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RELEVANCE AND RELIABILITY OF NO_2 AND NO MONITORING IN LOW-INCOME COUNTRIES USING LOW-COST SENSORS

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ABSTRACT

Assessing air quality's impact on human health involves monitoring pollutant concentrations such as NO_2, O_3, CO, SO_2, and particulate matter. While high-income countries rely on expensive reference instruments, low-income nations face technological limitations. This study explores the potential of low-cost scientific devices as a viable solution for these regions. The research focuses on evaluating the reliability of low-cost NO_2 sensors and consistency across five identical sensors. Calibration tests in controlled settings reveal a linear model with high coefficients of determination, contrasting with lower coefficients observed during field tests. Variability in intercepts and slopes is evident across time and campaign contexts. Time series analysis using low-cost NO_2 sensors showed that many of the tall peaks atop a fluctuating baseline correlates with peaks identified by reference instruments. Additionally, NO gas sensors are also able to identify pollution peaks in monitoring campaigns. Therefore, such affordable sensors provide valuable insights into pollutant concentration trends, offering indicative magnitude information. However, improving calibration and reliability of these sensors necessitates further research.

I. INTRODUCTION

Nitrogen dioxide (NO_2) typically ranges in average concentrations from 1 to 20 ppb in Belgian outdoor air, while nitric oxide (NO) averages between 1 and 2 ppb [1],[2]. These gases are released into the atmosphere through the high-temperature combustion of fossil fuels in a range of human activities, including mobility (cars, ships), power plants, industrial processes, residential heating, and cooking [3-7]. There are also several natural processes that contribute to their formation (e.g., lightning, volcanic activity, or forest fires). The sum of NO and NO_2 is usually expressed as NO_x (i.e., x is a variable) where the total concentration is expressed in NO_2 equivalents.

The air used to burn fuel contains around 21 vol% oxygen (O_2), 78 vol% nitrogen (N_2), traces of other gases such as carbon dioxide (CO_2) or argon (Ar), and varying amounts of moisture (H_2O). When the fuel reacts with the oxygen in the air, the main products formed are carbon dioxide (CO_2) and water vapor (H_2O), though small quantities of carbon monoxide (CO), volatile organic compounds (VOC) and black carbon (i.e., soot) are also produced. When the air is burned with the fuel, a fraction of the nitrogen (N_2) is transformed into nitrogen oxides [7]. According to the extended Zeldovich reaction mechanism, NO (nitric oxide) is produced when free radicals (e.g., O-atoms, N-atoms, H-atoms, and OH) attack N_2 in the flame region. The generation of NO is influenced by the air-fuel ratio, with higher levels observed when the oxygen content surpasses the ideal stoichiometric ratio for fuel combustion [8],[9].

This formation occurs until all available oxygen is consumed, particularly at temperatures exceeding 1300°C. At temperatures below 760°C, the production of NO is significantly reduced or sometimes nonexistent. In cooler areas (around 400-500°C) near the flame front, smaller quantities of NO might transform into NO_2 [10]. When NO is emitted in the atmosphere it converts through a reaction with O_3: $NO + O_3 \rightarrow NO_2 + O_2$.

NO_2 is a reactive gas involved in the formation of ozone and particulate matter. It is known to be irritating to the human respiratory system. Children and people with respiratory disease are the groups most at risk when exposed to NO_2 concentrations higher than 40-200 µg/m^3 [11],[12]. Moreover, there is a short-term and long-term effect of NO_2 on hospital admissions for cardiovascular diseases [13],[14].

To understand the impact of air on human health, monitoring NO_x concentrations is crucial. NO_x reference gas analyzers utilize chemiluminescence detection technology to measure ultra-low concentrations of NO_x in air. However, the cost of purchasing, operating and maintaining such devices can be prohibitive. An alternative method to analyze these pollutants at lower cost is to use gas sensors paired with an in-house developed data logger [15-20]. An important advantage of such devices is that they are affordable by low-income countries. As a result, such measuring devices contribute to the inclusivity of research. However, the reliability of low-cost measuring devices is often questioned in scientific literature, especially when such devices are compared to the gold standard. Therefore, it is not clear to what extent one can trust the collected data gathered. This contribution will explore the feasibility of using low-cost NO_2 and NO monitoring. The study will be illustrated with measuring campaigns conducted in Belgium and in Cuba.

