Evaluation of optical particulate matter sensors under realistic conditions of strong and mild urban pollution

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Abstract. In this paper we evaluate characteristics of three optical particulate matter sensors/sizers (OPS): high-end spectrometer 11-D (Grimm, Germany), low-cost sensor OPC-N2 (Alphasense, United Kingdom) and in-house developed MAQS which is based on another low-cost sensor – PMS5003 (Plantower, China), under realistic conditions of strong and mild urban pollution. Results were compared against a reference gravimetric system, based on Gemini (Dadolab, Italy), 2.3 m^3/h air sampler, with two channels (simultaneously measuring PM$_{2.5}$ and PM$_{10}$ concentrations). The measurements were performed in Sarajevo, the capital of Bosnia-Herzegovina, from December 2019 until May 2020. This interval is divided into period 1 - strong pollution (December 2019 – March 2020) and period 2 - mild pollution (March 2020 – May 2020). The city of Sarajevo is one of the most polluted cities in Europe in terms of aerosols: the average concentration of PM$_{2.5}$ during the period 1 was 83 μg/m^3, with daily average values exceeding 500 μg/m^3. During period 2, the average concentration of PM$_{2.5}$ was 20 μg/m^3. These conditions represent a good opportunity to test optical devices against reference instrument in a wide range of ambient particulate matter (PM) concentrations. The effect of an in-house developed diffusion dryer for 11-D is discussed as well. In order to analyze the mass distribution of aerosols, a scanning mobility particle sizer (SMPS), which together with the 11-D spectrometer gives the full spectrum from nanoparticles of diameter 10 nm to coarse particles of diameter 35 μm, was used. All tested devices showed excellent correlation with the reference instrument in period 1, with R^2 values between 0.90 and 0.99 for daily average PM concentrations. However, in period 2, where the range of concentrations was much narrower, R^2 values decreased significantly, to values from 0.28 to 0.92. We have also included results of a 13.5 month long-term comparison of our MAQS sensor with a nearby beta attenuation monitor (BAM) 1020 (Met One Instruments, USA) operated by the United States Environmental Protection Agency (US EPA), which showed similar correlation and no observable change of performance over time.

1 Introduction

Analysis of aerosols represents a key element for the studies of air pollution. Various studies shed light on their effect on health (Downward et al., 2018) and climate (Zhao et al., 2019). In many cases aerosols are a dominant pollutant among other components of pollution. Therefore, developing a strategy for reliable quantification of aerosols in ambient air is necessary. The traditional and most accurate approach to measuring the particulate matter concentration in the air is the reference method,

25 based on gravimetric measurements, after the collection of particulate matter by air samplers. The typical time resolution of such measurements is 24 hours. Although there are portable air samplers, these measurements are usually performed at fixed locations, such as research supersites. Reference systems are expensive and require a lot of laboratory work. Results are not immediately available, because of the time-consuming process of filter treatment. Taking that into account, various governmental institutions usually opt for more affordable and easier to use and maintain – equivalent methods. These are usually

30 fixed, semi-automatic stations equipped with BAM. The typical time resolution of such stations is 1 hour. If maintained and calibrated properly, the equivalent methods should achieve an acceptable level of agreement with the reference. For example, one long-term comprehensive study (Hafkenscheid and Vonk, 2014) performed at 14 different locations across Netherlands, showed that a linear correction $y = 0.91x - 1.6$, applied on raw readings from BAM, was necessary to achieve the requirements of the Guide to the Demonstration of Equivalence (ECWG, 2010).

35 Newer methods, based on optical particle sensors (OPS), are nowadays increasingly more popular, particularly low-cost variants (Zheng et al., 2019; Mukherjee et al., 2019; Tanzer et al., 2019; Morawska et al., 2018). Their typical time resolution is between 1 s and 1 min, and because of their price and size, they can be used in networks to provide better spatial coverage (Martin et al., 2019; Li et al., 2019). Furthermore, they provide information about multiple mass fractions of particulate matter simultaneously, unlike the concentration of single fraction in gravimetric system or BAM. However, there are concerns about

40 their suitability for measuring mass concentrations of ambient PM, since there is a significant measurement uncertainty arising from the principles of their operation.

Most commercially available OPS use the Mie scattering theory (Mie, 1908) to determine the size and number of particles within the unit volume of air. The Mie theory provides the solution of the Maxwell equations for the scattering of plane waves on spherical particles. The Mie solution is rather complex, but in order to illustrate the non-linearity of the theory, it will suffice

45 to consider the case where particles are much smaller than the wavelength (of light, since a red laser is commonly used in practice). In that case the intensity of scattered radiation is given by

$$I = I_0 \frac{1 + \cos^2 \theta}{2R^2} \left(\frac{2\pi}{\lambda} \right)^4 \left(\frac{d}{2} \right)^6 \left| \frac{m^2 - 1}{m^2 + 2} \right|^2, \tag{1}$$

where I_0 is intensity of the incident radiation, θ is the scattering angle, R is the distance between the particle and the observing point, λ is the wavelength, d is the particle diameter and m is the refractive index of the particle. Thus, in order to calculate the

50 diameter of the particle by measuring the intensity of the scattered radiation, one must assume a value for the refractive index of the particle. If the particle absorbs nothing from incoming radiation, its refractive index will be real, otherwise it is written in the form

$$m = n + i\kappa, \tag{2}$$

where κ is called the extinction coefficient and is related to the absorption coefficient α:

55 $$\alpha = \frac{4\pi\kappa}{\lambda}. \tag{3}$$

Once the size distribution is calculated accross K channels (bins), the total mass concentration of particles will be

$$c_m = \sum_{i=1}^{K} w_i \rho_i V_i N_i, \tag{4}$$

where V_i is the (average) volume, ρ_i is the density of the particles, N_i is the number of particles per unit volume and w_i is the weighting factor for channel i. Here we have another cause of OPS uncertainty: the density of particles must be assumed.

60 Regarding the weighting factors, sensor manufacturers calculate values to correct for certain effects, such as the fact that OPS cannot detect particles which are too small.

Laboratory tests and calibrations of OPS are performed under controlled conditions with known aerosols, such as polystyrene latex spheres (Walser et al., 2017; Bezantakos et al., 2018), continuously changing monodisperse, particles (Kuula et al., 2017, 2020) or multi-modal aerosols (Cai et al., 2019). Burning chamber is used in some investigations as well (Wang et al., 2015).

65 However, equation (1) is strongly non-linear in terms of refractive index, and in most practical cases corrections for different aerosol optical properties are impossible to implement. Furthermore, densities appearing in equation (4) are not known *a priori*. That explains why it is difficult to calibrate OPS for realistic ambient PM concentration measurements: any laboratory calibration may or may not be applicable to the changing outdoor conditions (Tryner et al., 2020; Crilley et al., 2020).

For outdoor applications, there is an additional problem: hygroscopic growth of aerosols (Jayaratne et al., 2018; Granados-
70 Muñoz et al., 2015; Di Antonio et al., 2018), which leads to overshoots of OPS if the ambient air humidity is (too) high. An obvious solution is to dry the air. However, any proper drying system would cost more than many models of OPS and it is rarely seen in combination with low-cost sensors. Analytical corrections are often used: humidity sensors are used to measure the relative humidity of ambient air and some analytical model, like Kohler's theory (Castarède and Thomson, 2018) or Hänel equation (Hänel, 1976), is applied. Later in this paper we will make some observations on this issue.

