour of the AQ nodes in terms of the monitored parameters,
 243 it was decided to visualize all meteorological and gaseous data in a way that can reveal
 244 dependencies and similarities of patterns; information that can be of value for the node
 245 validation and calibration. For this reason, the T-distributed Stochastic Neighbour Embedding
 246 (t-SNE) method was employed. This is a relatively new nonlinear mapping technique that is
 247 capable of preserving both the local and global structure of a high dimensional dataset (van der
 248 Maaten and Hinton, 2008). As multiple parameters (like air pollutant concentrations and
 249 meteorological conditions) are produced from the operation of the sensor boxes, it is impossible
 250 to simultaneously visualize them and thus investigate possible relationships. The t-SNE method
 251 is capable of visualizing this high dimensional feature space in a lower dimensional (2-D or 3-
 252 D) space. The main characteristic of the method is that the groups or clusters of features (here
 253 consisting of AQ nodes and reference measurements) appearing in t-SNE reflect similarities,
 254 thus the closer the attributes are to each other forming a group, the more they can be considered
 255 as similar or as belonging to the same cluster. Although the method itself requires multiple
 256 iterations and tests and should be only be used for data exploration purposes, ideally,
 257 measurements for the same parameter (e.g. NO₂) should be close to each other, regardless of the
 258 measuring unit (sensor) producing them.

259 *2.4.4 Multivariate calibration*

260 Air quality multisensor device suffer from several limitations that have their basis in the
 261 technological nature of the transducers they rely on. Cross sensitivities make the sensor
 262 response depend not only on the target gas, but also on the concentrations of so called
 263 interferent species. Environmental parameters like temperature, relative humidity and pressure
 264 have similar influence on the sensor to target gas response curve. A calibration that does not
 265 take into account these parameter values is prone to failure. Laboratory based calibration
 266 procedures use a limited number of combinations of target gas and interferent concentrations.
 267 The difficulties in replicating the exact conditions that a calibrating node will encounter in its
 268 operating life represent a significant limit to these procedures. In particular, the number of
 269 different configurations of target gases and interferent concentrations together with
 270 environmental conditions may undergo a “combinatorial” expansion. In order to overcome these
 271 limitations, several researchers (De Vito et al., 2008; Kamionka et al., 2006) proposed the use of
 272 field measurements taken with a gas multisensor device, as well as a collocated reference
 273 analyser, to build a data-driven, multivariate calibration procedure with the aid of neural
 274 networks (Webb, 2005). Recently, the use of machine learning approaches has become common
 275 practice in the field, primarily for the performance and cost benefits that can be obtained with
 276 respect to classic approaches (Vidnerová and Neruda, 2016). Those pioneering results were
 277 confirmed by Spinelle et al. (2015), in a series of multinode studies, highlighting the significant
 278 benefits of this approach when dealing with real world deployments to the point of partially
 279 reaching the DQO set by the EU directive for indicative measurements.

280 Another driver of sensor node performance limitation is the dynamic behaviour of the
 281 sensors. It is usually characterized by a limited responsiveness, thus minimising their ability to
 282 deal with rapid transients of pollutants concentrations that may occur in pervasive near-to-road

283 or mobile deployments. In addition, the responsiveness of a single sensor to different gases may
284 differ. To tackle this issue, Esposito et al. (2016) have shown the effectiveness of machine
285 learning approaches, improving the dynamic and overall performances of fast sampling nodes.

286 The manufacturing variability is a significant limitation to the scalability of each calibration
287 procedure. As previously mentioned, calibration operation for each different node may be
288 required, since their response behaviour to target gases as well as interferences may differ
289 significantly as also shown by Castell et al. (2017). Drift effects, and specifically those related
290 with ageing and poisoning may affect the long term performance of the sensor node requiring
291 relatively frequent recalibration actions (Tsujita et al., 2005; S. Marco et al., 2012). Data driven
292 approaches have been proposed for the improvement of long term performances, but the
293 problem remains open (S. Marco et al., 2012; De Vito et al., 2012).

294 The univariate calibration approach was implemented with the aid of a simple linear
295 regression (LR). For each sensor, a calibration function was established by assuming the
296 linearity of the sensor response with the reference measurement for each pollutant. Orthogonal
297 linear regression with the minimization of square residuals of the sensor response versus
298 reference measurement was also used.

299 The multivariate calibration approach was implemented with the aid of Computational
300 Intelligence (CI) methods from the Machine Learning field. Preliminary computational
301 experiments, and literature methods (Spinelle et al., 2015; De Vito et al., 2018) led to use of two
302 CI algorithms; Random Forests (RF) (Tin Kam Ho, 1995) and shallow Feed Forward Neural
303 Networks (FFNN) (Bishop, 2006). Model results were evaluated with the aid of appropriate
304 model performance indices as described in detail in Borrego et al., 2016.

305 Random Forest (RF) is an algorithm belonging to the ensemble-based classifiers that makes
306 use of decision trees and then estimates the value of the target parameter as the average of the
307 forecasts of each individual tree of the ensemble (Breiman, 2001). The RF algorithm uses
308 bootstrapping; the initial feature space consisting of M observations of N input parameters
309 (features) sampled with replacement to generate a number of M training sets. Each set is then
310 used to train a decision tree. For each tree, a random number of features, n , is selected with
311 replacement (the so called bagging procedure) thus formulating a random subset of the initial
312 instances and then a decision tree is fitted (trained) on each subset and is therefore used in order
313 to predict the parameter of interest. Here the random number of features is calculated as
314 $n = \text{int}(\log_2(N+1))$, where int is the integer part of a real number, according to Breiman, 2001.
315 An unlimited number of levels and nodes are used for each of the aforementioned random trees.
316 The final prediction is calculated as a (voted or averaged) sum of individual predictions, thus
317 making RF an ensemble-based meta-classifier. It should be noted that node split per tree, i.e.,
318 the splitting per node based on feature threshold values, was the optimum among a random
319 subset of the features of size n . On this basis, the variance decreases due to the averaging in the
320 ensemble, leading to an overall improvement in results.

321 Following the same modelling approach, shallow Feed Forward Neural Networks (FFNN)
322 (Bishop, 2006) have been trained and tested for each of the AQ nodes. FFNN has become a
323 reference tool for multivariate regression in the machine learning community making it a useful
324 comparison method. Given its small operative computational complexity, it is arising as a tool
325 of choice for implementing on board embedded intelligence when availability of computational
326 resources is scarce e.g. when targeting mobile/wearable deployments (De Vito et al., 2018). In
327 our implementation, we made use of 5 sigmoidal neurons in the single hidden layer for the 1-h
328 dataset, and a three layer FFNN with 5 hidden layer neurons and a single output for the 1-min
329 dataset. The reported performance indices are averaged along multiple implementations of the
330 cross validation procedure, due to the inherent dependence of the performance on the random
331 choice of the initial network weights parameter. No hyper parameter optimization procedure
332 (Bishop, 2006) has been implemented.