II. MATERIALS AND METHODS

II.1. DESIGN OF THE LOW-COST DATA LOGGER

A first low-cost, in-house-developed data logger utilizes an Arduino Mega 2560 as a microcontroller board as its central component. In order to transform this microcontroller board into a versatile data logger that can be tailored to the user's specific requirements, we have designed a compact, custom-made expansion shield. This design has been previously published and no changes have been made on the hardware during this work [21-23]. This shield serves the purpose of connecting sensors to the data logger and converting sensor signals into a format that the microcontroller board can interpret. For air quality monitoring campaigns, sensors that measure temperature, relative humidity, inorganic gaseous pollutants, and particulate matter are connected to the expansion shield. In this setup, NO was not measured while NO_2 has been measured with an A-series Alphasense gas sensors. The gas sensors are inserted into an analog conditioning board (Analog Front End no. 810-0023-00 from Alphasense), and that board is connected to the expansion shield using a flat cable and suitable connectors. The advantage of this data logger is that the user can insert or remove sensors and configure the data logger according to his needs.

A second data logger design uses a Raspberry Pi 3B+ as its central processing unit that is connected to a larger PCB sensor shield. The sensors are directly interfaced to the sensor shield without external wiring. Since the array of sensors fixed to the sensor shield is extensive, this single-purpose data logger can be used across a diverse range of applications. Among the various sensors, it features six Alphasense B-type gas sensors that targets CO, NO_2, OX ($NO_2 + O_3$), NO, H_2S, and SO_2. Each gas sensor is connected to an Alphasense Individual Sensor Board, which in turn is seamlessly incorporated into the architecture of the sensor shield. This design has been described in earlier publications [24],[25].

II.2. SENSOR CALIBRATION

The Alphasense gas sensors generate a concentration dependent signal at the working electrode WE and an internal signal that is supposed to be concentration independent at the auxiliary electrode AE. For both electrodes, the signal at zero pollution is denoted by WE_0 and AE_0 respectively. The sensor signal is calculated as $(WE - WE_0) - (AE - AE_0)$ [26]. For the sake of simplicity, it is assumed that the temperature has no effect on the signal. Since WE_0 and AE_0 are only constant for shorter periods, the values are either measured during the calibration experiments, or determined as the minimum value of WE and AE of the time series. Although Alphasense furnishes calibrations for each individual gas sensor, these calibrations must be subject to scrutiny and necessitate periodic verification [27]. Therefore, the reliability of these sensors must be evaluated by calibration experiments. For this reason, the NO_2-sensors have been submitted to 4 different calibration methods [28-32]. The collected data have been processed by linear regression:

- **Low-cost laboratory-based calibration:** In addition to the low-cost data logger, cost-effective calibration methods have been devised to ensure the inclusivity of air quality research in lower-income countries. The calibration setup uses a closed plastic box containing the NO_2 gas sensor. Within the calibration box, the air is initially purified by passing it through a $Ca(OH)_2$-saturated solution. The cleaned air is used to determine WE_0 and AE_0 (i.e., zero calibration). Subsequently, controlled amounts of NO_2 are generated within a closed setup constructed from medical disposables [33],[34]. The generated NO_2 gas, held within a syringe, is then introduced into a second plastic box for dilution. After approximately 20 minutes, a sequence of gas volumes (0.6, 1, 1.6, 1.6, 2, and 2.6 mL) is introduced into the calibration box. The corresponding concentrations inside the calibration box can be calculated from the ideal gas law. Each injection results in a calibration point where sensor signal $(WE - WE_0) - (AE - AE_0)$ and corresponding pollutant concentration is known (i.e., span calibration). The calibration is determined by a linear regression through these points;

- **Calibration in a high-end climate chamber:** The two data logger configurations have been subjected to calibration within a climate chamber at the laboratories of VITO, Belgium, enabling precise regulation of temperature and relative humidity. Certified calibration gas cylinders holding specific concentrations of the target gas in nitrogen are blended with pure nitrogen to achieve the desired concentration level. The gas mixture is then introduced into the calibration chamber. Through modulation of the calibration gas dilution, a step-like function is generated over a defined time span. Furthermore, the concentration of the target gas within the calibration chamber is continuously tracked with the Airpointer, which consists of several reference instruments [24]. At every step in the staircase function, the average reference concentration is calculated and the corresponding sensor signal $(WE - WE_0) - (AE - AE_0)$ is determined as well where WE_0 and AE_0 are the measurements in zero air. As a result, every step results in a calibration point

through which calibration can be determined by linear regression;