75 Due to all the above-mentioned factors, it is always interesting to check how OPS perform in different realistic scenarios. Numerous papers deal with laboratory calibrations and outdoor evaluations of OPS (Karagulian et al., 2019; Borghi et al., 2018; Chatzidiakou et al., 2019; Magi et al., 2020; Sousan et al., 2016b; Malings et al., 2020; Kelly et al., 2017; Sayahi et al., 2019; Crilley et al., 2018; Zheng et al., 2018; Tasic et al., 2012; Cavaliere et al., 2018; Mukherjee et al., 2017; Sousan et al., 2016a; Zhang et al., 2018; Holstius et al., 2014; Badura et al., 2018). Reported results vary depending on the composition of
80 aerosol pollution, range of concentrations and meteorological factors.

The novelty of this research is a unique combination of instruments and conditions of extremely high urban pollution. The city of Sarajevo is situated in a valley and is affected by strong temperature inversions that appear typically 150 m-300 m above ground level with a very strong temperature gradient in the inversion layer, exceeding 30 K/km (Masic et al., 2019). The inversion episodes were present during most of January 2020. As a consequence, the average monthly concentration of
85 $PM_{2.5}$ was very high: 167.3 $\mu g/m^3$. In contrast to that, monthly average values for March and April 2020 were 21.6 $\mu g/m^3$ and 19.6 $\mu g/m^3$, respectively. This presented an excellent opportunity to test the performance of OPS in very different pollution levels. Simultaneously with OPS and reference gravimetric measurements, an SMPS was employed to detect nanoparticles. It can detect particles with diameters from few nm up to 1 μm. While SMPS can count very small particles, 11-D can count larger particles, from 0.25 μm to 35 μm in diameter. When they work simultaneously, they can detect (almost) the full range of

90   aerosol diameters, with a span of more than three orders of magnitude. This will give detailed insights on the mass distribution
       of aerosols.

2   Methodology and experimental setup

The experimental facility was located at the Faculty of Mechanical Engineering in the central part of Sarajevo valley (N
43.85424, E 18.39607, 540 m above sea level) and represents well the overall conditions in the city. The reference instrument

95   for measurements of PM concentrations was a Dadolab Gemini air sampler. It is a single device with two completely indepen-
       dent channels ($PM_{2.5}$ and PM_{10} in this campaign). The filter preparation and gravimetric analysis are performed in separate
       laboratory of Faculty of Science, Department of Chemistry. The air sampler, gravimetric laboratory and all filter procedures
       satisfied requirements of the standard EN 12341:2014.

Grimm 11-D is a high-end optical particle sizer, with sophisticated construction and ability to count individual particles

100   from 250 nm to 35 μm in 31 equidistant (on logarithmic scale) channels. It uses a proprietary algorithm and the manufacturer
       does not share information about the refractive index, density or weighting factors. It was factory calibrated, and equipped with
       firmware version 12.50. Data was recorded in 1 minute intervals (6 seconds is also possible). Since we use the common term
       OPS occasionally, it should be noted that 11-D belongs to different category of devices (in comparison to low-cost sensors).

Alphasense OPC-N2 belongs to the category of low-cost sensors. The manufacturer transparently shared most specifications.

105   It has a much simpler construction than 11-D: instead of regulated pump, air flow is provided by 25 mm fan. The device has
       16 channels, from 380 nm to 17 μm. Firmware version 18.2 was used. Refractive index was $n = 1.50 + i0$ and density was
       1.65 g/cm^3. All other parameters, including weighting factors, were used as firmware default values.

The Plantower PMS5003 could be termed a very low-cost sensor, since its price is lower by an order of magnitude than that
       of the OPC-N2. Limited specifications don't reveal all operating parameters. From the specification sheet we can conclude that

110   the device uses Mie scattering theory, with detection limit of 300 nm, and has 6 channels. It is likely that PMS5003 works in
       photometric mode, rather than counting individual particles. By disassembling one PMS5003 sensor we have discovered some
       components: red semiconductor laser which pulsates approximately once per second, photodetector and 32-bit processor. Air
       flow is provided by a 20 mm fan. The PMS5003 has two data outputs, one is called SM (standard material, CF = 1), and another
       AE (atmospheric environment), without further explanation from the manufacturer. The latter mode is used in our work. Based

115   on PMS5003, we have designed MAQS (Mobile Air Quality System) smart sensor. Essentially, it is a modular platform for
       PMS5003, with options for additional sensors (pressure, temperature, humidity, carbon dioxide, wind speed), GNSS receiver,
       flash memory, Wi-Fi module and 3D-printed enclosure. Eight MAQS sensors were made and tested prior to the main campaign
       in order to evaluate consistency between units. Figure 1 shows the results of preliminary outdoor measurements for a batch of
       8 MAQS sensors. They showed very good consistency: the coefficient of determination, R^2, between any two sensors from

120   the batch was greater than 0.99 and average readings from all sensors are within $\pm 10\%$ from the average value of the batch of
       sensors. Data was recorded every minute on a local SD card and remote cloud server simultaneously. Recording interval can
       be as short as 1 second, but there was no need for that.

Grim 11-D and Alphasense OPC-N2 could not be used outdoors without shelter, while MAQS has a special case which provides basic protection for outdoor use. Furthermore, netbook PC was used to record data from the OPC-N2. Outdoor shelter

125 had to be constructed to accommodate 11-D with power supply, OPC-N2 with PC and SPI adapter, and MAQS (for better protection). Stevenson screen like wooden structure was designed for that purpose. Another MAQS sensor was used at a remote location, for reasons that will be explained later.

For low-cost sensors (OPC-N2 and MAQS) there was no air dryer or heater, since they are typically used in such conditions. We have designed and constructed diffusion dryer for application on 11-D, which consists of porous stainless steel tube

130 surrounded with 1 kg of silica gel. The dryer is compact, 25 cm in length with 8 cm external diameter and does not reduce the mobility of the instrument. It was installed only during the period of mild pollution.

Meteorological parameters were measured using Vantage Pro2 (Davis Instruments, USA) weather station with recording intervals of 15 minutes.

SMPS is a complex system which consists of a condensation particle counter (CPC), a differential mobility analyzer (DMA)

135 and a charge conditioner (often inadequately called "neutralizer"). Depending on the characteristics of the DMA, the SMPS can be configured for certain span of particle diameters. We have used Grimm 5.416 high-end SMPS with long DMA which is able to separate particles from 10 nm to 1000 nm in 129 channels, equidistant on a logarithmic scale. Despite the fact that aerosols with diameter below 10 nm play an important role in nucleation and growth studies (Tiszenkel et al., 2019), their contribution to the mass budget is negligible. Another (larger) in-house developed diffusion dryer was installed at the inlet of

140 SMPS. A soft X-ray device was used as the charge conditioner. Scanning mode (alternating upscan and downscan) was used for all measurements. One scan takes about 4 minutes (8 minutes for both upscan and downscan). When working parallelly, SMPS and 11-D form a powerful wide-range spectrometer, which covers a range of aerosol diameters from 10 nm to 35 μm in 160 channels. Additionally, there is an overlapping area between 250 nm and 1000 nm where we can see how well these two instruments match. The complex SMPS system was kept indoors (an unavoidable necessity, since both X-ray charger and

145 DMA use very high operating voltages). The air was sampled from outside using a conductive tube of shortest possible length, to avoid aerosol losses. It was running continuously, except for the periods of maintenance.

A rigorous data validation procedure was used. All instruments were inspected periodically and data logs were analyzed thoroughly. When calculating daily average values, complete and consistent data series were required.

3   Results and discussion

150 During this campaign 296 filters were used in the reference air sampler. After the removal of several blank filters used for periodic verification and those with incomplete sampling (pneumatic system of air sampler failed to load new filters automatically couple of times), 288 filters remained: 143 $PM_{2.5}$ and 145 PM_{10} samples. Figure 2 shows $PM_{2.5}$ and PM_{10} daily average concentrations, together with hourly and daily values of ambient air temperature and relative humidity.

