Response to Anonymous Referee #2

We would like to thank the reviewer for the thoughtful and constructive examination of our paper! Please find below our responses to each comment individually and please note that:

- **Blue bold font** represents comments of the Referee,
- Black regular font represents the response to each referee's comment,
- ~~strikethrough font~~ represents removed text from the manuscript according to referee's comment,
- Red font represents added text in the manuscript according to referee's comments.

General comments: This paper focuses on the performance of 3 OPS devices in a highly polluted area. I think this paper will be helpful to the sensor/air monitoring community as it is at higher ambient concentrations than much previous work with a suite of collocated reference measurements. The authors present a highly valuable dataset. Overall, this is a nice paper with scientific significance, good presentation quality and a few changes to statistical methods/discussion and discussion of previous work will strengthen the scientific quality.

Response: Thank you for the general comments! We will try to implement your suggestions.

Line 114: Can you provide any justification as to why you used the AE channel? The two channels have a nonlinear relationship (Tryner 2020 https://doi.org/10.1016/j.atmosenv.2019.117067) and I wonder if this is some of the reason you have underestimation at high concentration (Figure 3).

Response: The manufacturer of PMS5003 sensor, Plantower, has not explained publicly what is the difference between AE and SM modes, but in private communication they explicitly confirmed that AE is the correct channel for ambient air measurements, while the SM mode is recommended for industrial production workplaces (metal particles or other higher density particles). We have performed laboratory test of these two modes on PMS5003 using the burning chamber and incense scents as the source of PM, and here are the results:

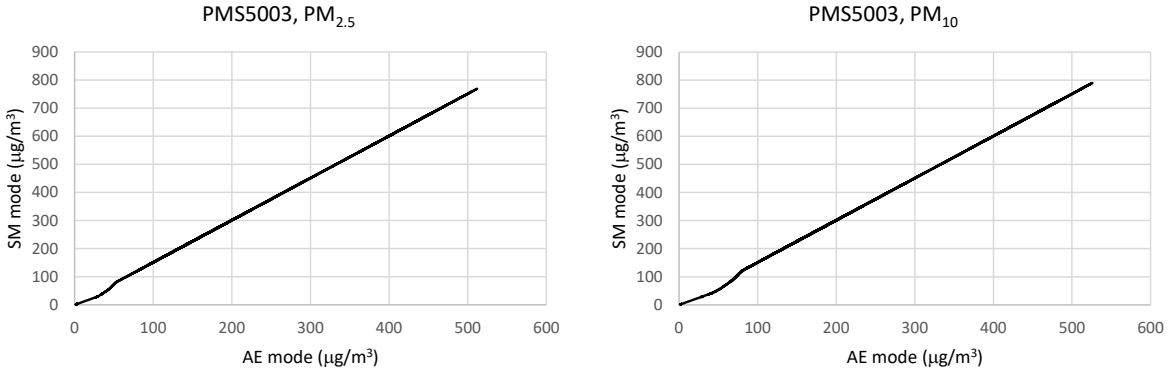


Figure 2. AE and SM modes of PMS5003 sensor.

The relationship between SM and AE modes can be deduced from our test results:

$$\mathrm{SM_{PM2.5}} = \begin{cases} \mathrm{AE_{PM2.5}\ for\ AE_{PM2.5}} \leq 30 \\ \text{nonlinear for } 30 < \mathrm{AE_{PM2.5}} \leq 50 \\ 1.5 \times \mathrm{AE_{PM2.5}\ for\ AE_{PM2.5}} > 50 \end{cases}$$

$$\mathrm{SM_{PM10}} = \begin{cases} \mathrm{AE_{PM10}\ for\ AE_{PM10}} \leq 43 \\ \text{nonlinear for } 43 < \mathrm{AE_{PM10}} \leq 77 \\ 1.5 \times \mathrm{AE_{PM10}\ for\ AE_{PM10}} > 77 \end{cases}$$

Investigation of Tryner et al (2020) https://doi.org/10.1016/j.atmosenv.2019.117067 has limited range of PM concentrations, where we can't see this third segment of direct proportionality between SM and AE mode for higher PM concentrations. In conclusion, for low concentrations these two modes are the same, in the narrow, intermediate range the relationship between SM and AE mode is nonlinear, while for higher concentrations SM mode simply gives 50% higher value than AE mode, for both PM2.5 and PM10. If applied to our measurements, SM mode would overestimate actual PM concentrations more than AE mode. These results are added in section 2.

The authors have a heavy reliance on R2 throughout this paper even though it is well known that this is not the best comparison between measurement methods (Bland & Altman "STATISTICAL METHODS FOR ASSESSING AGREEMENT BETWEEN TWO METHODS OF CLINICAL MEASUREMENT" Lancet, 1986; i: 307-310). They do also discuss bias (% difference) but I think it would also be helpful to not rely so heavily on discussion of R2 and add another metric of scatter MAE (or RMSE or another metric the authors prefer).

Response: we agree completely. For all graphs on figures 3, 4 and 8 in manuscript we have added MAE value and 1:1 line. These MAE values are discussed in sections 3.1, 3.2 and 3.6. Furthermore, Figure 8 is modified for some later discussion on humidity influence. Here are the new graphs:

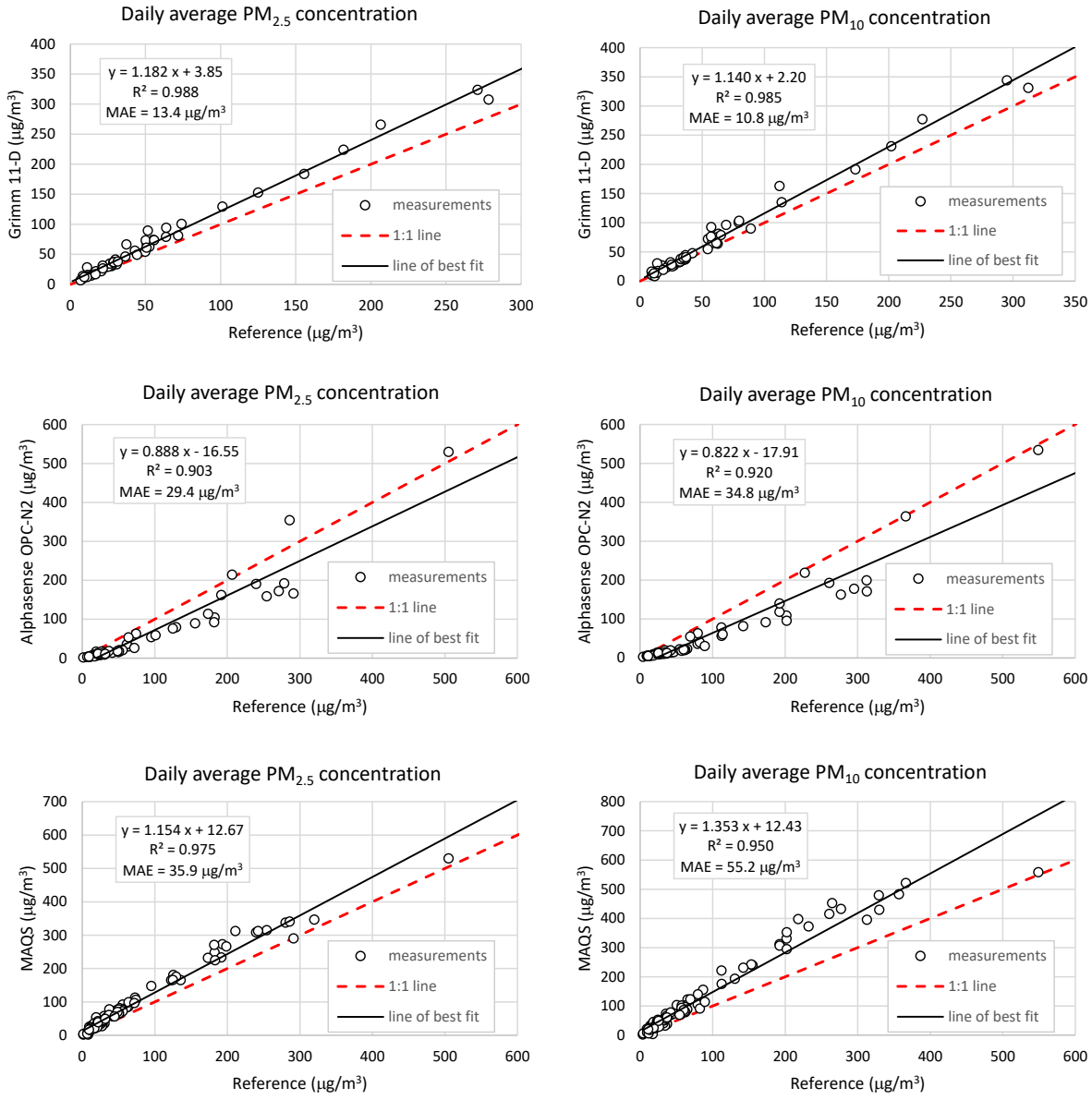


Figure 5. OPS performance during the period of strong pollution (12/2/2019–3/12/2020).