- **Field calibration in outdoor air:** One NO_2 sensor underwent field calibration by installing a data logger device in the proximity of a reference measurement station whose data is publicly available [35]. Both devices measure the same ambient pollutant concentrations in outdoor air under realistic conditions. This calibration procedure took place at station 42R801 in Antwerp, Belgium, that is operated by the Flemish Environment Society (Vlaamse Milieu Maatschappij VMM). The natural variations in pollutant levels present in the outdoor air assures for a calibration over a specific concentration range. That range is usually smaller than the one used in the laboratory calibration methods. WE_0 and AE_0 can be determined as the minimum value of WE and AE within the time series when it does not contain low sensor values due to instrumental errors. The time series of the reference instruments and of the low-cost data logger are 1 hour and 2 minutes respectively. With the function VLookUp in Microsoft Excel, the sensor signals WE and AE could be resampled so that values for WE and AE are obtained at the same timestamps as the measurements in the reference time series. The resampling allows the integration of the sensor signal (WE - WE_0) - (AE - AE_0) and the corresponding reference concentration in a single database. The calibration is determined by the linear regression through the large set of data points;
- **In situ calibration:** The quick-and-dirty in situ calibration method assumes that the minimum and average pollutant concentration in the region where the measuring campaign takes place are known. In Antwerp, the minimum sensor signal (WE - WE_0) - (AE - AE_0) of NO_2 in a time series is associated to 0 ppb, while the average value of (WE - WE_0) - (AE - AE_0) is associated to 16.22 ppb. The average concentration has been determined by measurements at other VMM reference stations in the neighborhood, or from literature information. These two calibration points define the linear calibration curve. In principle, this method can be improved by actual in situ measurements of the minimum and average concentration but low-cost methods to perform such field measurements are not yet part of our possibilities.

II.3. COLLECTED DATA

Several experiments and measurement campaigns have been conducted to calibrate the gas sensors and evaluate the reliability of the collected data. The description of these tests are previously published [25],[34]. In this contribution, only the results for NO_2 will be emphasized.

A measurement campaign was carried out on board a 36-year-old ship that is dedicated to near shore operations at the Belgian coast. During the measuring campaign at March 15-18, 2021, the Raspberry Pi-based data logger has been installed in the engine room. In parallel, a measurement campaign is performed with the Air pointer, which was used as a reference-grade instrument [24]. They both measure NO_2 and NO.

In Cuba, a field measuring campaign has been performed in Cienfuegos, from March 14 to April 22, 2022 with a sampling time of 2 minutes using the Arduino-based data logger.

III. RESULTS AND DISCUSSIONS

III.1. SENSOR CALIBRATION

To comprehend the influence of the situational context in which a measurement campaign is conducted, calibration of the A-type NO_2 gas sensor in combination with the Arduino-based data logger has been undertaken using four distinct methods. When looking at the peaks in Figure 1a, almost all NO_2 peaks observed in the field campaign, resulted in a peak in the sensor's signal except when the peak is below the detection limit of the sensor. This suggests that the NO_2 low-cost sensor is able to generate reliable information about the dynamic pattern that can be observed in Figure 1a. However, some tall peaks in the reference measurements resulted in smaller sensor peaks and vice versa. The high scattering in Figure 2a and the low coefficient of determination (i.e., 0.1976) confirm this complicated relationship. In addition, the calibrations performed in laboratory conditions (VITO and low-cost calibration) have a slope that is substantial higher when compared to the linear regression of the field data. There is a factor 5 difference between the VITO and field calibration. However, the VITO-calibration is characterized by a coefficient of determination that is close to 1 (i.e., 0.9997). The in-situ calibration, which is considered as quick and dirty, seems to approach the field calibration the best. It should be remarked that micro-conditions can substantially deviate from the average pollution concentration that is used in the in-situ calibration. The differences between calibration methods suggest that the context of laboratory and real-life outdoor conditions has a substantial effect. The well-controlled conditions in laboratory conditions do not seem to be representative for outdoor conditions.