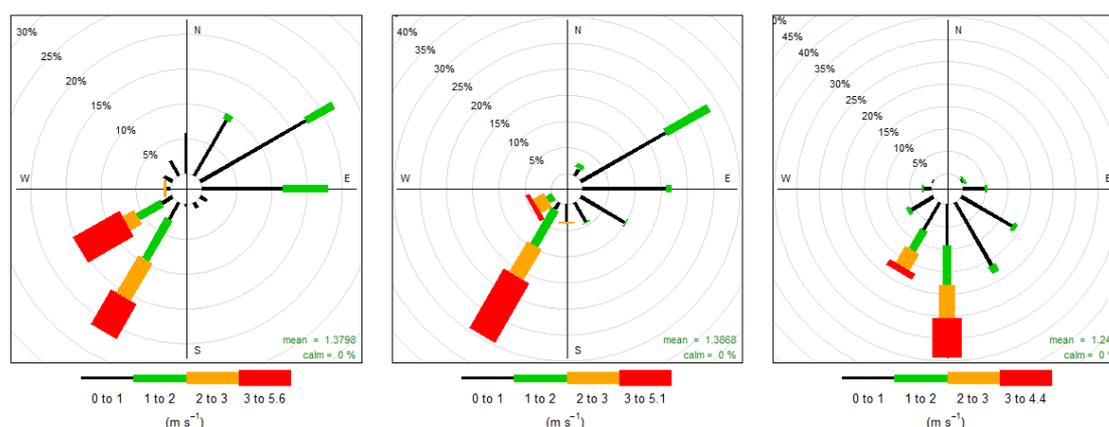
333 Performance estimation for both RF and FFNN was done on the basis of a 10-fold cross
 334 validation procedure: the initial data set was randomly divided into 10 equal subsets. Nine of
 335 them were used for model training while the tenth was used for model evaluation. This
 336 procedure is repeated ten times, each time leaving a different subset out, to be used for model
 337 evaluation. In this way, ten models were trained and evaluated, and the overall prediction scores
 338 were calculated as averages of the individual models.

339 Sensor responses and IDAD reference values have been used for the cross validation based
 340 training and performance assessment. It is worth noting that for each sensor box, all available
 341 and meaningful raw sensor responses have been used to provide the multivariate input to the
 342 above mentioned CI methods. For the purposes of the present study, the WEKA computational
 343 environment was employed (Hall et al., 2009) for RF while Matlab was used as the
 344 computational environment for ANNs.

345 3. Results and discussion

346 3.1. Identification of pollutant sources

347 As mentioned in section 2.4 during the Aveiro campaign, principal meteorological variables
 348 were measured by the IDAD LabQAr van, using a Vaisala WXT 520 weather station.
 349 Additionally, CAM_10 and CAM_11 boxes included direct observations of wind speed, wind
 350 direction, temperature and humidity. The results are presented in Fig. 2 and Fig. 3, allowing
 351 comparison of meteorological variables and source apportionment.

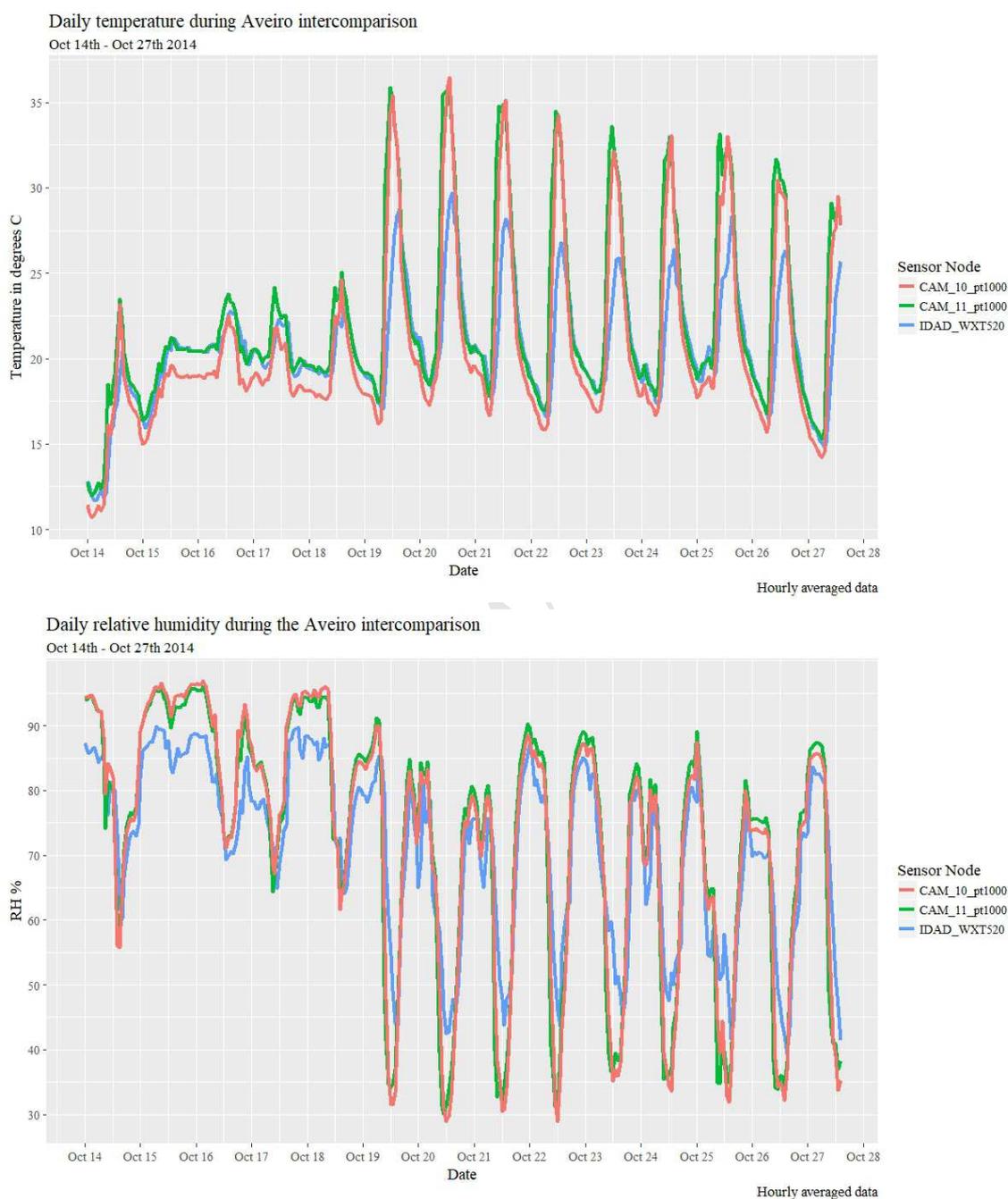


352
 353 **Fig. 2.** Wind speed and wind direction measured (from left to right) by IDAD LabQAr and CAM_11 and CAM_10 sensors. Wind
 354 speeds are split by the coloured intervals shown in each panel, the grey circular lines show the directional frequency (%).

355 From the wind speed and direction data, it can be seen that both the CAM_11 and IDAD
 356 sensors capture the split between west/south-westerly (W/SW) and east/north-easterly (E/NE)
 357 winds in terms of frequency, with the strongest winds from a SW direction during the first
 358 week. The sonic anemometer on CAM_11 was partially blocked to the west by the telescopic
 359 pole, hence it has missed the frequent winds to the WSW measured by the WXT520. In
 360 contrast, CAM_10 measured predominantly southerly winds (S) throughout the study period –
 361 possibly as a result of its mounting position lower down on the van roof.

362 In Fig. 3 CAM sensor boxes demonstrate good agreement with the WMO certified
 363 WXT520, well capturing the diurnal trends. Nevertheless, there is a clear positive bias of 5°C in
 364 temperature readings from both CAM boxes relative to the WXT520, which is especially
 365 prevalent during the second week of measurements when the diurnal range was around 20°C. A
 366 smaller positive bias (~ +2 – 3%) is also observed in relative humidity readings compared to the
 367 WXT520.