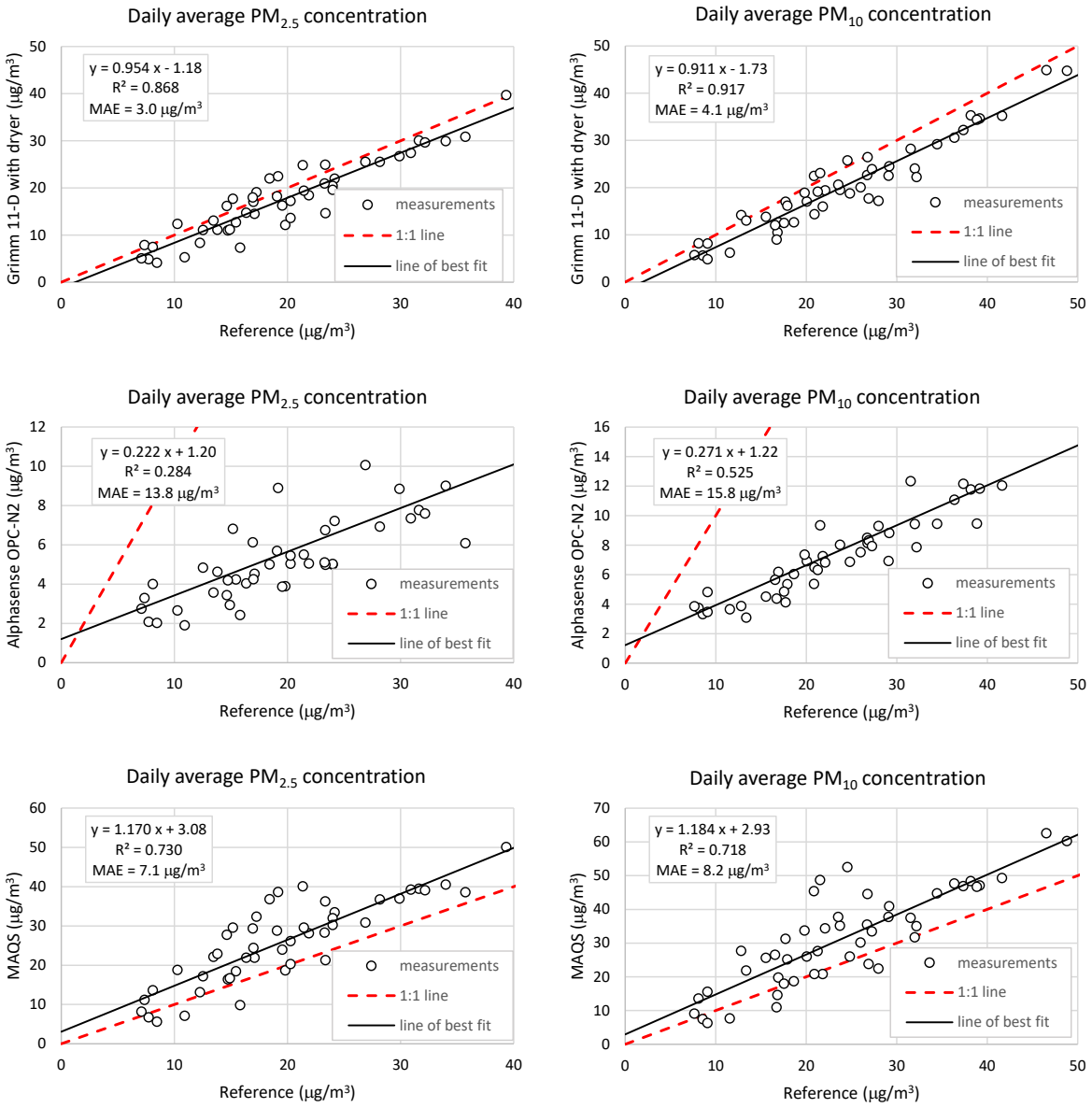


Figure 6. OPS performance during the period of mild pollution (3/13/2020–5/4/2020).

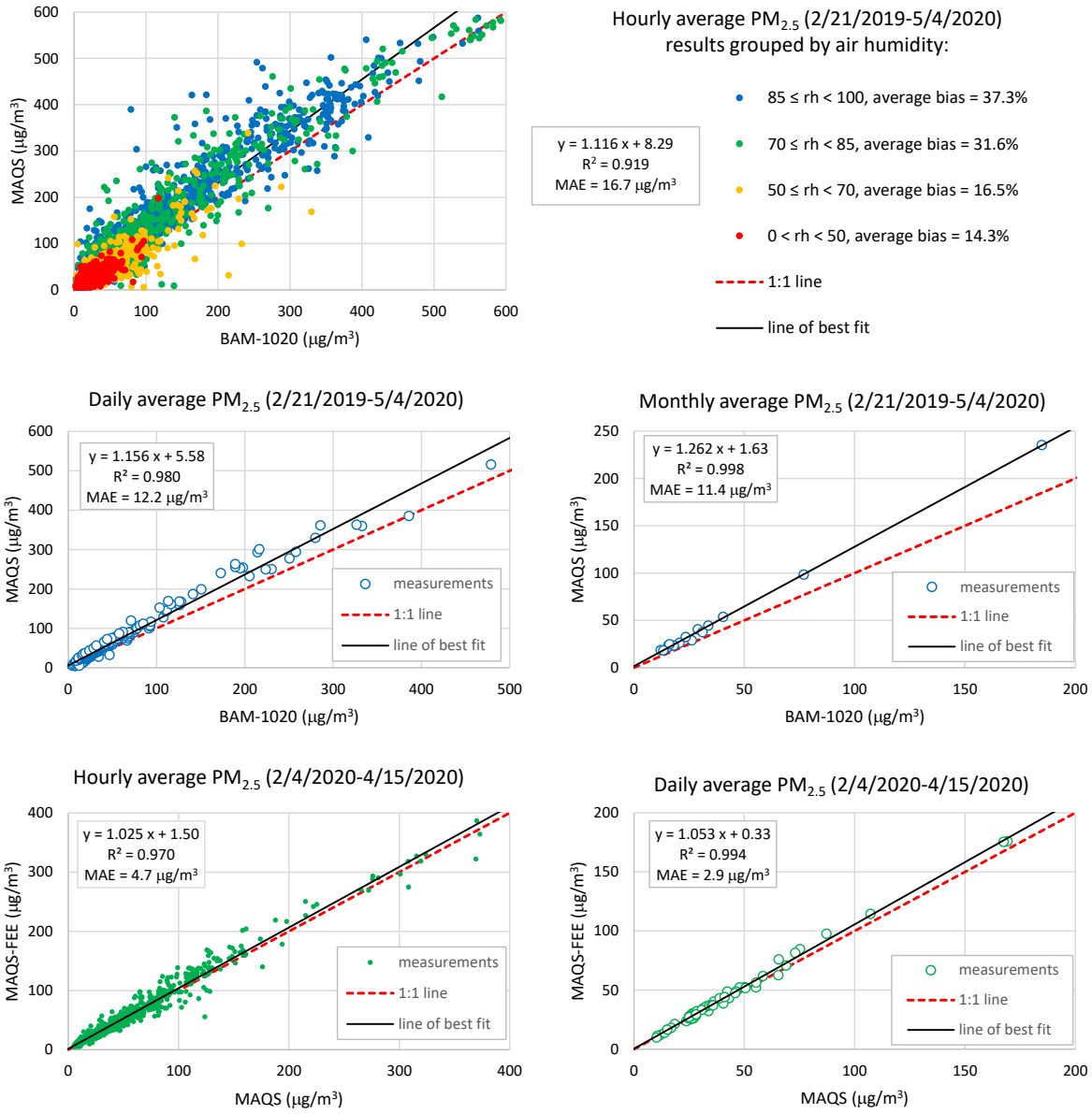


Figure 8. Long-term comparisons of MAQS sensor with BAM operated by US EPA at the nearby location: hourly, daily, monthly average values and comparison of hourly and daily average values of two MAQS sensors: first one (MAQS) at our main facility and second one (MAQS-FEE) at Faculty of Electrical Engineering in the immediate vicinity of BAM-1020.

Line 159-161: I don't think this paragraph provides enough details to understand how you calculated this. I'm guessing this is 3 standard deviations but of what? Just zero concentration experienced in the field? Please elaborate as I think these results will be particularly of interest to the field. It seems like the Bulot paper reports LOD from a bunch of previous work with LCS not just PMS5003/N-2 it might be helpful to strengthen the discussion here. More recent work has also explored the LLOD of PMS5003 sensors (e.g. Magi 2019 https://doi.org/10.1080/02786826.2019.1619915). Also did you want to provide any details on what you do with data below the LLOD (throw out, replace, etc)?