Some modifications of the shelter for 11-D and OPC-N2 were necessary, making those instruments unavailable periodically

155 during December and January. Additionally, more frequent maintenance, such as cleaning of 11-D, was needed when working

in extreme conditions. The same stands for SMPS, which was maintained according to the recommendations of the manufacturer. Taking into account difficult operating conditions, the amount of data collected is satisfactory during the period of strong pollution and excellent during the period of mild pollution.

The lower limit of detection (LLoD) of $PM_{2.5}$ concentration for evaluated optical aerosol devices is estimated based on their
160   actual field performance using standard methodology (3σ), and the results are: $5\,\mu g/m^3$ for PMS5003, $2\,\mu g/m^3$ for OPC-N2 and $1\,\mu g/m^3$ for 11-D. These values are comparable to those reported by Bulot et al. (2019).

3.1   Strong urban pollution

During the period of strong urban pollution (12/2/2019–3/12/2020), the average value of $PM_{2.5}$ concentration was $82.9\,\mu g/m^3$, with minimum daily average value $1.3\,\mu g/m^3$ and maximum value $504.9\,\mu g/m^3$. In the same period, the average PM_{10} con-
165   centration was $95.5\,\mu g/m^3$, with minimum value $3.6\,\mu g/m^3$ and maximum value $549.0\,\mu g/m^3$. The ratio of average values of concentrations $PM_{2.5}/PM_{10}$ was 0.87. Very good correlations were observed for all three OPS against the reference instrument. Such a range of ambient PM concentrations was favorable for achievement of high R^2 values, but non-linear effects of low-cost sensors were observed too.

Grimm 11-D produced results with R^2 values 0.988 and 0.985 for $PM_{2.5}$ and PM_{10} concentrations, respectively. Absolute
170   values were larger than the reference, on average 17.6% for $PM_{2.5}$ and 25.5% for PM_{10}. The average ratio $PM_{2.5}/PM_{10}$ measured by 11-D was 0.93.

Alphasense OPC-N2 undershoots with respect to the reference values, on average 31.0% for $PM_{2.5}$ and 36.8% for PM_{10}, but R^2 coefficients are relatively high: 0.903 and 0.920 for $PM_{2.5}$ and PM_{10} respectively. The OPC-N2 measured the ratio $PM_{2.5}/PM_{10}$ to be 0.97.
175   MAQS sensor produced surprisingly good R^2 values of 0.975 for $PM_{2.5}$ and 0.950 for PM_{10}. In terms of absolute values, it overshoots by 31.9% for $PM_{2.5}$ and 49.3% for PM_{10} (on average). The calculated ratio $PM_{2.5}/PM_{10}$ was 0.76. It seems that the Plantower PMS5003 can not accurately determine the PM_{10} fraction. One possible explanation is provided by a laboratory test of PMS5003, where it was found that its size bin [$2.5\,\mu m$-$10\,\mu m$] is noisy and inaccurate (Kuula et al., 2020). Further investigation of this behavior would be useful.
180   None of the tested OPS were equipped with an air dryer, and this certainly contributes to overprediction. Yet, Alphasense OPC-N2 with default firmware settings underpredicts values, despite the aerosol hygroscopic growth effect. Figure 3 shows comparisons of all three OPS against reference for $PM_{2.5}$ and PM_{10} during the period of strong pollution.

3.2   Mild urban pollution

The correlation coefficients changed dramatically in the period of mild pollution (3/13/2020–5/4/2020), as Figure 4 shows.
185   The much narrower range of particulate matter concentrations plays an important role, and even the reference method is less accurate, since the mass difference of loaded and blank filters becomes very small (smaller than 1 mg for 24 h sampling period if PM concentration is below $18\,\mu g/m^3$). The average concentration of $PM_{2.5}$ was $19.7\,\mu g/m^3$ with a minimum daily average value of $7.1\,\mu g/m^3$ and a maximum value of $39.3\,\mu g/m^3$. During this period, the average value of PM_{10} concentration was

24.2 μg/m^3, with minimum and maximum values of 7.6 μg/m^3 and 48.8 μg/m^3, respectively. The ratio PM$_{2.5}$/PM$_{10}$ was 0.81
190 on average.

This time Grimm 11-D was equipped with a dryer, whose effects will be discussed in the next subsection. The device
produced relatively high R^2 values 0.868 for PM$_{2.5}$ and 0.917 for PM$_{10}$. The absolute readings underestimated concentrations
of PM$_{2.5}$ by 16.3% on average, while PM$_{10}$ were underestimated by 10.9% on average. The PM$_{2.5}$/PM$_{10}$ ratio was 0.87. This
test clearly shows that 11-D is a completely different class of instrument (in comparison to low-cost sensors). When equipped
195 with dryer, 11-D shows level of performance comparable to BAM, at least those reported by Hafkenscheid and Vonk (2014).

Alphasense OPC-N2 did not perform well during the period of mild pollution. Coefficients of determination, R^2, were only
0.284 for PM$_{2.5}$ and 0.525 for PM$_{10}$. Absolute readings are worrying: the OPC-N2 underpredicted PM$_{2.5}$ by 67.6% and PM$_{10}$
by 71.6% on average. The ratio PM$_{2.5}$/PM$_{10}$ was 0.73.

MAQS sensor demonstrated mediocre performance, with R^2 values of 0.730 for PM$_{2.5}$ and 0.718 for PM$_{10}$. On average,
200 this sensor overpredicted PM$_{2.5}$ by 30.5% and PM$_{10}$ by 32.6%. The PM$_{2.5}$/PM$_{10}$ ratio was 0.83, very close to the reference
value (contrary to the performance of the sensor in the period of strong pollution).

It would be interesting to test low-cost sensors with a proper dryer as well, but that combination is rarely seen in practice.

3.3   Humidity influence

One of the important factors in ambient measurements of PM concentrations is humidity, since the particles reflect more light
205 (i.e. appear larger) during measurements due to hygroscopic growth. This can be described using Hänel equation:

$$f_\zeta(RH) = \left(\frac{1 - RH}{1 - RH_{\text{ref}}} \right)^{-\gamma}, \tag{5}$$

where f_ζ is enhancement factor for aerosol property ζ. Here RH represents the relative humidity and RH_{ref} is a reference
relative humidity:

$$f_\zeta(RH) = \frac{\zeta(RH)}{\zeta(RH_{\text{ref}})}. \tag{6}$$

210 It is important to note that the coefficient γ, which is an indicator of the hygroscopicity of aerosol, depends on the type of
aerosol (and changes whenever composition of ambient aerosols is changed).

If we compare results produced by 11-D relative to the reference, during period 1 (without dryer) and period 2 (with dryer),
we can see that readings of the 11-D were reduced by more than 30%. However, we can not conclude whether it was the
effect of the dryer or the consequence of significantly different ambient conditions. Unfortunately, we have only one 11-D,
215 so we couldn't measure simultaneously with and without dryer (that's the reason why we used the instrument with the dryer
only in one of the two periods). If we take into account two intervals with similar ambient conditions, with and without dryer
we get following values: from 2/27/2020 to 3/12/2020 average ambient concentration of PM$_{2.5}$ was 21.1 μg/m^3 while 11-D
(without dryer) measured 21.5% more. In the second interval, from 3/13/2020 to 4/1/2020, ambient concentration was similar,
21.0 μg/m^3 while 11-D (with dryer) measured 1.4% smaller value. This comparison indicates that the effect of the dryer could
220 be around 23%. A similar analysis for PM$_{10}$ concentrations gives an estimate of about 20% for the dryer effect.