368 The temperature bias appears to be systematic with the PT1000 thermocouples used in this
 369 study, and for future measurements will need to be corrected, especially under periods of high
 370 insolation as experienced during the second week of the campaign (Borrego et al., 2016). It is
 371 possible that the lack of an adequately ventilated and screened enclosure for the temperature and
 372 humidity sensors could contribute to these biases, although this requires further investigation.



373

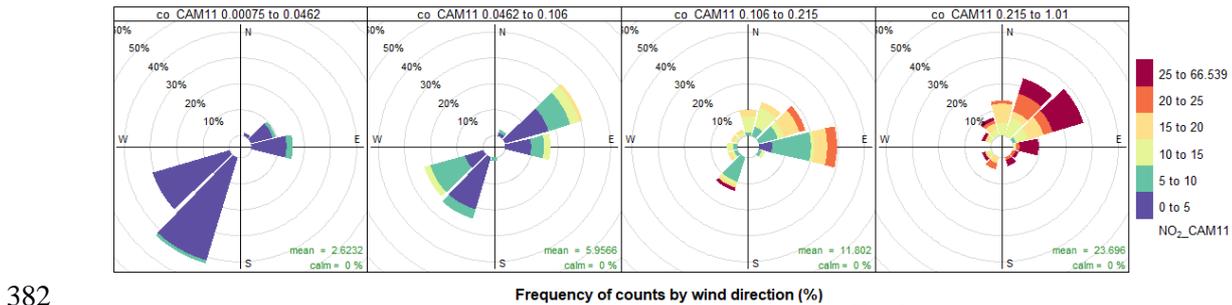
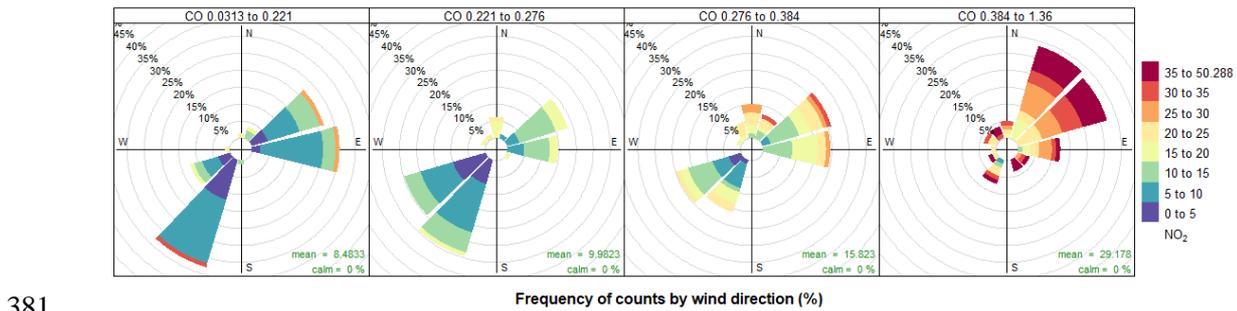
374
375

Fig. 3. Temperature °C (top) and relative humidity % (bottom) measured by IDAD LabQAr, CAM_10 and CAM_11 sensor boxes.

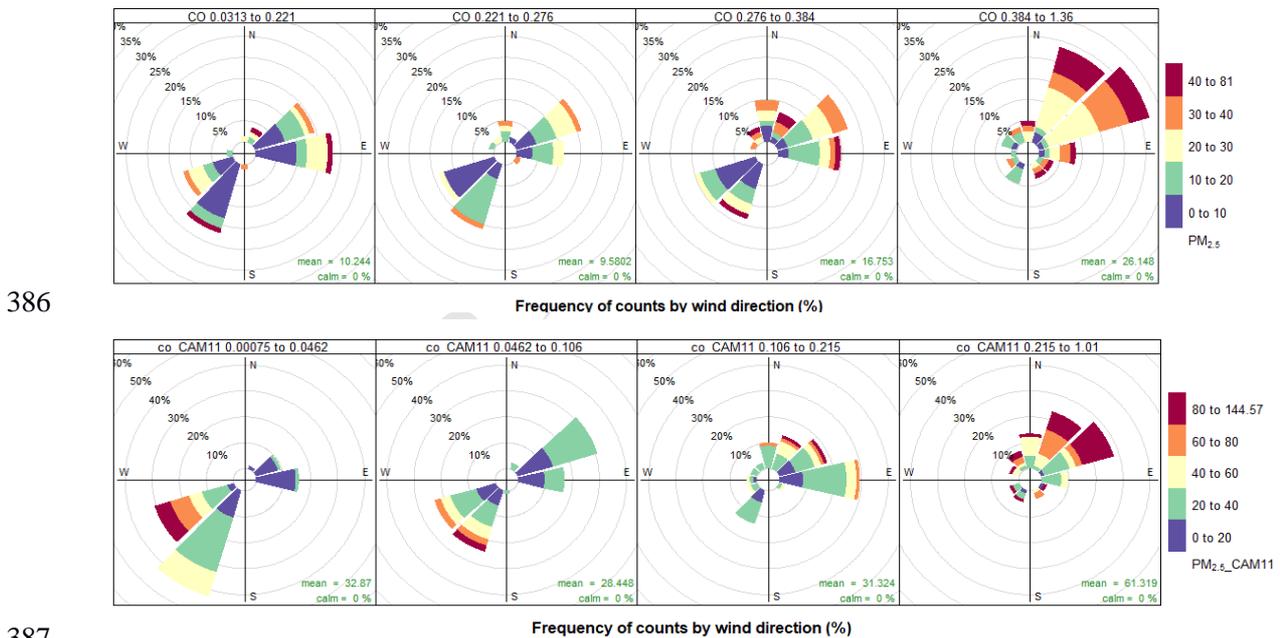
376 Nevertheless, the wind speed and direction data can be used to highlight local or regional
 377 sources of pollution. For example, Fig. 4 and Fig. 5 show pollution rose plots¹ of NO₂ and
 378 PM_{2.5} conditioned with CO. This process effectively plots x and y variables against a third

¹ <http://www.openair-project.org>

379 variable, in this case CO, as concentrations increased during the period of monitoring. As CO
 380 can be used as a tracer for combustion processes, this helps to pinpoint clean and polluted air.



383 **Fig. 4.** Pollution rose plots of NO₂ (ppb) vs wind direction conditioned by CO. IDAD LabQAr (top) and CAM_11 (bottom). The
 384 concentration of CO (ppm) increases from left-to-right. Concentration of NO₂ is shown by the coloured intervals, and wind
 385 directional frequency by the grey contour lines. Hourly averaged data.



388 **Fig. 5.** Percentile rose plots of PM10 ($\mu\text{g}\cdot\text{m}^{-3}$) vs wind direction derived from CAM_11 sensor data, split for day and night periods.
 389 In this case, one-minute averaged data were used. The percentile intervals are shaded and are shown by wind direction, with the
 390 concentration in ppbv indicated by the contours.