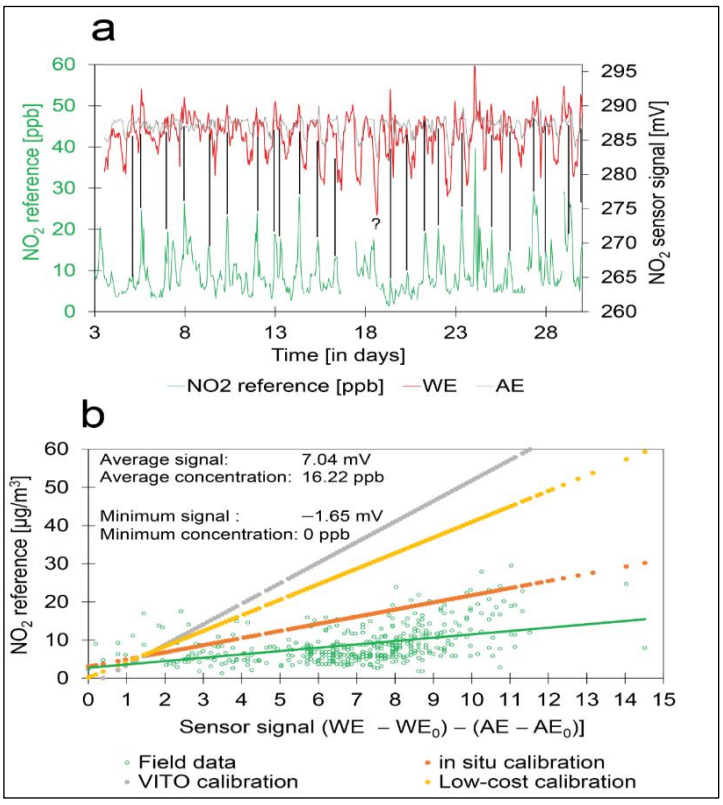

Figure 1: Different calibration methods applied on the same NO_2 sensor. a) NO_2 time series obtained at a VMM air quality monitoring station where the environmental conditions fluctuate in an uncontrolled way; b) Calibration of the low-cost NO_2 sensors using the field data and a superposition of the calibration curves obtained with the other methods.
Source: Authors, (2024).

To understand the impact of the selection of a random sensor from a pool of identical gas sensors and the sensor's history, five distinct B-type NO_2 sensors coupled to the Raspberry Pi-based data logger have been calibrated by VITO. For 3 of them, the calibration has been done twice. Figure 2a shows that the signal of WE at zero air does not restore to the same value. It is also noticed that after exposure to an elevated pollutant concentration, the sensor needs time to regain its equilibrium. This suggest that the sensor's history has an impact on current calibration. Figure 2a shows that each calibration results in a calibration with a high coefficient of determination. However, there is a factor 2 difference between the lowest and highest sensor signal generated by the same NO_2 concentration. In addition, the repeated calibration experiments also resulted in different slopes. Apparently, the sensors do not (always) generate the same response under identical conditions. This suggests that regular in situ calibrations are required to enhance the reliability of quantitative measurements.