Grim 11-D has a very useful feature: internal temperature and humidity sensor. Figure 5 shows self-heating and diffusion dryer effect on 11-D, by comparing internal and external measurements of temperature and humidity. The average ambient air temperature from 2/27/2020 to 4/1/2020 was $7.02\,^0$C while the average 11-D internal temperature was $14.27\,^0$C, which shows a significant difference of $7.25\,^0$C. This self-heating effect reduces internal humidity significantly, and we can see that it

225 rarely goes beyond 50%. Once the dryer is installed, internal relative humidity is further reduced: the average value of internal humidity without dryer (2/27/2020-3/12/2020) was 36.2% and with dryer (3/13/2020-4/1/2020) was 21.8% (the ambient air humidity also dropped in the later period, but nevertheless the effect of the dryer is evident).

After roughly a month, the dryer's performance degraded and the silica gel needed a regeneration (but it wasn't performed since we didn't want to interrupt measurements when the end of the campaign was near).

230 ### 3.4 SMPS data and wide-range spectrometer

The wide-range spectrometer (SMPS+11D) produced very valuable results. Figure 6 shows the continuous concentration and mass distributions. It is created from hourly average measurements from SMPS and 11-D. A relative density of 1.65 was applied in SMPS software (based on LabVIEW) for the mass calculation. No other corrections were performed and all settings were factory defaults. Selected histograms (hourly average values) are shown in Figure 7. What we can see from figures 6 and

235 7 is that in the period of strong pollution the dominant mass contribution comes from particles with diameters around 300 nm. In terms of concentrations, particles around 100 nm appear in greatest numbers, with occasional secondary peaks coming from even smaller particles.

In the period of mild pollution, however, we can see that particles larger than 2.5 μm often appear on histograms (usually about 3 μm in diameter). Number concentrations still have peaks about 100 nm, but sometimes the distribution is different in

240 favor of even smaller particles, as Figure 7 shows. Again, the largest mass contribution comes from particles around 300 nm.

In the overlapping area, SMPS and 11-D matched very well, almost perfectly for concentrations. Their match was not as good for mass calculations, but that is understandable, taking into account all the factors explained in section 1. Overall, the combination of SMPS and 11-D worked very well and gave the full spectrum of aerosols, both for number concentrations and mass distribution.

245 The obtained mass distribution of particles, especially during the period of strong pollution, rises the question on suitability of OPS for measurements of mass concentrations, and resolving different fractions, since they cannot detect small particles that significantly contribute to the total mass. For example, the Alphasense OPC-N2 has a detection limit of 380 nm, and is likely to miss the particles around 300 nm which form the dominant contribution to the mass budget. The Grimm 11-D, with a detection limit of 250 nm, has a far better potential to resolve mass fractions.

250 ### 3.5 Long-term performance

Another question about OPS, especially low-cost types, is the drift of performance over time. The PMS5003 sensor uses a semiconductor laser (diode laser) which has a limited lifetime. We have some long-term comparisons of the MAQS sensor with MetOne BAM-1020 operated at a nearby location by the US EPA. Strictly speaking, their station is not collocated with

our equipment, but for the distance of only 300 m it is reasonable to assume that the air composition is very similar at these two

255   points, since they are located in the same neighborhood. In order to verify that assumption, we have installed another MAQS sensor at the location of Faculty of Electrical Engineering, University of Sarajevo, which is in immediate vicinity of the US EPA site, and at the same distance from us (about 300 m). Figure 8 shows long-term comparisons of MAQS sensor and BAM-1020, and additional verification of correlation between readings of two MAQS sensors, which was very high ($R^2 = 0.970$ for hourly average values and $R^2 = 0.994$ for daily average values) confirming our assumption that these two locations share the

260   same air, in terms of PM concentrations and properties.

       During 13.5 months of continuous comparisons, the correlations for hourly, daily and monthly average values of PM2.5 are 0.919, 0.980 and 0.998, respectively, with absolute values overestimated by 20% on average. This leads us to the conclusion that time averaging reduces a lot the influence of variation of aerosol composition and meteorological variations. If we use a longer time average period, we lose one of the major advantages of low-cost sensors (time resolution), but it is a more natural

265   approach to correcting readings compared to using artificial algorithms like neural networks (Badura et al., 2019) or machine learning (Si et al., 2020). An excellent viewpoint of this issue is given by Hagler et al. (2018). The calibration of a larger number of low-cost sensors can be simplified if they show similar relative performance (to each other) in laboratory and field (Sousan et al., 2018). Floating corrections, even physically justifiable interventions, such as the instantaneous correction for humidity growth of aerosols, insert a lot of noise, and the benefit is questionable. Depending on ambient conditions, self-

270   heating of the sensor and some other factors, relative humidity may not be accurately determined. Even if we have a very accurate humidity measurement, the hygroscopic growth coefficient will change whenever the composition of aerosol changes, inevitably injecting noise into the results.

       We can also see strong non-linear effects at very high concentrations, above $500 \, \mu\text{g/m}^3$. In that case a quadratic regression fit will be more suitable.

275   During this period of 13.5 months of continuous outdoor operation, the MAQS sensor worked flawlessly without performance drifts. Designed enclosure sufficiently protected the sensor outdoors, while not obstructing air sampling.

4   Conclusions

A comprehensive experimental study was carried out with the aim of evaluating the performance of three very different OPS: high-end Grimm 11-D, low-cost Alphasense OPC-N2 and in-house developed MAQS sensor, which is based on another low-

280   cost sensor, the Plantower PMS5003 sensor. The study was performed in realistic conditions of strong and mild urban pollution. The reference instrument was a dual-channel air sampler with gravimetric analysis in separate laboratory. In total 288 filters were collected from 12/2/2019 to 5/4/2020.

       During the period of strong urban pollution all three instruments produced very high R^2 values. However, during the period of mild urban pollution, these correlation factors dropped significantly, especially for Alphasense OPC-N2 sensor measuring

285   PM2.5 parameter. The OPC-N2 underestimated the mass concentrations badly, especially during the period of mild pollution.

MAQS sensor overshoots PM$_{2.5}$ concentrations by approximately 30% on average, which is likely to be partially caused by hygroscopic growth.

The wide-range spectrometer, which consists of SMPS and 11-D, produced valuable information about distribution of aerosols, both in number and mass concentrations. Particles with diameters around 100 nm (and sometimes below) repre-
290   sent the dominant fraction in pure numbers, while particles with diameter of around 300 nm give the highest contribution to mass. In the period of mild pollution, particles larger then 2.5 μm gave a larger contribution than in the period of strong pollution.

Grimm 11-D performed well in all conditions, and when equipped with dryer, it performed at a comparable level to the beta attenuation monitor. For the calibration of low-cost sensors, especially those based on PMS5003, we propose a linear or
295   quadratic correction (in case of high pollution levels) with steady coefficients, since the instantaneous corrections insert noise into results.

Future measurements should further investigate characteristics of OPS in different ambient conditions, influence of humidity and effect of micro-dryers specifically designed for low-cost sensors and mass distributions by means of wide-range spectrometer.

300   *Data availability.* The underlying datasets for this publication are available at https://doi.org/10.5281/zenodo.3897379

Furthermore, data from the BAM measurements by US EPA are availble at https://cfpub.epa.gov/airnow

Author contributions. AM participated in all phases of this research and wrote the manuscript with contributions from all co-authors. DB and BP performed field work together with AM. AB designed, manufactured and analysed the performance of two diffusion dryers and evaluated the influence of humidity on the readings of 11-D. SZ and JH performed gravimetric measurements, including preconditioning and
305   treatment of the filters, ensuring strict fulfilment of the standard EN 12341:2014

Competing interests. The authors declare that they have no conflict of interest.