391 The IDAD LabQAr reference and CAM_11 sensors both indicate that as the CO+NO₂
 392 burden increases, greater than 60% of the highest concentrations originate in the E/NE direction.
 393 Traffic flow was NE to SW alongside the measurement site (Fig. 1), so under low wind speeds
 394 local emissions will dominate. In contrast under S/SW conditions (Fig. 2), the CO+NO₂ burden
 395 is reduced dramatically (less than 15 ppb NO₂) indicating the importance of higher wind speeds

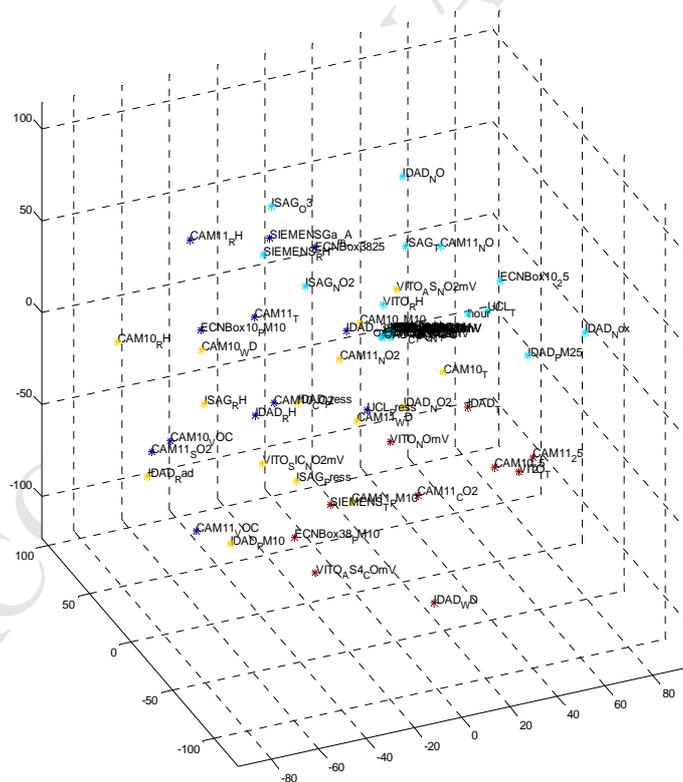
396 for transporting cleaner background air and subsequent mixing and transport of these local
 397 emissions away from the site.

398 Similarly, the CO+PM_{2.5} concentrations are greatest in the E/NE direction for both sensor
 399 nodes, suggesting traffic and/or other local sources of combustion. During the second week of
 400 measurements, low wind speeds, high solar insolation and stagnant conditions lead to an
 401 inversion and build-up of pollutants (Borrego et al., 2016). However, there is also a source of
 402 PM_{2.5} to the WSW of the CAM_11 sensor node, seen to lesser extent in the IDAD data. The
 403 CAM_11 particle sensor uses an optical, not gravimetric technique and shows enhanced
 404 response under high relative humidity (experienced during the first week) and this interference
 405 cannot be ruled out.

406 The examples above show how local meteorological data combined with micro-sensor
 407 networks, can be of use to pinpoint pollution sources, and could be a valuable tool for policy
 408 decisions regarding urban traffic control and development. However, there are caveats.
 409 Meteorological sensors need to be properly sited with adequate protection from direct sunlight
 410 and rain, and need to be close to the pollution sensors themselves, ideally in the same enclosure.
 411 Care needs to be taken with post-processing of the data (for example, with the CAM PT1000
 412 thermocouple temperature sensors used in this study) and where possible a WMO certified
 413 weather station should be used for quality checks.

414 3.2. Parameter dependencies

415 The analysis with the aid of the t-SNE method led to a number of visualizations like the one
 416 presented in Fig. 6.



417
 418 **Fig. 6.** A visualisation of the Aveiro dataset based on the t-SNE method. Proximity is a measure of similarity. Labels by the dots
 419 indicate the AQ node and the parameter (feature) monitored.

420 Although such graphs are not straightforward to analyse, it is possible to identify parameters
 421 that are visualised as being “close” to each other, via rotation and figure magnification. This
 422 “closeness” should actually be interpreted as the tendency that the data points have to organise

423 themselves in neighbourhoods (clusters) of data, on the basis of a mathematically defined
424 similarity. On this basis, we identified that when it comes to meteorological parameters like
425 pressure, measurements of UCL as well as of ATh-ISAG are close to the reference node of
426 IDAD. For gaseous pollutants, it was evident that CAM_11 is close to the reference node, thus
427 indicating a good overall agreement (this being suggested already by the findings of Borrego et
428 al., 2016).

429 Such similarities can also be identified regarding different sensor types belonging to the
430 same node, as it is the case with the VITO sensors for CO (MiCS-5525, MiCS-5521, Figaro
431 2201, Alphasense CO-BF), all of which produce mV as initial output: all four sensors were
432 visualised very close to each other, thus suggesting that they can be considered as similar and
433 providing “equivalent” information. Such findings are useful for selecting, in a next step, the
434 sensor that is also more easily calibrated and possesses additional characteristics like response
435 time for example, in order to deploy a node implementation in a future application.

436 *3.3. Calibration of sensors*

437 To transform the sensor responses into air pollutant concentrations, it is necessary to create
438 a calibration function. In some cases, the calibration function is provided by the manufacturer
439 and it is usually determined by comparing the sensor response versus reference values in a
440 laboratory environment. In the laboratory, the conditions of temperature and relative humidity
441 are controlled and the pollutant concentrations are precisely regulated. However, the laboratory
442 calibration in most cases is not enough to cope with environmental variability and unpredictable
443 interferences found in the field. In these cases, the sensor performance (in terms of
444 concentration estimation quality) often decrease dramatically; presenting in some cases biases
445 that can be partially corrected if a field calibration is employed as described in Spinelle et al.
446 (2015). In this section, the results are presented from the field calibration obtained comparing
447 the data from the sensor platforms with the data from the reference instruments in Aveiro.

448 The results from the linear regression univariate calibration, implemented with the aid of a
449 simple linear regression, highlight the need for field calibration, as most of the sensors present a
450 slope and an intercept that significantly differ from the optimal target values of 1 and 0,
451 respectively. This confirms the findings of the target diagram regarding the bias of sensor values
452 (Borrego et al., 2016). Actions also must be taken to correct both the zero and range of sensors
453 in field calibration conditions. Overall results suggest that the CO sensors are generally the most
454 effective, with intercept close to 0 and uncertainty in the slope within 35%.

455 To minimize the concentration estimation errors, we made use of the (1-h) IDAD dataset of
456 reference meteorological measurements, plus microsensor data, to construct multivariate
457 calibration models. As mentioned in section 2.4, two computational intelligence (CI) algorithms
458 were employed, namely RF and FFNN. We compared the results received with those obtained
459 via the basic lab-based and linear calibration methods. Table S2 defines all statistical indicators
460 and Table S3 shows the values of all four calibration methods used (see Supplementary
461 Material), while Fig. 7 compares all methods for all sensor nodes and pollutants in terms of
462 correlation coefficient.

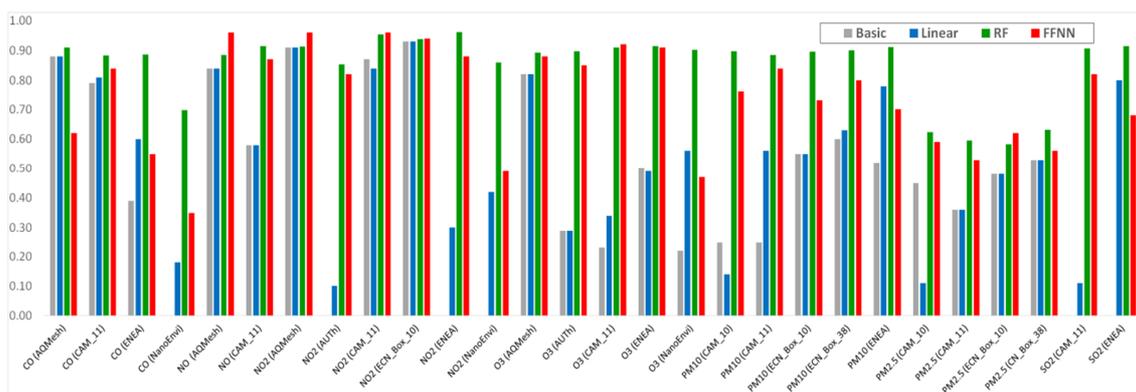


Fig. 7. Comparison of the basic, linear, RF and FFNN oriented calibration methods in terms of reference correlation coefficient achieved for all sensor nodes and pollutants based on hourly measurements. Missing values indicate insufficient data.