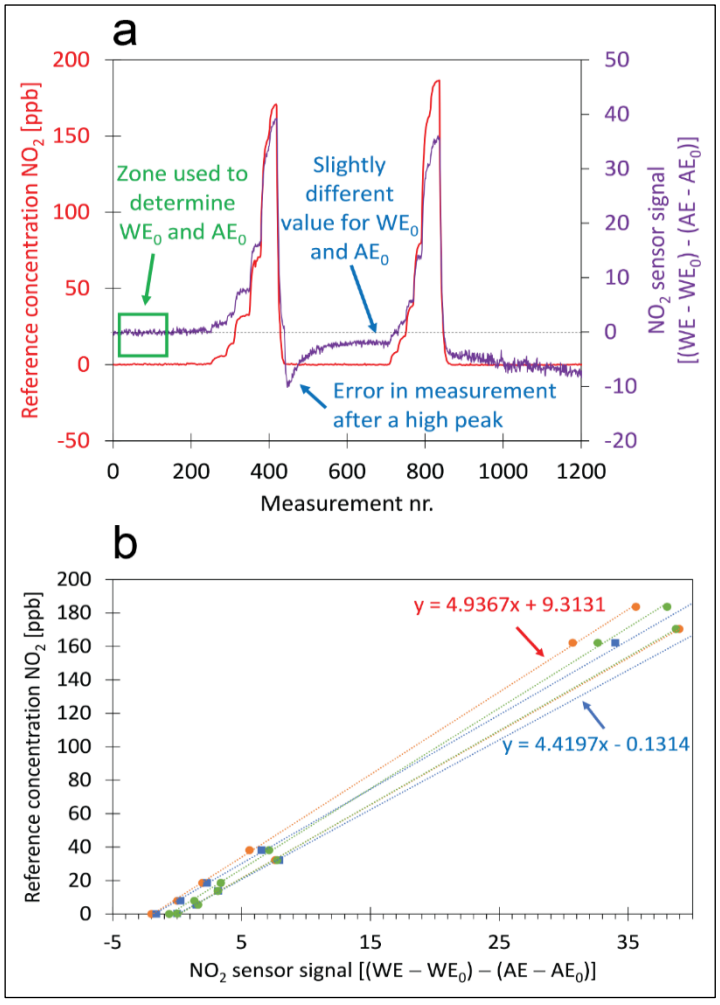

Figure 2: Calibration of 5 NO_2 gas sensors; a) Raw signal of a NO_2 sensor during 2 consecutive calibration experiments; b) Calibration curves for a pool of 5 sensors where the calibration of 3 sensors have been repeated.
Source: Authors, (2024).

III.2. MEASURING CAMPAIGN IN BELGIUM

Figure 3 presents the outcomes of a measuring campaign conducted in the engine room of a ship. More comprehensive details of this campaign are available elsewhere [24],[25]. The time series depicted in Figure 3 reveals that the structure within these series has arisen from a dynamic process. This can be observed in the NO and NO_2 series, where narrow peaks at elevated concentrations are superposed over a slowly fluctuating baseline. From the dynamics and absolute values of Figure 3, the following can be learned:

- **Precision and accuracy of sensor measurements:** Regarding NO_2, both peaks and background concentrations as measured by the sensor consistently remained above the detection limit. In contrast, the background concentrations of NO frequently approach or even fall below the detection limit, but detectable peaks occur regularly at elevated concentrations. The most obvious difference with the reference measurements is that the NO and NO_2 sensors severely underestimate the peak maxima. This is in contrast with the field calibration in Figure 2b where the in-situ calibration overestimates the concentrations. This is another indication that the calibration in laboratory conditions cannot be extrapolated to other situational contexts in a simple way. The underestimation introduces an uncertainty when the measurements are compared with health-related thresholds. This uncertainty can be reduced by improving the calibration of the gas sensors;
- **Reliability of the dynamic pattern:** The field campaign (see Figure 1a) has shown that peaks superposed on a slowly fluctuating baseline can be identified and that these peaks are observed by the low-cost NO_2 gas sensor. Also, in the measurement campaign on board the ship, a dynamic pattern consisting of tall peaks superposed on a baseline is observed. The position of the peaks as measured with the low-cost NO and NO_2 gas sensors are in register with the ones observed in the reference measurements. Moments where such peaks occur are characterized by a poorer indoor air quality. The dynamics in the structure of the time series contain valuable information and for that reason it is worthwhile to perform monitoring campaigns with a high temporal resolution;
- **Occurrence of events:** The peaks in the time series can be interpreted as events corresponding to instances when the ship's exhaust emissions or the exhaust gas from a passing ship enter the engine room via the ventilation inlet. In most cases, NO and NO_2 peaks occur concurrently. However, events where only one of the NO or NO_2 peaks occur have been observed as well. For the periods that both peaks occur simultaneously, the NO_2/NO ratio varies from 0.16 to 1.5 and in the majority of cases, the peak maximum of NO surpasses that of the corresponding NO_2 peak. While the synchronicity of most NO and NO_2 peaks implies a common pollution source, the variable ratio suggests that this source generates a diverse mixture of pollutants. This brief study suggests that the distribution of valuable information in the time series is not homogeneous. Events contain valuable information about the pollution source and moments of poorer air quality. Therefore, it seems worthwhile to develop mathematical methods that can extract and analyze events in time series.