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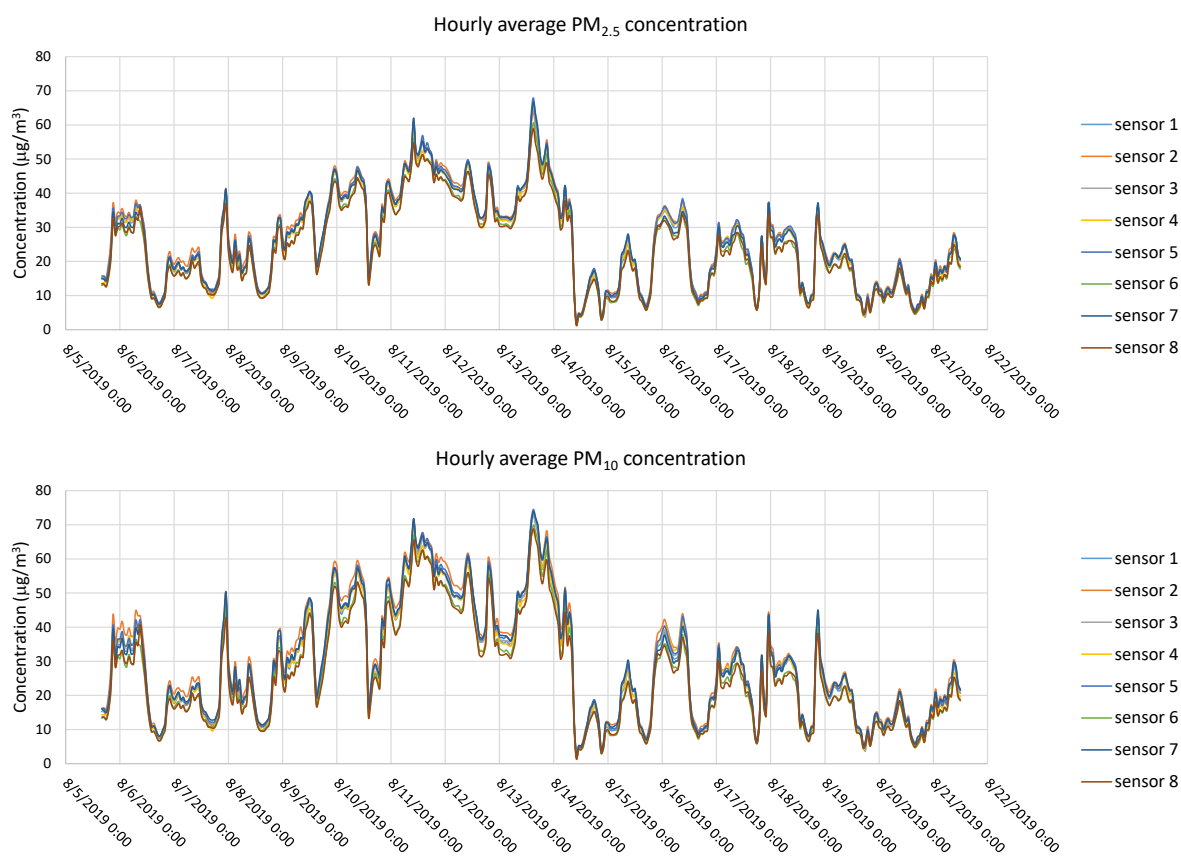
Figure 1. Preliminary test of 8 MAQS sensors, outdoor measurements.
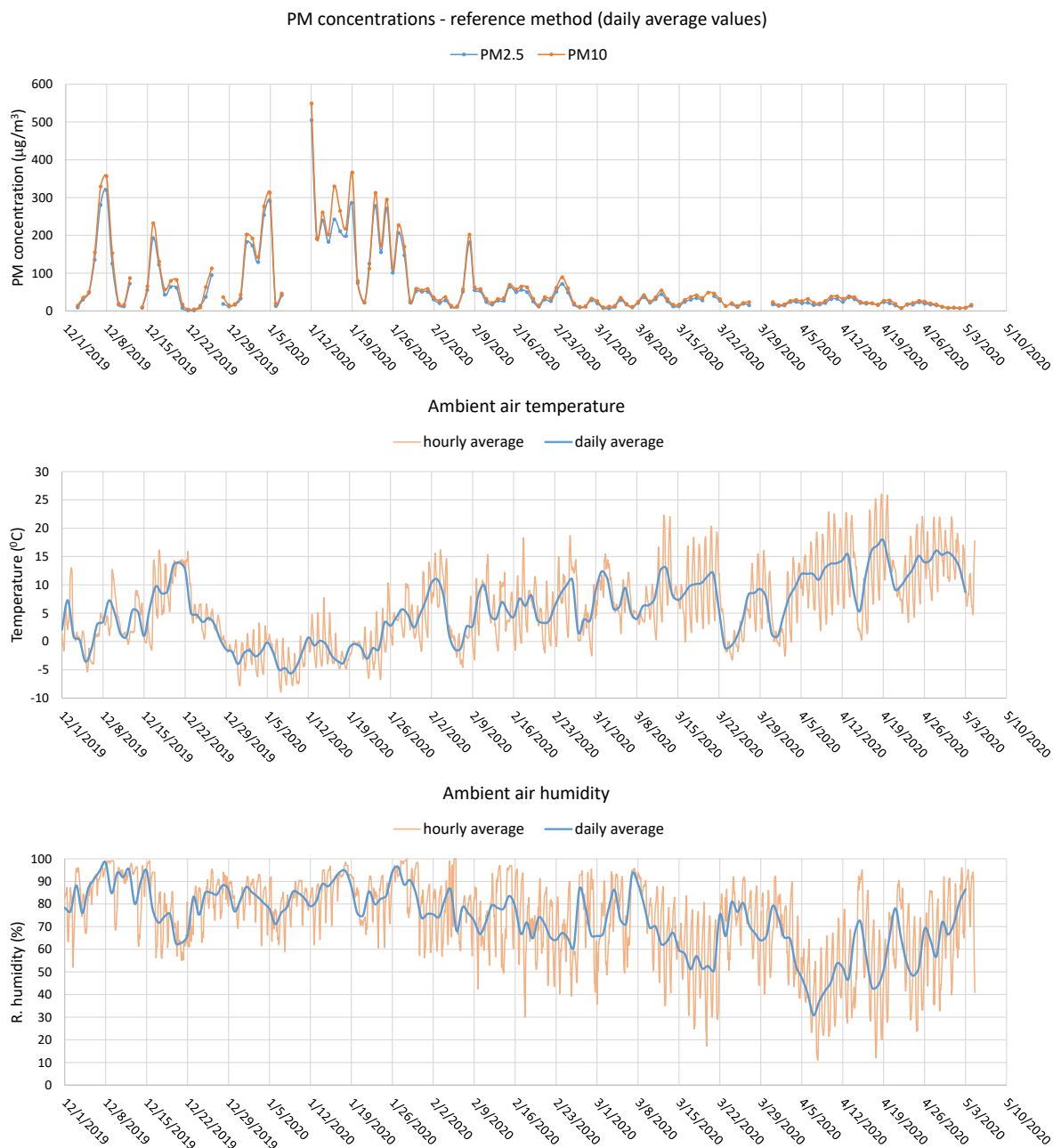
Figure 2. Reference PM concentrations with ambient air temperature and humidity.
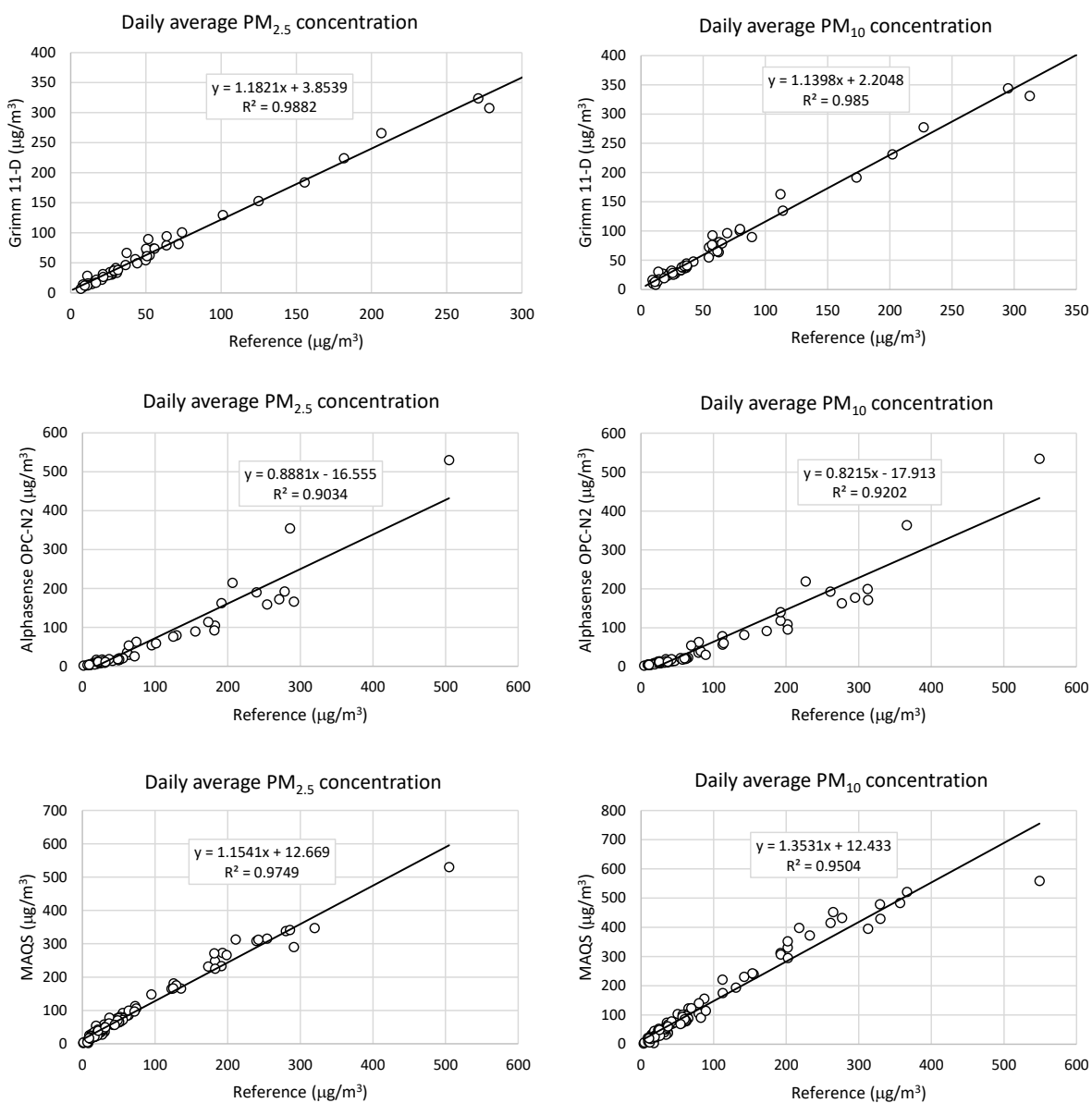