463
464
465

466 In the case of CO the good performance of the basic and linear calibration procedures
467 (max(r)=0.88) is further improved via RF and FFNN (max(r)=0.91). The latter demonstrates a
468 higher MRE and a lower FOEX (in absolute values) when compared to FFNN for AQMesh and
469 CAM_11 thus indicating that CI-based calibration procedures are sensitive to errors and require
470 further improvement.

471 Regarding NO sensors, both RF and FFNN improve the already good performance of the
472 basic and the linear calibration method in terms of almost all statistical indices. Thus basic and
473 linear methods reach a max(r)=0.84 that rises up to r =0.96 with FFNN.

474 All six NO₂ sensor nodes demonstrate good performance indicators (max(r)=0.91 with basic
475 and linear methods, reaching up to max(r)=0.96). It is worth noting that the application of RF or
476 FFNN algorithms leads to a strong improvement in the correlation coefficient especially for
477 those nodes that demonstrated the worst performance in basic calibration mode, thus greatly
478 improving their operative capabilities. This is the case for AUTH-ISAG, ENEA and NanoEnvi,
479 where the calibrated values reach now r >0.7.

480 For O₃ the correlation coefficient achieved for basic calibration ranges from r =0.22 up to
481 r =0.82, while with linear calibration ranges from r =0.29 up to 0.82. RF leads to very good
482 correlation coefficients ranging between r =0.89 to r =0.91 while FFNNs demonstrate a range of
483 r =0.47 to r =0.92. The MAE of both RF and FFNN calibration results is lower than basic or
484 linear calibration results. On this basis, the two CI methods improve the calibration outcomes
485 for O₃.

486 For PM10, results indicate a good overall correlation between the reference and the
487 available measurements, A correlation coefficient of r =0.91 was the maximum value (for
488 ENEA) and of r =0.88 was the minimum value (for CAM_11) achieved with the use of RF. On
489 the other hand, a correlation coefficient of r =0.84 was the maximum value (for CAM_11) and
490 r = 0.7 was the minimum value (for ENEA) achieved with the use of FFNN.. In all cases, FFNN
491 surpasses basic and on field linear calibration methods, while RF performs even better than
492 FFNN, for all sensor boxes. On the other hand, the CRMSE (Centred Root Mean Square Error)
493 achieved with the use of FFNN (being in all cases below 25) is much lower than the one
494 achieved via RF (being in all cases above 145) for all sensor nodes, the best one observed for
495 CAM_11. Concerning the MBE, RF led to lower values in comparison to FFNN. The FOEX
496 index of over/under estimation is closer to the ideal one (zero) for RF results with respect to
497 FFNN results, yet both are clearly closer to zero in comparison to basic calibration, while linear
498 calibration is similar to FFNN.

499 For PM2.5, results indicate that both RF and FFNN improve the correlation coefficient
500 ranging from r =0.53 up to r =0.63 in comparison to the best one achieved with basic calibration
501 (ranging from r =0.36 up to r =0.53). The CRMSE with FFNN is again much better compared to
502 the one achieved with RF.

503 Both sensor boxes (CAM_11 and ENEA) with SO₂ measurement demonstrate high
 504 correlation coefficient (with a maximum surpassing 0.9), low MBE, and low CRMSE. The
 505 FOEX index is also improved, with RF providing with the best values.

506 Considering fast sampling systems suitable for mobile and pervasive deployments, results at
 507 1-min sampling rate have been computed. Basic statistical performance indicators are reported
 508 for the sensors that provided 1-min data in Table 2, considering the same indices adopted by
 509 Borrego et al. (2016). In this case, the sensor performance analysis was carried out using the
 510 observed data from sensor platforms with 1-min sampling rate and the reference data provided
 511 by the 1-min IDAD dataset. As a further comparison baseline, univariate linear regression
 512 models have been applied to targeted sensors to estimate the target concentration using 1-min
 513 data. Results are reported in Table 3.

514 **Table 2.** Basic performance indicator for target sensors using off the shelf basic calibration (1-minute sensor data used). No off the
 515 shelf calibration was provided for Siemens node sensors. Acronyms are explained in Table S2 of the supplementary material.

Pollutant	Sensor node	MBE	r	r ²	CRMSE	NMSE	FB	FOEX	MAE	MRE
O ₃	CAM_11	22.22	0.24	0.06	21.22	1.78	1.30	-2.93	24.19	1.40
O ₃	VITO	-16.05	0.09	0.01	10.91	36.45	-1.92	-40.36	16.10	0.82
NO ₂	CAM_11	-2.28	0.82	0.67	6.76	0.43	-0.37	-29.04	4.56	0.34
NO ₂	ECN	0.93	0.87	0.75	6.56	0.26	0.14	-22.62	4.28	0.33
NO ₂	VITO (NO ₂ MiCS2710)	-9.28	-0.30	0.09	11.44	5.27	-1.66	-28.15	9.95	0.70
NO ₂	VITO (Figaro)	-10.01	-0.39	0.15	11.88	9.15	-1.77	-28.49	10.86	0.81
CO	CAM_11	-0.21	0.80	0.64	0.17	3.06	-1.38	-46.88	0.24	0.86
CO	Siemens	-	-	-	-	-	-	-	-	-
CO	VITO (Alphasense)	1.36	0.43	0.19	0.24	0.61	1.82	48.03	1.36	5.43
CO	VITO (MiCS5525)	1.81	0.30	0.09	0.25	0.69	1.88	48.03	1.82	7.11
CO	VITO (MiCS5521)	2.47	0.59	0.35	0.22	0.75	1.93	47.89	2.47	9.62
CO	VITO (Figaro)	1.03	0.77	0.59	0.18	0.54	1.73	47.81	1.03	3.95
NO	CAM_11	9.91	0.47	0.22	20.65	1.54	1.12	27.99	15.25	8.69
SO ₂	CAM_11	30.14	-0.23	0.05	12.54	6.92	1.99	39.05	30.15	19.15

516 **Table 3.** Linear regression outcomes (1-minute sensor data used). Acronyms are explained in Table S2 of the supplementary
 517 material.