III.3. MEASURING CAMPAIGN IN CUBA

The measurement campaign conducted in Cienfuegos, as depicted in Figure 4, has been calibrated using a combination of low-cost methods and VITO calibrations. The structure of the temperature and relative humidity exhibit peaks and valleys in counterphase. Obvious differences with campaigns in Belgium are the elevated levels of O_3 and SO_2. The high concentrations of SO_2 are attributed to the combustion of sulfur-containing fuels, a type

of fuel that is not used in the Western world (except for seagoing ships). In the case of NO_2, a prominent peak reaching up to 200 ppb is evident at the onset of the campaign, which suppresses the minor fluctuations in the remainder of the time series. The NO_2 peaks coincide with the ones of relative humidity. It is unknown if this is due to an environmental cause or an instrumental artifact. The average concentration of NO_2 (47 ± 16 ppb) is higher than the average value of the Belgian field campaign (19 ± 11 ppb).

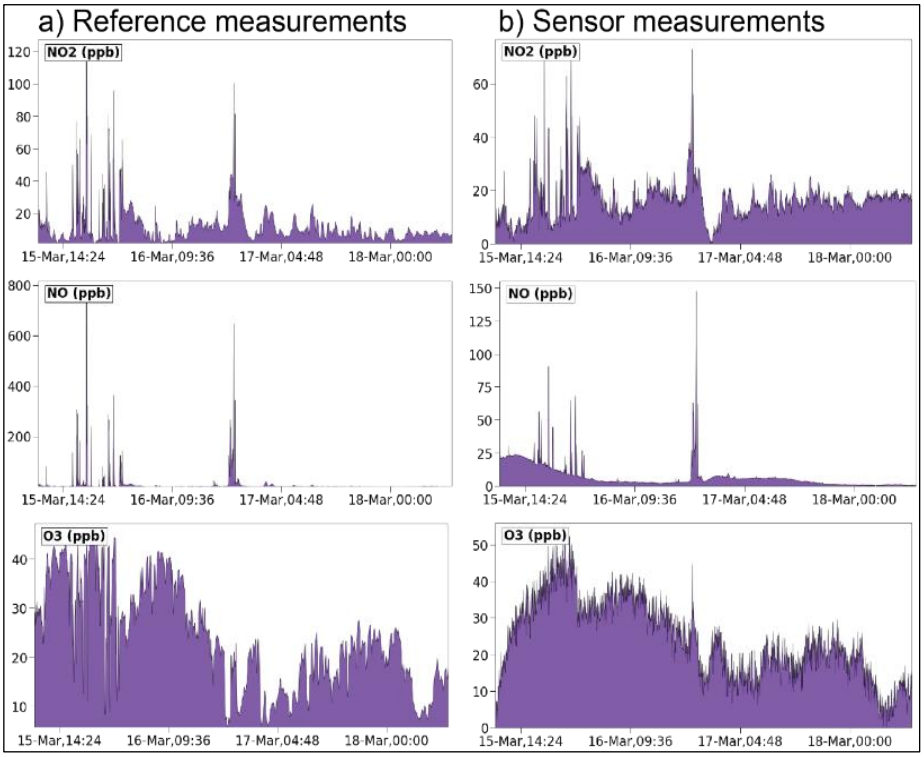

Figure 3: Measurement campaigns of NO_2, NO and O_3 at the same location in the engine room. The measuring campaign covers the period March 15, 2021 – March 18, 2021; (a) 1-minute data collected by the Airpointer containing reference instruments; (b) 3-minute data collected by the Raspberry Pi based data logger.
Source: Authors, (2024).