Response: we agree and here are the changes:

Standard deviation (σ) was calculated for periods with near-zero ambient PM concentration and average value of 3σ is estimated LLoD. For PMS5003 our final estimation is 5 μg/m^3. The same value is an estimation of (Magi et al., 2020), calculated by averaging segmented regressions and (Bulot et al., 2019) by combining results from several previous studies. This method applied on OPC-N2 yields LLoD of 2 μg/m^3 and 1 μg/m^3 for 11-D. For reference gravimetric system LLoD was calculated using the blank filters, which were treated exactly the same way as real samples (except the sampling of particulate matter), and the calculated value of LLoD is 0.7 μg/m^3. All measurements below LLoD were discarded during the quality assurance phase.

Line 177: Also see recent paper on PMS5003 and large particles that may be helpful (Kosmopoulos 2020 https://doi.org/10.1016/j.scitotenv.2020.141396)

Response: this is a very relevant reference, and we have added it in the revised manuscript. Investigation in Patras, Greece with narrower range of ambient PM concentrations and different aerosol composition with more frequent episodes of Sahara dust. PurpleAir (PMS5003) showed high bias (similar to our results) and bad performance during the Sahara Desert dust episode. We have also registered one intensive desert dust episode on 3/27/2020 from the Aralkum Desert. Here is the new text:

However, different conditions were observed on 3/27/2000 when the dust from Aralkum Desert covered part of Europe, including our test location. During this episode, OPC-N2 performed much better than PMS5003, which wasn't able to determine large fraction of coarse particles correctly (Figure 11). Similar observation about PMS5003 was reported by (Kosmopoulos et al., 2020), when Sahara dust covered Greece.

The discussion of previous work appears fairly limited. It would be helpful to discuss how the high bias of the PMS5003 and low bias of the OPC-N2 and overall performance compare to studies in other locations as both these devices have been studied fairly extensively.

Response: the following text is added in section 1:

In (Mukherjee et al., 2017) OPC-N2, PMS7003 and 11-R were compared against BAM-1020 during 12 weeks in the Cuyama Valley, California, USA. Grimm 11-R performed well, while both OPC-N2 and PMS7003 (which is a miniaturized version of PMS5003) produced mediocre performance with heavy low bias. PurpleAir (PMS5003) was tested in (Tryner et al., 2020) using laboratory and field tests. High bias of PMS5003 was observed. In (Magi et al., 2020) PurpleAir (PMS5003) was analyzed for 16 months in Charlotte, North Carolina, USA against BAM-1022, high bias of PMS5003 that increases with humidity was reported. High mean bias of PurpleAir (PMS5003) was reported in (Kosmopoulos et al., 2020) as well.

Section 3.3: You only discuss the Humidity influence on the Grimm it would be helpful to discuss the influences on the PMS5003 and OPC-N2 as well.

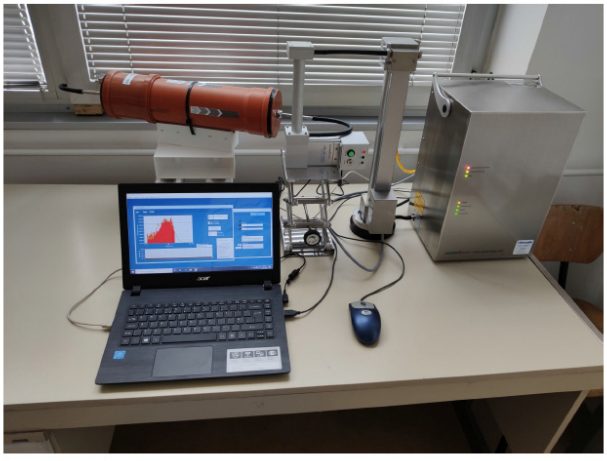
Response: we have constructed dryers only for 11-D and SMPS (new Figure 1). The design and construction of dryers for small, low-cost sensors, such as OPC-N2 and PMS5003 is in our plans for future work. From the dataset of this study, we can include ambient air humidity as the parameter in Figure 8.

Figure 8 shows the long-term (13.5 months) comparison of MAQS and BAM-1020 with time resolution of 1 hour, together with measured values of ambient air humidity. By averaging all this data we can estimate the influence of humidity on the MAQS sensor: if we sort the measurements by humidity, subset of points where humidity is below 50% has average bias of 14.3%, for humidity range 50%-70%, bias is 16.5%; for humidity range 70%-85% bias is 31.6% and for humidity rang 85%-100% bias is 37.3%. If we subtract bias of least humidity subset from bias of highest humidity subset, we can estimate that humidity influence adds up to 23% on PM2.5 readings from MAQS sensor, which is similar result to the analysis of humidity influence on our 11-D with dryer installed. While this influence can not be neglected, it is still relatively modest. Reason for

this is the composition of particles, where we have mostly fine particles below 300 nm, for which hygroscopic growth is less effective (Kosmopoulos 2020).

(a) Air sampler and Stevenson screen.

(b) Devices under test.



(c) SMPS with dryer.

Figure 1. Experimental setup: a) co-located air sampler and Stevenson screen, b) devices under test inside of Stevenson screen: 11-D with dryer, OPC-N2 with SPI adapter and (white-orange) enclosure, MAQS (white enclosure with grey front panel), and c) indoors SMPS with dryer.

It seems like you also have the opportunity to discuss the influence of particle size distribution on the performance of the OPC-N2 and PMS5003 but you have limited your discussion to the Grimm. You mention this briefly in lines 245-249 but it seems like instead of just commenting that small particles could be an issue you can look to see of the OPC-N2 is specifically underestimating more because more of the particles are too small. In addition, both the OPC-N2 and PMS5003 have binned data that could be discussed.

Response: We have data bins for OPC-N2 and PMS5003. However, there are some doubts if the comparison of data bins from PMS5003 and OPC-N2 to 11-D are appropriate, due to the following reasons:

- these devices are constructed differently, 11-D has hydrodynamic focusing (sheath flow), regulated flow rates and high-performance optics, while the low-cost sensors couldn't count individual particles in the same way

– PMS5003 is not a particle counter, it is a nephelometer (Tanzer, 2019) doi.org/10.3390/ijerph16142523

– OPC-N2 has detection limit of 380 nm, PMS5003 from 300 nm, while 11-D can count particles from 250 nm

– when calculating PM mass concentration from individual data bins manufacturers use different weighting factors. Only Alphasense provides values of weighting factors for data bins, assumed particle density for each bin and index of refraction (especially important for correct determination of the particle's diameter). These values for 11-D and PMS5003 are not known.