Pollutant	Sensor node	MBE	r	r ²	CRMSE	NMSE	FB	FOEX	MAE	MRE
O ₃	CAM_11	2.86	0.34	0.12	8.87	10.47	0.28	-19.97	6.69	0.66
O ₃	VITO	-0.19	0.09	0.01	10.89	108.35	-0.02	-0.26	8.80	1.85
NO ₂	CAM_11	0.26	0.83	0.68	6.25	0.45	0.04	5.81	4.25	0.60
NO ₂	ECN	1.85	0.88	0.78	5.92	0.31	0.26	-6.00	3.76	0.38
NO ₂	VITO (NO ₂ MiCS2710)	-0.02	0.30	0.09	10.75	9.82	0.00	11.97	8.29	1.31
NO ₂	VITO (Figaro)	0.00	0.39	0.15	10.38	5.64	0.00	13.22	7.72	1.12
CO	CAM_11	0.02	0.80	0.64	0.16	0.58	0.12	-4.39	0.11	0.40
CO	Siemens (AppSens1)	0.00	0.50	0.25	0.23	2.90	0.01	4.90	0.16	0.55
CO	Siemens (AppSens2)	0.00	0.58	0.33	0.22	2.08	-0.01	4.97	0.15	0.54
CO	Siemens (Ga2O3hp)	0.00	0.34	0.12	0.25	7.29	0.00	8.14	0.18	0.64
CO	Siemens (Ga2O3hpHeater)	0.00	0.30	0.09	0.25	10.54	0.00	10.22	0.18	0.63
CO	Siemens (MicronasPht_L)	0.00	0.26	0.07	0.26	14.69	0.00	8.67	0.18	0.65
CO	Siemens (MicronasPt_L)	0.00	0.17	0.03	0.26	31.35	0.00	8.68	0.18	0.69
CO	VITO (Alphasense)	0.00	0.43	0.18	0.24	4.43	0.01	5.65	0.19	0.71
CO	VITO (MiCS5525)	0.00	0.30	0.09	0.25	9.25	0.00	9.58	0.18	0.70
CO	VITO (MiCS5521)	0.00	0.60	0.35	0.21	1.73	-0.01	6.17	0.16	0.61
CO	VITO (Figaro)	0.00	0.78	0.59	0.17	0.65	-0.01	3.02	0.12	0.46
NO	CAM_11	0.26	0.40	0.16	20.05	5.15	0.04	16.85	11.00	4.53
SO ₂	CAM_11	-0.01	0.22	0.05	0.52	21.85	-0.003	-0.75	0.44	0.27

518

519 By using the same experimental design for the 1-minute data, we also report the results
520 obtained by:

- 521 (a) the application of the Random Forest algorithm (Table 4)
522 (b) a FFNN (three layers with 5 hidden layer neurons) (Table 5)

523 In all cases both RF and FFNN greatly improve the node performance in comparison to the
524 basic calibration and the linear correlation based-calibration.

525 Obtained results show a significant improvement of the performance indices with both CI
526 architectures. The use of RF as well as FFNN, strongly improves the performances obtained by
527 linear regression estimators: in all cases (with the exception of CAM_11 monitored CO for
528 FFNN), both RF and FFNN greatly outperform linear and basic calibration performance,
529 confirming the power of multivariate non-linear regression when coupled with field data for
530 microsensor based air quality monitor calibration. Moreover, the obtained results indicate that
531 the RF approach outperforms the Neural one in several cases. To the best of our knowledge this
532 is the first study to report this advantage in this particular field; providing an insight into on
533 field calibration for further investigation.

534 **Table 4.** Random Forests (RF) based non-linear multivariate regression outcomes. Acronyms are explained in Table S2 of the
535 supplementary material.

Pollutant	Sensor node	MBE	r	r ²	CRMSE	NMSE	FB	FOEX	MAE	MRE
O ₃	CAM_11	0.00	0.98	0.97	4.20	0.03	0	0.77	1.32	0.20
O ₃	VITO	0.01	0.94	0.89	14.17	0.11	0	2.68	2.05	0.29
NO ₂	CAM_11	0.01	0.94	0.89	14.39	0.11	0	11.06	1.97	0.17
NO ₂	ECN	-0.04	0.94	0.88	14.73	0.12	0	5.46	2.31	0.29
NO ₂	VITO	-0.02	0.92	0.85	19.48	0.16	0	10.92	2.38	0.27
CO	CAM_11	0.005	0.94	0.88	0.01	0.13	0.01	4.98	0.07	2.80
CO	Siemens	0.003	0.92	0.85	0.01	0.15	0.01	0.37	0.07	4.73
CO	VITO	0.002	0.79	0.62	0.03	0.38	0	4.81	0.11	6.35
NO	CAM_11	0.07	0.86	0.74	112.22	0.26	0	21.47	4.76	1.27
SO ₂	CAM_11	-0.002	0.97	0.95	0.02	0.05	0	0.39	0.09	0.05

536 **Table 5.** Feed Forward Neural network (FFNN) based non-linear multivariate regression outcomes. Acronyms are explained in
537 Table S2 of the supplementary material.

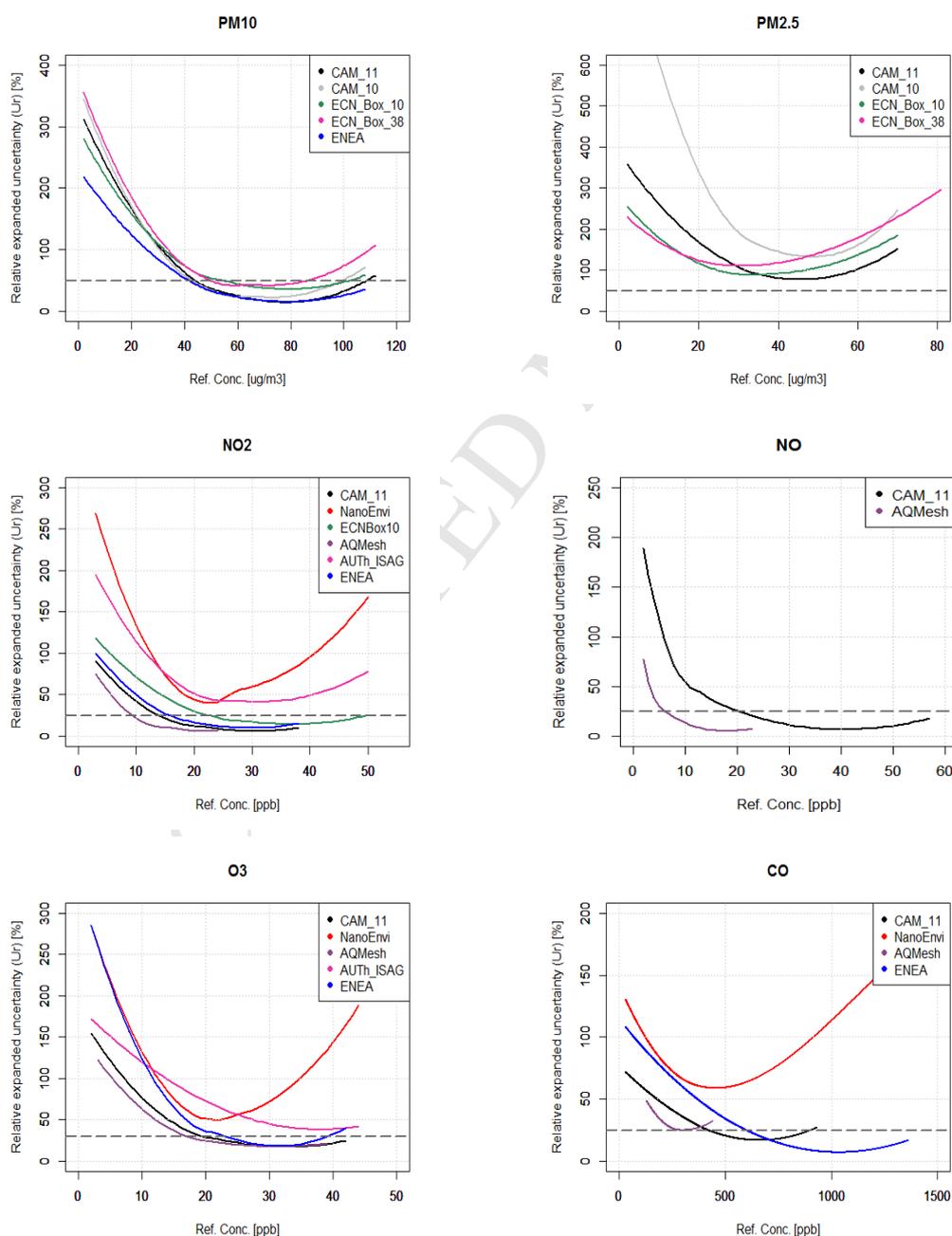
Pollutant	Sensor node	MBE	r	r ²	CRMSE	NMSE	FB	FOEX	MAE	MRE
O ₃	CAM_11	2.79	0.93	0.86	4.49	0.16	0.28	-27.72	2.60	0.18
O ₃	VITO	-0.16	0.98	0.96	2.31	0.05	-0.02	0.25	1.64	0.20
NO ₂	CAM_11	-3.50	0.90	0.81	4.90	0.23	-0.61	-24.77	2.00	0.25
NO ₂	ECN	0.78	0.90	0.81	4.96	0.24	0.11	-31.69	3.25	0.31
NO ₂	VITO	-0.03	0.91	0.84	4.50	0.19	0.00	5.15	2.84	0.30
CO	CAM_11	-0.06	0.71	0.51	0.14	0.91	-0.35	-25.80	0.09	0.34
CO	Siemens	0.11	0.76	0.58	0.17	0.71	0.04	1.47	0.12	0.44
CO	VITO	0.00	0.86	0.74	0.13	0.33	-0.01	4.28	0.10	0.37
NO	CAM_11	-6.26	0.62	0.39	7.91	1.56	-1.34	-22.37	2.39	0.84
SO ₂	CAM_11	-0.06	0.93	0.86	0.22	0.16	-0.07	-27.32	0.16	0.10