Figure 3. OPS performance during the period of strong pollution (12/2/2019–3/12/2020).
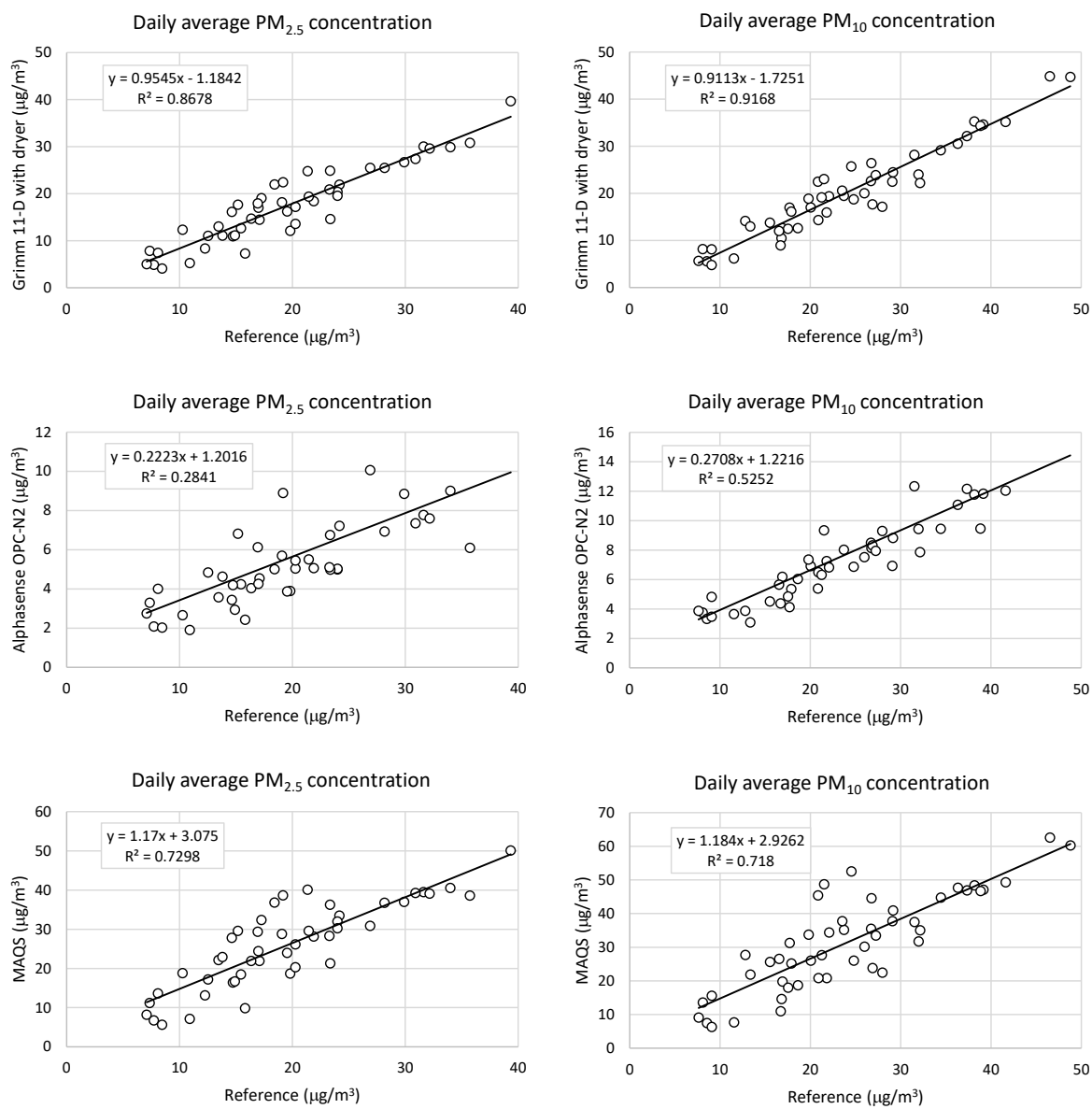
Figure 4. OPS performance during the period of mild pollution (3/13/2020–5/4/2020).
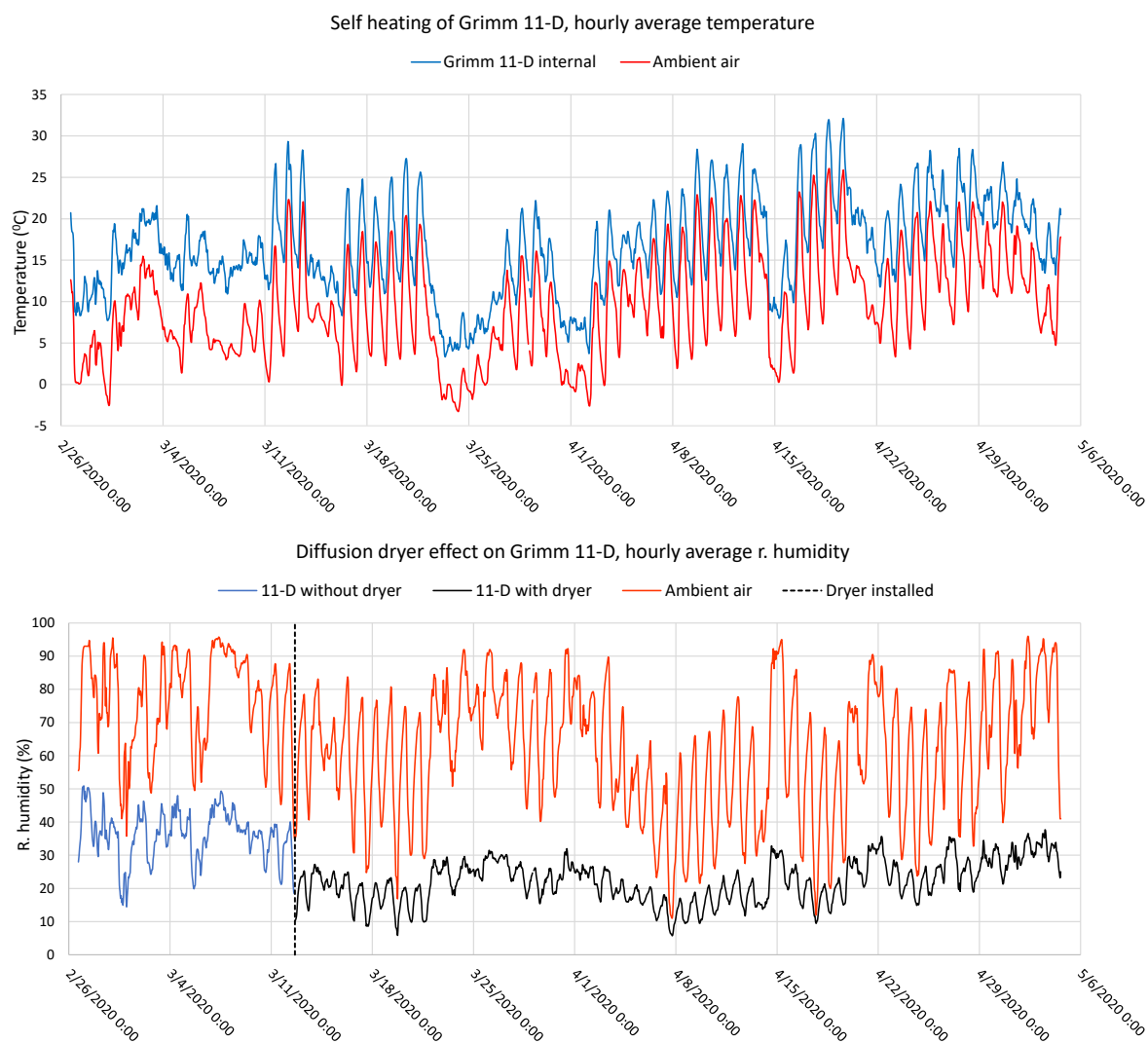
Figure 5. Self-heating and diffusion dryer effect of Grimm 11-D. Dryer was installed on 3/12/2020 15:30.
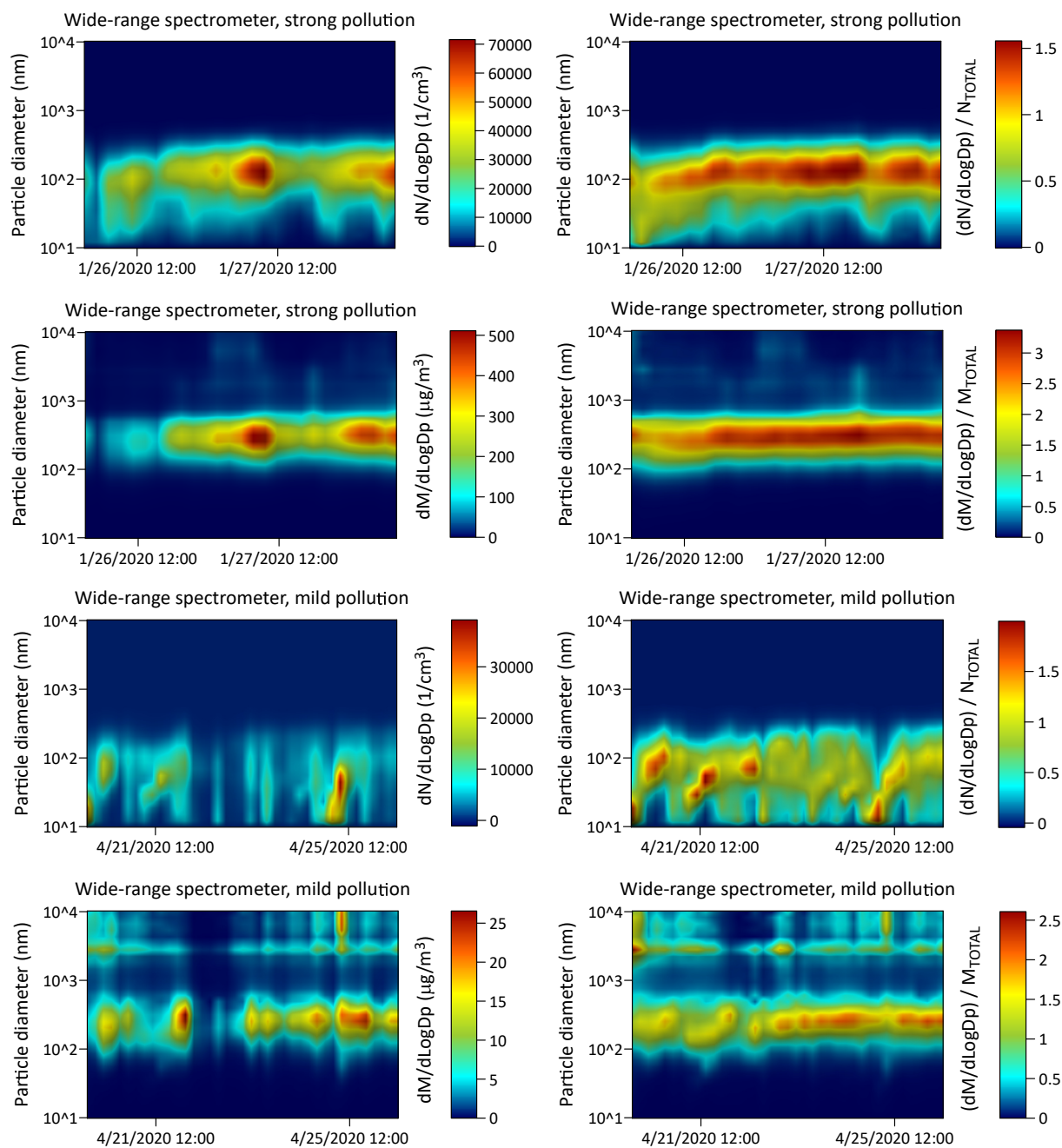