Taking into account these notes, we present new subsection in the revised manuscript:

3.5 OPS histograms and Aralkum Desert dust

All tested OPS have data bins, with different number of channels, as described in section 2. Figure 11 shows histograms that compare data bins from 11-D, OPC-N2 and MAQS on 1/18/2020 (strong pollution) and 4/16/2020 (mild pollution). It should be noted that we compare here data bins from devices with different specifications and category. As expected, 11-D has ability to count particles below 300 nm, which appear in greatest numbers. Counting efficiency of OPC-N2 is investigaetd in laboratory conditions using PSL particles in (Sousan et al., 2016a), and the results were good for particles larger than 0.8 μm while for particles with diameter of 0.5 μm OPC-N2 the device showed lower detection efficiency (detection limit of OPC-N2 is 0.38 μm). In our realistic scenario, dominant contribution to the mass comes from particles much smaller than 0.8 μm (Figures 9 and 10) which is not favorable to OPC-N2.

Contrary to OPC-N2, PMS5003 has problems with coarse particle, as indicated in laboratory test (Kuula et al., 2020). If the fraction of coarse particles is small and steady, PMS5003 performs much better. Ambient conditions in Bosnia-Herzegovina are such most of the time, since the primary source of PM is combustion of coal and biomass. That could explain why PMS5003 performs better than OPC-N2 most of the time. However, different condition were observed on 3/27/2000 when the dust from Aralkum Desert covered part of Europe, including our test location. During this episode, OPC-N2 performed much better than PMS5003, which wasn't able to determine large fraction of coarse particles correctly (Figure 11). Similar observation about PMS5003 was reported by (Kosmopoulos et al., 2020), when Sahara dust covered Greece.

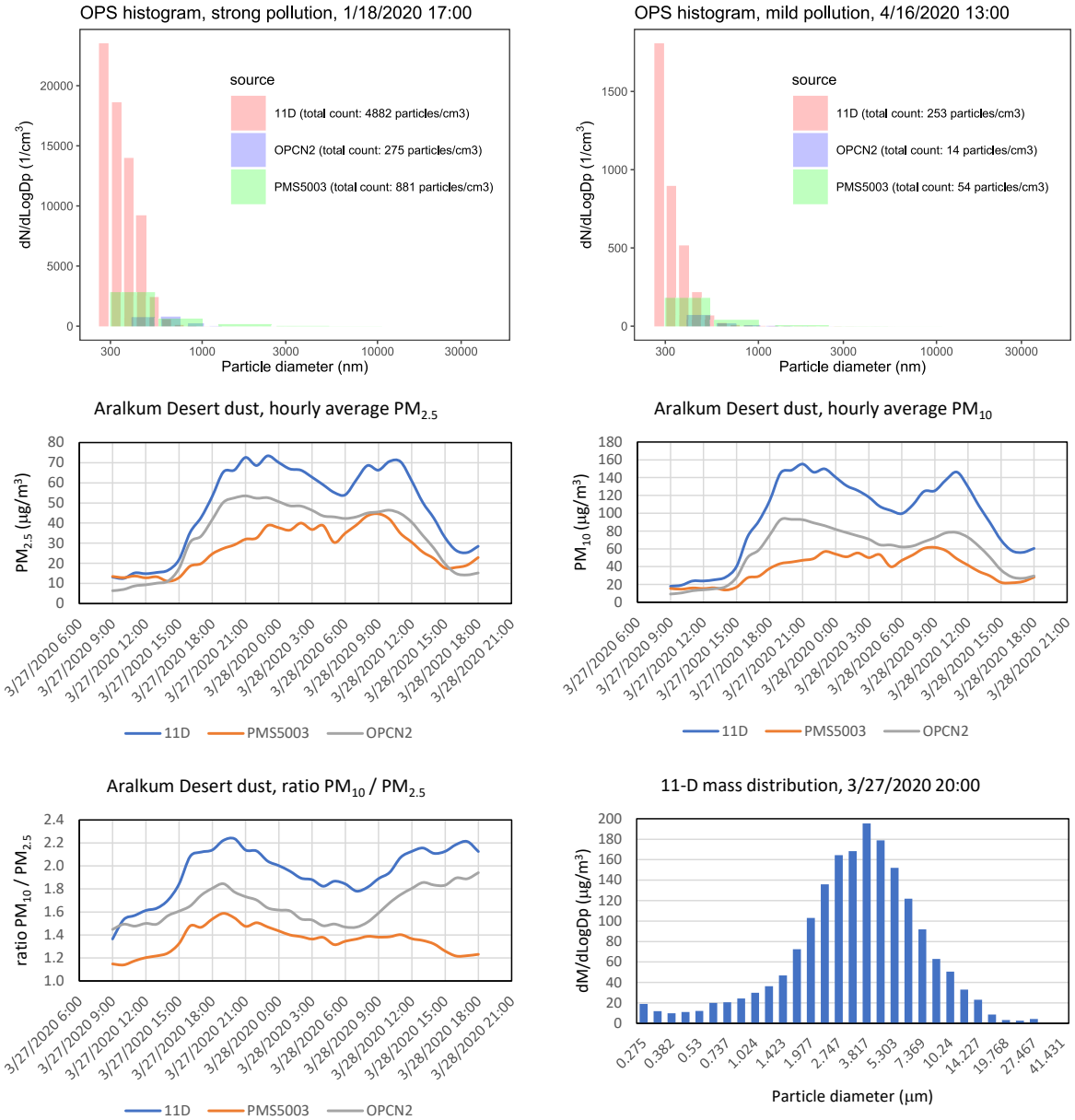


Figure 11. OPS histograms and Aralkum Desert dust episode, hourly average values.

Technical corrections: Line 30: grammatical error "equipped with BAMs" and you should probably spell out what BAM stands for the first time you use it. Line 123,221: missing m on Grimm Line 181: It may be helpful to mention the figure earlier on in the paragraph before discussing the results so that readers can look at the figure and follow along.