538 3.4. Measurement of nodes expanded uncertainty

539 The results for the relative expanded uncertainty (see EU, 2018; EC WG, 2010) of the single
540 sensors show that, generally, the relative uncertainty of CO targeted sensors is the lowest among
541 the different target gases under analysis. Slightly higher values have been recorded for the
542 uncertainty of some of the NO and NO₂ sensors. AQMesh and ECN nodes demonstrate the
543 lower relative expanded uncertainty U_r for NO₂, with U_r reaching below 30% and 60% for
544 concentrations above 20 ppb, for the two aforementioned nodes. However, the AQMesh NO₂
545 sensor seems to increase the U_r for concentrations over 30 ppb. For PM10, the results vary
546 considerably from platform to platform; the ECN platform, for example, presents lower relative
547 uncertainty for concentration values above 80 µg.m⁻³ approaching DQO limits, while the CAM

548 platforms present an U_r over 600% for all the concentration range. For O_3 , AQMesh and
 549 NanoEnvi nodes show a reduction in U_r values for concentrations over 10 ppb, while with
 550 AQMesh presents the lowest relative uncertainty. SO_2 sensors are generally characterised by the
 551 highest relative uncertainty values for all the gas sensors analysed, with uncertainties values
 552 over 5000%: it should be underlined that these results are influenced by the very low
 553 concentrations recorded during the exercise. Overall, the aforementioned analysis verifies that
 554 even when some of the sensors demonstrate promising uncertainty results, sensor platforms are
 555 still not in compliance with the DQO imposed by the European Air Quality Directive (AQD) for
 556 indicative measurements when off the shelf calibration is used (EU, 2008).

557 Considering the results of the FFNN processed multisensors response, the results show a
 558 significant improvement with respect to off the shelf calibration. Figure 8 represents the relative
 559 expanded uncertainty of the platforms versus reference data with the FFNN processed
 560 multisensors response. As usual, figures show results obtained on test samples that have not
 561 been used in the network training phase.