Figure 6. Wide-range spectrometer (SMPS+11D), hourly average values. Relative density 1.65 was applied on SMPS to calculate mass of particles.
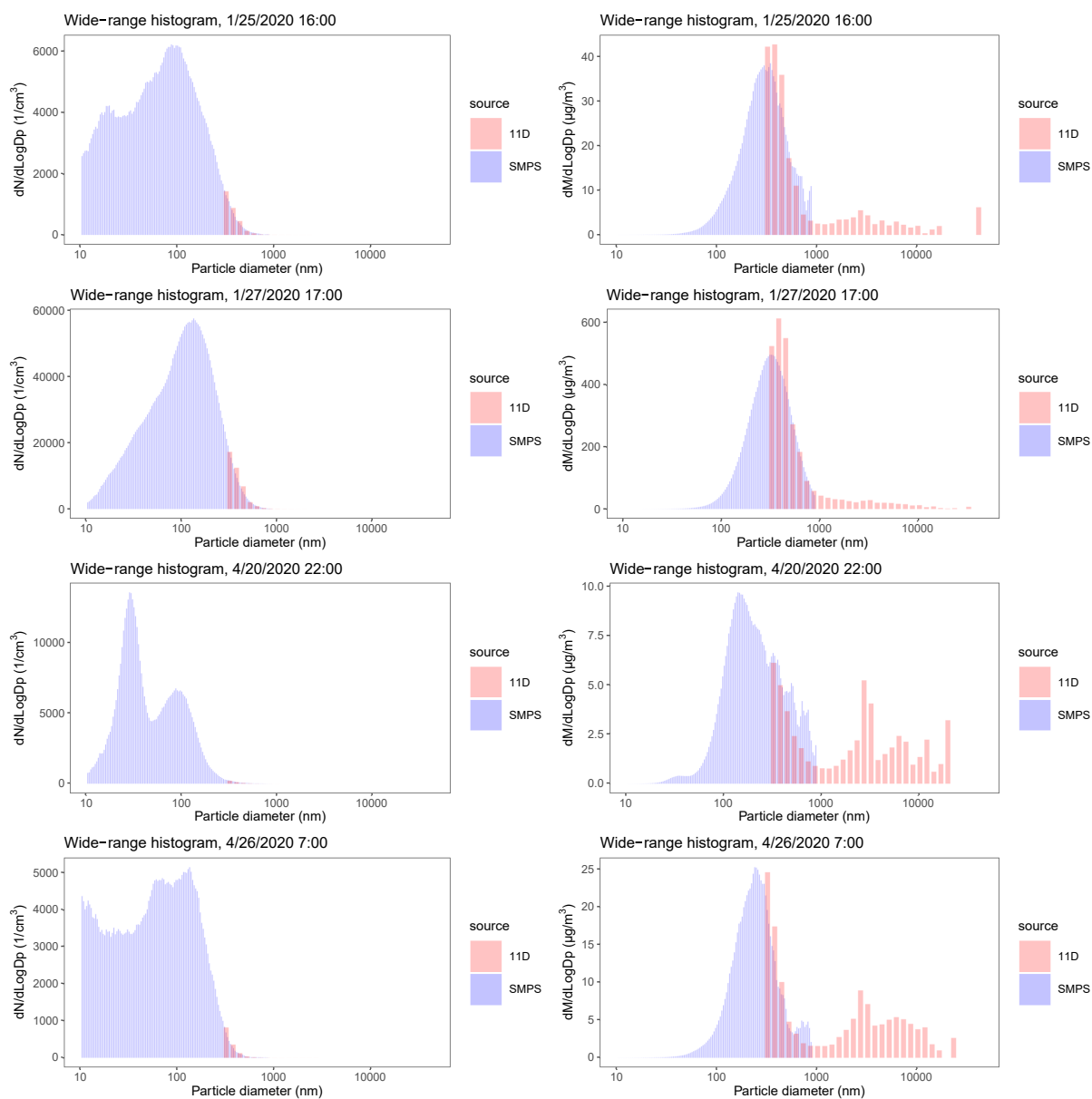
Figure 7. Wide-range histograms, hourly average values. Relative density 1.65 was applied on SMPS to calculate mass of particles.
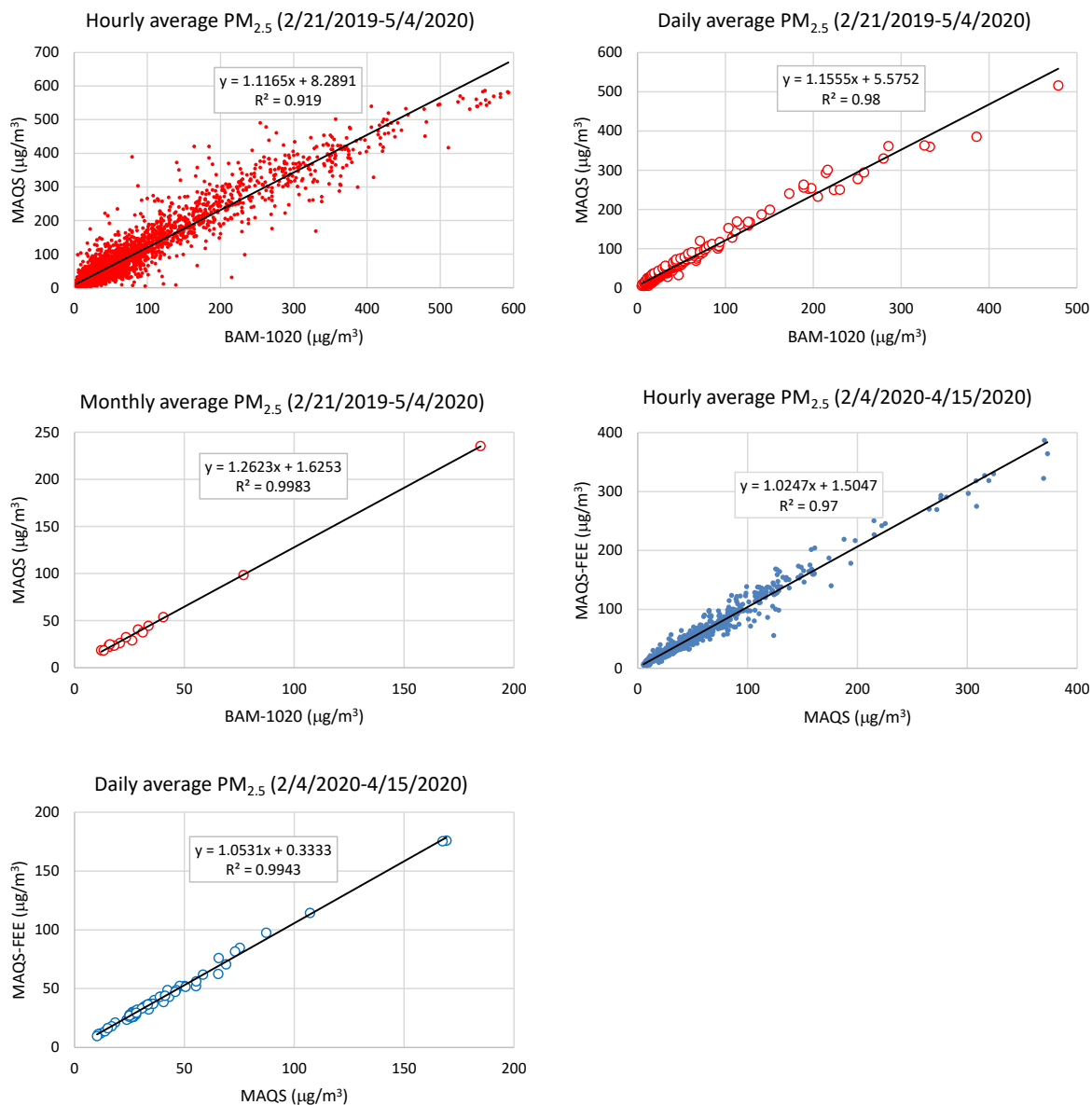
Figure 8. Long-term comparisons of MAQS sensor with BAM operated by US EPA at the nearby location: hourly, daily, monthly average values and comparison of hourly and daily average values of two MAQS sensors: first one (MAQS) at our main facility and second one (MAQS-FEE) at Faculty of Electrical Engineering in the immediate vicinity of BAM-1020.