Response: we accept all these suggestions and appropriate corrections have been made for the revised text.

Response to Anonymous Referee #3

We would like to thank the reviewer for the thoughtful and constructive examination of our paper! Please find below our responses to each comment individually and please note that:

- **Blue bold font** represents comments of the Referee,
- Black regular font represents the response to each referee's comment,
- ~~strikethrough font~~ represents removed text from the manuscript according to referee's comment,
- Red font represents added text in the manuscript according to referee's comments.

The manuscript describes the evaluation of some optical particulate matter sensors in high and low pollution episodes. The topic of very limited novelty as some of the sensors have already been extensively tested in the peer reviewed literature. 2 of the sensors are already not commercially available anymore and follow-up models are being sold. Novelty and generalizable findings would need to be emphasized because right now, there appears little true scientific discussion on fundamentals that would easily be transposable to justify publication of the manuscript on essentially outdated sensors.

Response: We couldn't agree with this general comment. These sensors are not outdated and they are commercially available: as of today, both Grimm 11-D and Plantower PMS5003 are fully available, while Alphasense OPC-N2 is available in limited quantities for existing customers.

The authors should address the following issues

- What is the true novelty here and insights that were not already documented in the existing papers on the Grimm, the alphasense or the PMS?

Response: the true novelty here is the range of ambient air concentrations, which is much wider than any previous work that we could find in existing articles. This unique dataset is relevant for many countries in Eastern Europe, which share the same problem: air pollution with PM as a dominant component. Performance of optical PM sensors greatly depends on composition of PM. In Eastern Europe many houseolds still use coal as the source of energy for heating. Furthermore, network of govermnetal air quality measuremt stations is relativelae sparse. Considering these facts, it is very importnat to systematicaly analyze performance of OPS in this region. Furthermore, wide-range spectrometer, which consists of SMPS and OPC 11-D is for the first time successfully applied in realistic scenario under such ambient air conditions.

- You use in the comparison figures linear regressions with non-zero intercepts, some of these intercepts are substantial! >10 ug/m3 for PM10 (figure 3) both positive and negative. This needs to be explained.

Response: yes, indeed, the intercept values for the lines of best fit in Fig. 3. in the manuscript vary from -17.9 μg/m^3 to 12.7 μg/m^3. But if we take into account that the range of measurements is from 0 to 600 μg/m^3, these intercepts are relatively small. Please have a look at revised pictures where we have added 1:1 line, according to your suggestion. From these graphs we can see that these intercepts are visually close to the ideal situation (1:1 line).

Overall for all the comparison figures, why not indicate a 1:1 line and please do a deeper analysis. It looks like these figures mostly show non linearity with at low concentrations most data points above the lien and at high below or vice versa. There seems to be clear non linearity without any discussion, instead these weird linear regressions with intercepts that are not explained. Even weirder that the authors acknowledge in the text that there is non linearity likely.

Response: thank you very much for this suggestion! We have new graphs below with included 1:1 line. Regarding the linearity, actually it is surprisingly good across the wide range of measurements for all 3 tested devices. Figures 3 and 8 show some non-linear tendency of tested sensors only above 300 μg/m^3 of $PM_{2.5}$.