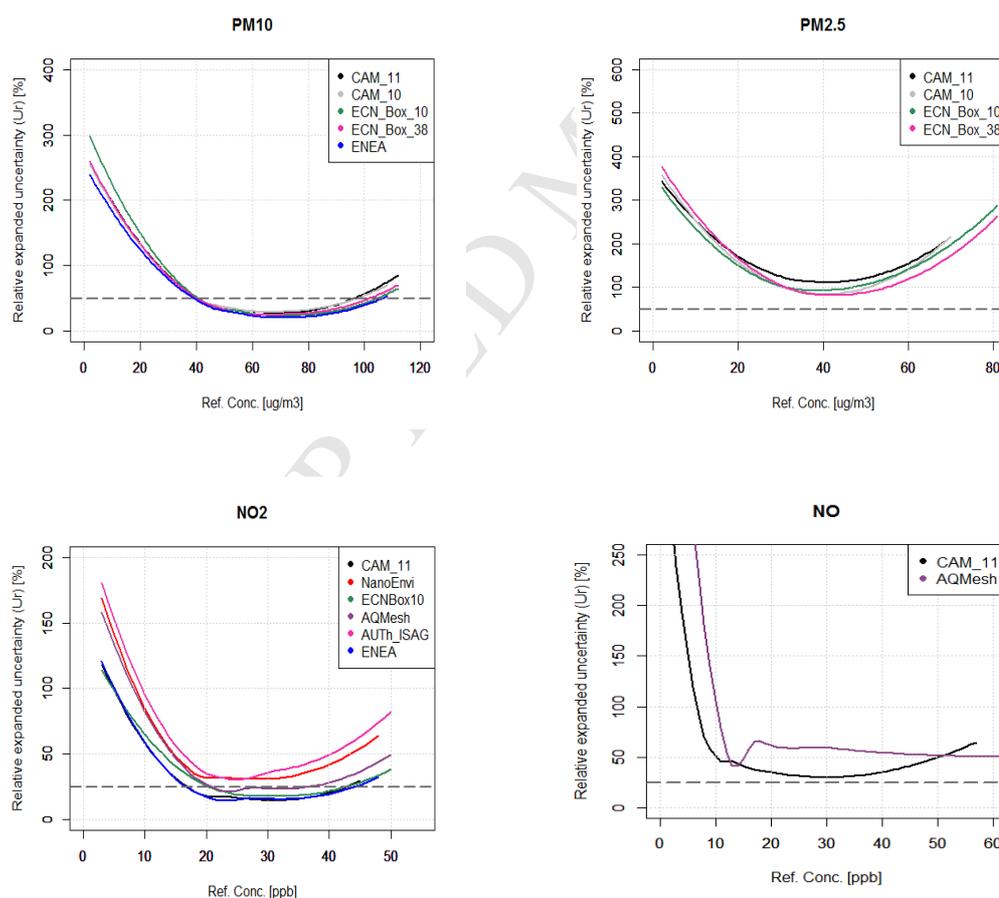


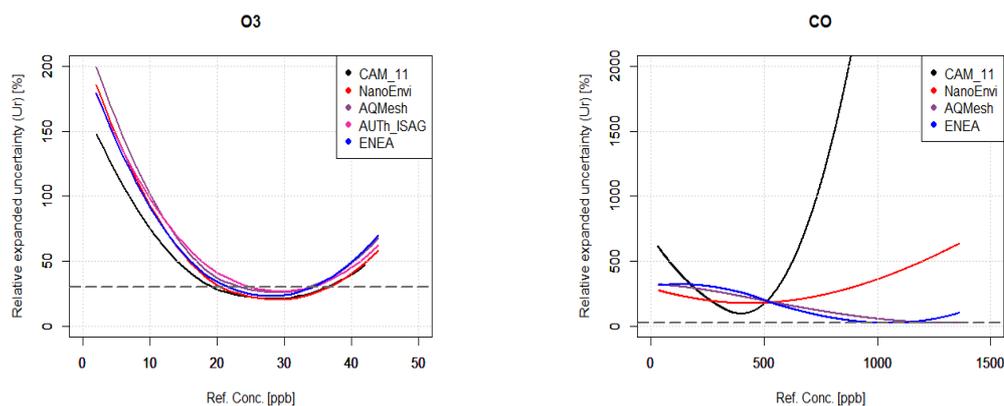
562 **Fig. 8.** Relative expanded uncertainty (%) of the FFNN processed multisensors *versus* the reference data, for PM10 ($\mu\text{g}\cdot\text{m}^{-3}$), PM2.5
 563 ($\mu\text{g}\cdot\text{m}^{-3}$), NO₂ (ppb), NO (ppb), O₃ (ppb) and CO (ppb). Each colour identifies a specific platform: black (CAM_11), red
 564 (NanoEnvi), grey (CAM_10), green (ECNBox10), magenta (ECNBox38), blue (ENEA), orchid (AQMesh), pink (AUPH_ISAG).

565 For PM10 sensors, the estimated uncertainties generally meet the AQD data quality
 566 objective in part of the measurement range (40 - 100 $\mu\text{g}\cdot\text{m}^{-3}$). On the contrary, for PM2.5,
 567 despite the improvement obtained with FFNN calibration, the estimated uncertainty is still
 568 higher than the AQD objective for all values in the relevant range (1 - 80 $\mu\text{g}\cdot\text{m}^{-3}$).

569 With regards to NO₂, the majority of platforms show results that match with the uncertainty
 570 criteria defined in the AQD when concentration exceeds a node dependent threshold. In this
 571 case, only the results for NanoEnvi and ISAG sensors are above the AQD objective across the
 572 entire measurement range (2 - 50 ppb). For O₃ sensors, an equivalent result is obtained, with
 573 some of the platforms showing very promising results.

574 With the NO sensors, the limited number of data available is reflected in the uncertainty
 575 results. In this case the uncertainty is below the reference value in part of the measurement
 576 range when concentration exceeds 5 ppb and 18 ppb respectively for the AQMesh and CAM_11
 577 node. For CO, all but the NanoEnvi and CAM11 nodes meet the AQD objective at least in a
 578 small concentration interval starting from 300 ppb for AQMesh node, 400 ppb for CAM_11
 579 node and 600 ppb for ENEA node. Given the usual relevant concentration range for CO (0.5-10
 580 ppm) the ENEA node gives the best results.





581 **Fig. 9.** Relative expanded uncertainty (%) of the RF processed multisensors *versus* the reference data, for PM10 ($\mu\text{g}\cdot\text{m}^{-3}$), PM2.5
 582 ($\mu\text{g}\cdot\text{m}^{-3}$), NO₂ (ppb), NO (ppb), O₃ (ppb) and CO (ppb). Each colour identifies a specific platform: black (CAM_11), red
 583 (NanoEnvi), grey (CAM_10), green (ECNBox10), magenta (ECNBox38), blue (ENEA), orchid (AQMesh), pink (AUTH_ISAG).

584 Similar results are obtained by the use of the RF algorithm (Fig. 9). For PM10, the results
 585 reflect what has already been shown with the aid of FFNN, with an increased homogeneity
 586 among the node's performances. For PM2.5, again none of the nodes is capable of reaching the
 587 DQO levels. For NO₂, slightly worse results are obtained with RF, however it can be seen that
 588 most of the nodes are able to reach the DQO in relevant concentration ranges. Results for the
 589 NO sensors could not reach the quality objective achievement with the use of RF. On the other
 590 hand, results obtained for O₃ sensors suggest that by the use of RF regression, all the nodes,
 591 including NanoEnvi and AUTH-ISAG are able to meet the intended objectives. Finally, for the
 592 carbon monoxide, results with RF reflect the higher MRE already presented in Table 3.

593 4. Conclusions

594 Continuing developments in Air Quality (AQ) microsensor technologies present the need to
 595 evaluate their ability to support and complement standard monitoring procedures. This paper
 596 introduced the second part of the results of an intercomparison of AQ microsensors with
 597 reference methods during an AQ monitoring campaign in Aveiro (Borrego et al., 2016).

598 Microsensor nodes are low-cost devices with considerable application potential, offset by
 599 important limitations when applied to urban air quality monitoring. Heterogeneity of hardware
 600 and calibration procedures call for an additional "calibration layer" that should be implemented
 601 in any AQ information system that uses them. Sensor uncertainty and performance indicators
 602 need to be addressed in a structured way allowing for field-based sensor comparison and use.

603 This work focused on the analysis of the uncertainty estimation along with the development
 604 of a Computational Intelligence-based "calibration" layer focusing on the hourly data, while
 605 also addressing high temporal resolution (1-min) data. The aim was to estimate the uncertainty
 606 of the measurements according to the DQO of the European Air Quality Directive and to
 607 improve their performance with the aid of a computationally-oriented methodology. For this
 608 purpose, linear regression in addition to Feed Forward Neural Networks and Random Forests
 609 algorithms were employed, in an effort to improve sensor performance based solely on sensor
 610 data and local meteorological data from a reference station.

611 The results confirm that the standard in-factory calibration performances can be strongly
 612 ameliorated by CI-based algorithms with positive outcomes on several statistical indices, as
 613 already reported in the literature (De Vito et. al., 2018). Improved results can also be obtained
 614 for sensor nodes for which no in-factory calibration has been performed. For the first time, this
 615 is confirmed with tests on several multisensor nodes based on different type of solid state

616 sensors and from different independent institutions, including academia and commercial
617 companies, adding an unprecedented generalization value to the presented result. The latter also
618 allows for the adoption of the proposed sensor calibration methodology in order to make sensor
619 readings compliant with the DQO, thus also for the first time suggesting a method that may
620 render AQ microsensors as appropriate for the support of official AQ monitoring tasks.

621 Nonetheless, this procedure should be regarded as an on-site calibration, as it requires
622 ground truth data from the area of interest, thus posing a considerable challenge in terms of
623 method scalability, required for rapid deployment.

624 Generalization of these results should take into account possible sensor and concept drift
625 impacts on concentration estimation quality. It is well known, actually, that the ageing (sensor
626 drifts) and seasonal changes in the pollutants and/or environmental variables joint probability
627 distribution (concept drifts), may lead to suboptimal estimations negatively affecting
628 performance estimator values (Esposito et al., 2017).

629 Furthermore, the performance degradation rate due to ageing effects may be different for
630 each sensor. This behaviour could only be observed and quantified during long-term
631 deployment campaigns and are not the subject of this study.

632 Further work should evaluate the robustness of the methodologies described herein. Sensor
633 profiling with the aid of CI methods in addition to SOM can support aforementioned tasks. In
634 addition, model performance may take benefit from an ensemble approach where different
635 models based on different algorithms can be used in combination.

636

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651

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Highlights

- Several air quality microsensors were tested against reference methods
- Improved correlation between CO, NO₂, O₃, PM10, PM2.5, SO₂ sensors and reference methods through calibration with machine learning techniques
- Parameter dependencies and measurement uncertainty of sensors were evaluated
- Possibility of compliance with DQO of the AQD for indicative measurements
- Microsensors can improve spatiotemporal data resolution to complement current monitoring networks