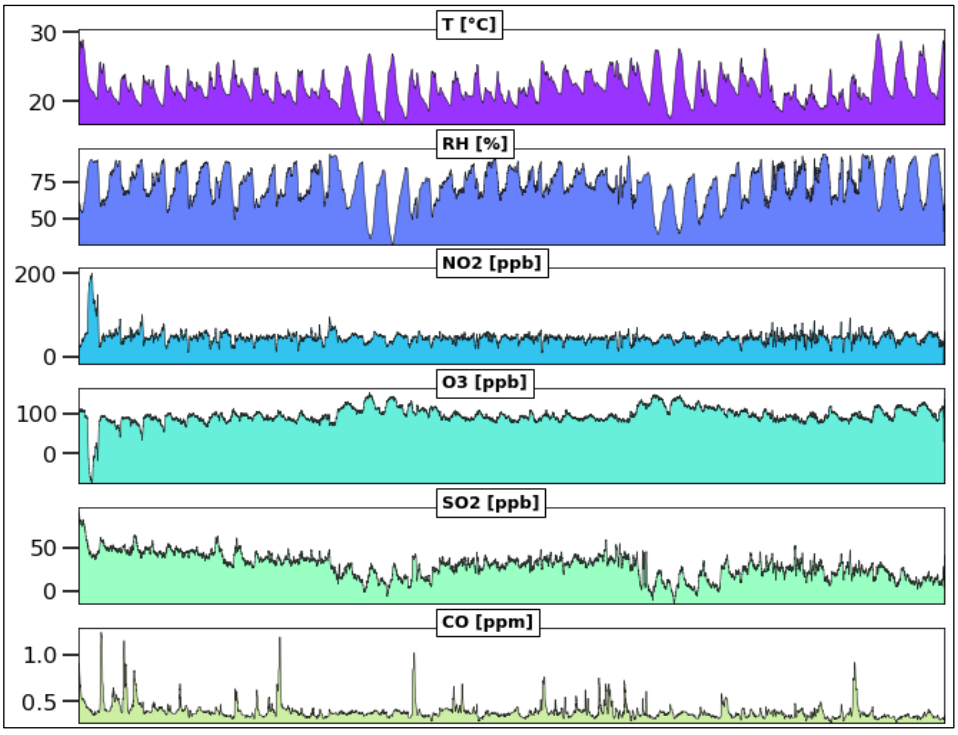

Figure 4: Measurement campaign performed at Cienfuegos, Cuba with the low-cost monitoring system from March 14 to April 22, 2022.
Source: Authors, (2024).

V. CONCLUSIONS

The coefficient of determination of the linear calibration in the field campaign is lower due to variable climatic conditions. This study has shown that the calibration of low-cost gas sensors in laboratory conditions result in linear calibration curves with a coefficient of determination close to 1. Despite this good correlation, intercept and slope appears to vary over time, with the situational context, the sensor's history, and from sensor to sensor. The situational sensitivity introduces an additional uncertainty in the measurement of pollutant concentration.

The dynamic patterns observed in time series measured by low-cost gas sensors appear to be sufficiently reliable to extract meaningful information. Specifically, the NO gas sensor appears to provide valuable insights by detecting peaks that surpass the detection limit. The peaks in time series contain elevated amounts of information regarding both pollution source and instances of poorer air quality.

Future research is necessary to enhance our understanding of gas sensor behavior in zero air under varying conditions of temperature, relative humidity, and pressure. This will lead to an improved accuracy of zero calibration (i.e., intercept of the calibration curve). In addition, also the effect of environmental parameters on the slope of the calibration curve should be studied. Such calibration experiments might be hampered by an equilibrium reaction of NO_2, NO and SO_2 with moisture, resulting in the formation of acids that might not be detectable by the respective sensor. Considering the relatively low reactivity of CO under ambient conditions, it is advisable to assess the span calibration for this gas first. Additionally, it is important to evaluate the replicability of zero and span calibrations by conducting a series of repeated experiments using zero air and a constant target gas concentration. The scientific literature contains numerous publications on the calibration of low-cost gas sensors, illustrating the importance of this topic.

VI. AUTHOR'S CONTRIBUTION

Conceptualization: Olivier Schalm.
Methodology: Rosa Amalia González-Rivero, Erik Hernández-Rodríguez, Mayra C. Morales-Pérez, Daniellys Alejo-Sánchez, Alain Martinez
Investigation: Olivier Schalm, Rosa Amalia González-Rivero, Erik Hernández-Rodríguez
Discussion of results: Olivier Schalm
Writing – Original Draft: Olivier Schalm.
Writing – Review and Editing: Olivier Schalm, Werner Jacobs, Luis Hernández
Supervision: Olivier Schalm
Approval of the final text: Olivier Schalm

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