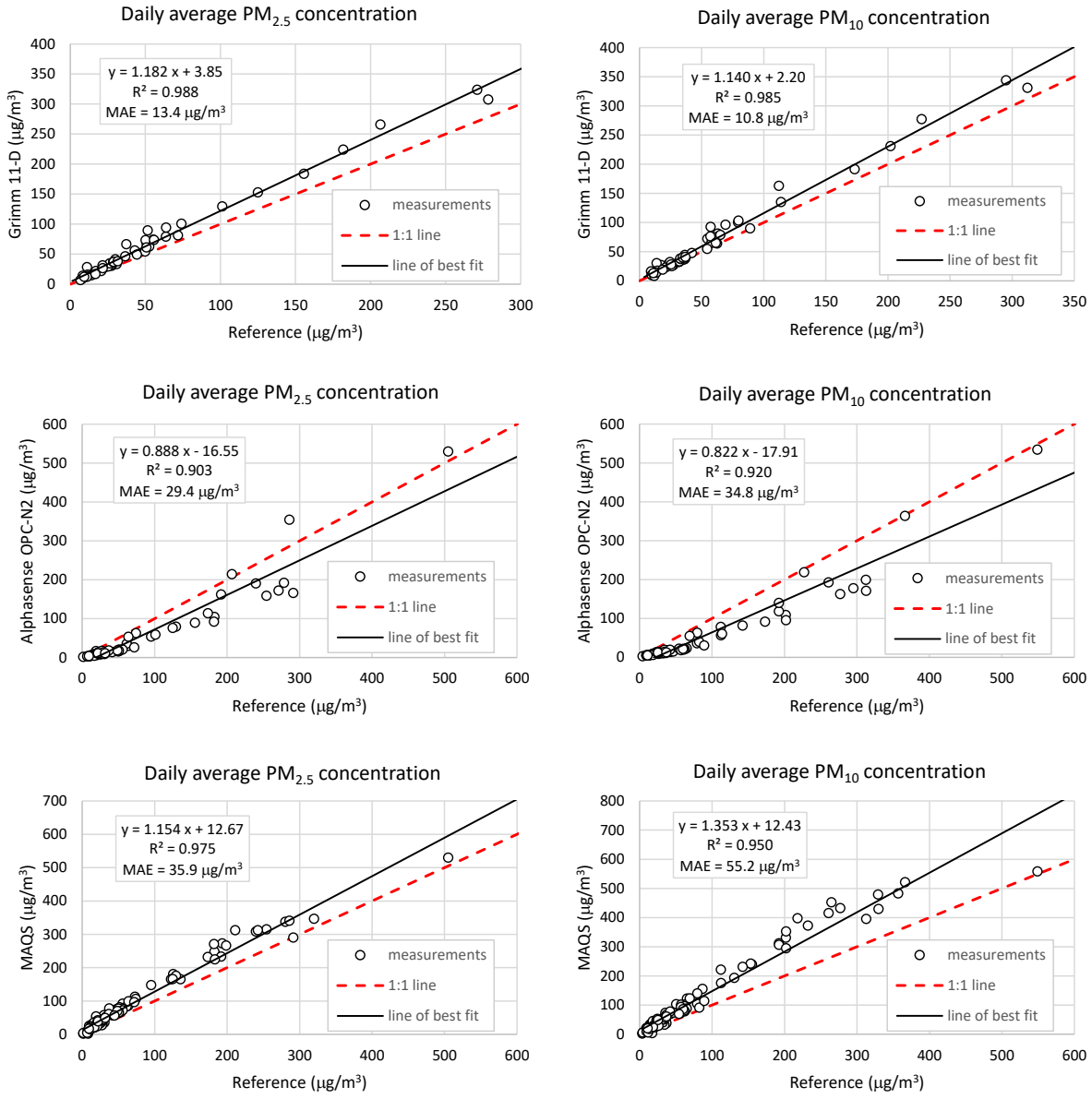
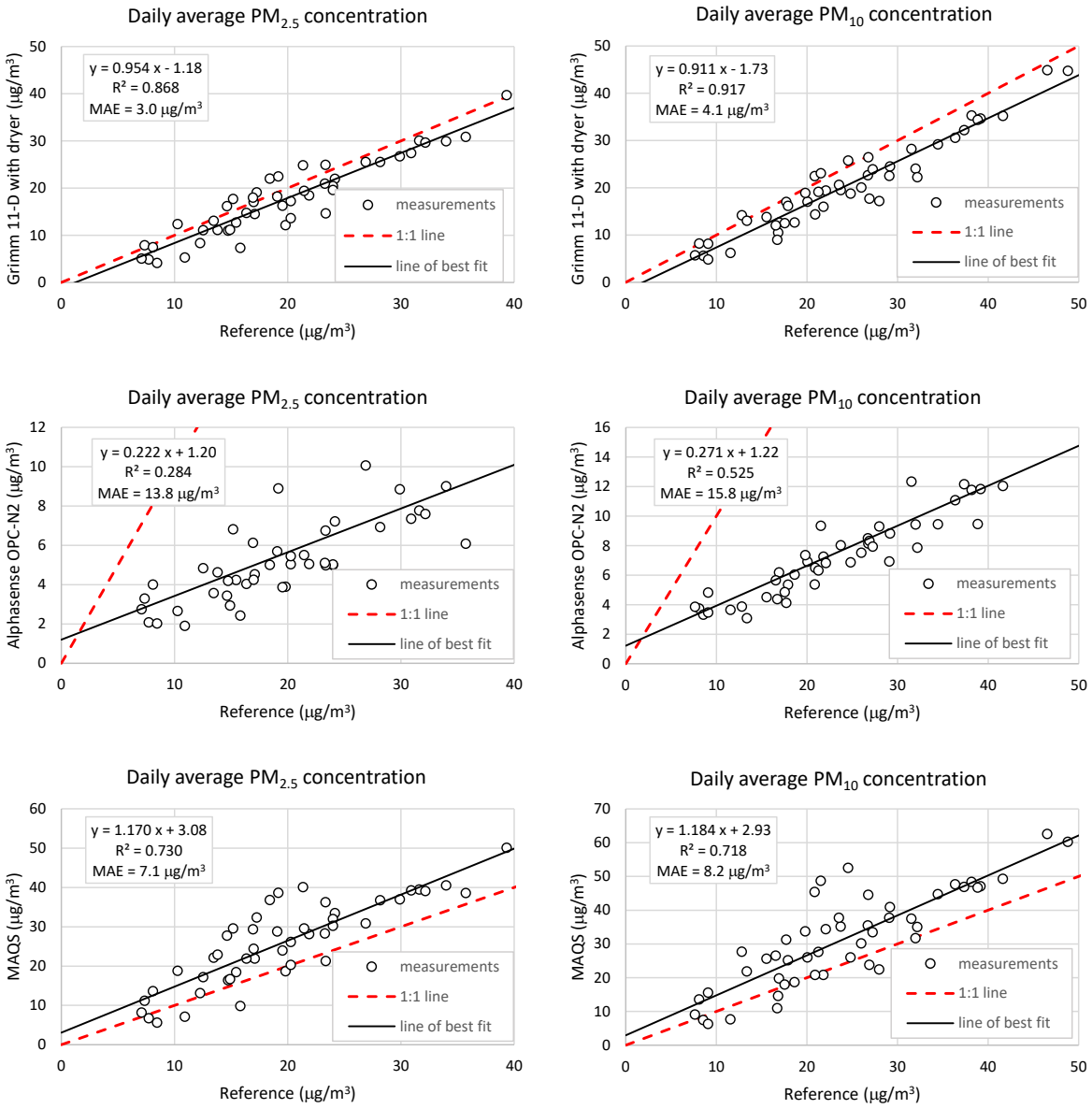


Figure 5. OPS performance during the period of strong pollution (12/2/2019–3/12/2020).



185 **Figure 6.** OPS performance during the period of mild pollution (3/13/2020–5/4/2020).

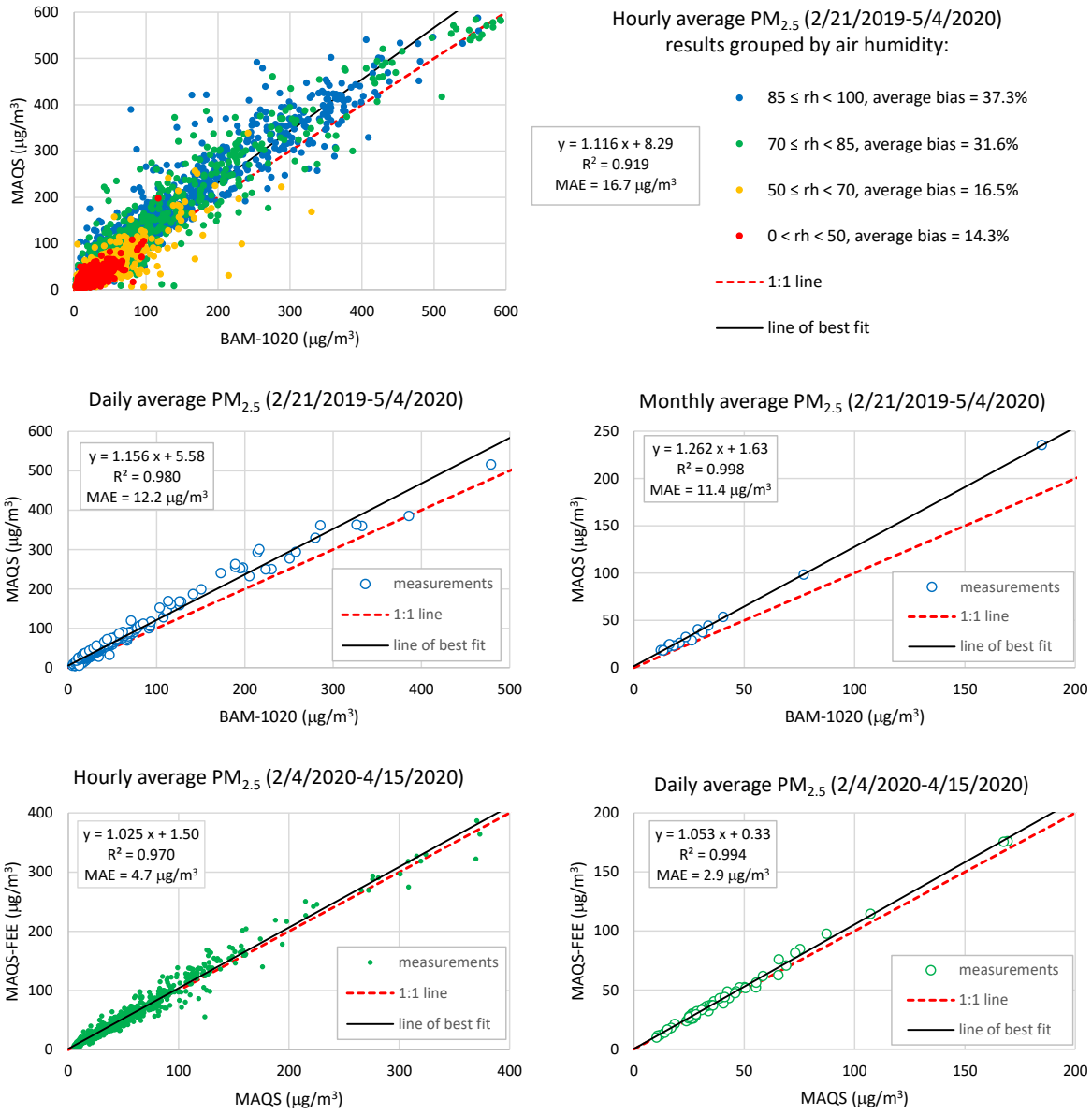
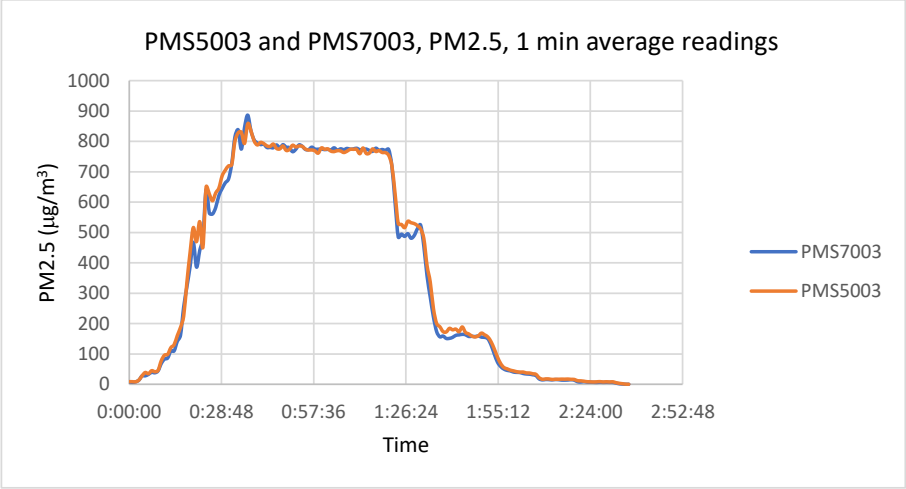


Figure 8. Long-term comparisons of MAQS sensor with BAM operated by US EPA at the nearby location: hourly, daily, monthly average values and comparison of hourly and daily average values of two MAQS sensors: first one (MAQS) at our main facility and second one (MAQS-FEE) at Faculty of Electrical Engineering in the immediate vicinity of BAM-1020.

- The Alphasense is now on version OPC-N3 and it is hard to find information on earlier version idem on the PMS5003, they are now at PMS7003. Could you comment if you expect the observations here to be transposable otherwise they are useless.

Response: OPC-N3 is a successor to OPC-N2. By analyzing the specifications of both, we expect that these results are applicable to OPC-N3 as well. Major differences between OPC-N3 and OPC-N2 are internal temperature and humidity sensor in OPC-N3 and slightly lower detection limit, 350 nm (N3) instedad of 380 nm (N2). While these are useful features, they certainly don't make OPC-N2 obsolete.

Plantower PMS7003 is a miniaturized version of PMS5003. The only advantage of PMS7003 is its smaller size. Because of that PMS7003 is preferred if the size is critical, for example the AirBeam project (Mukherjee, 2017, Sensors). But for our research PMS5003 is more appropriate, because it is more rugged and it has larger intake fan. PurpleAir (Tryner, 2020, AE) is good example of integration of PMS5003 into the network of sensors. We have many laboratory test results of PMS7003 and PMS5003 and conclusion is clear: these sensors give the same results. Please have a look at the results of one of our tests, where we compare these two sensors. Burning chamber with incense scents as the source of PM was used.



Comparison of PMS7003 and PMS5003

- The description of the PMS device seems very speculative? This is very weird when a simple google gives clear descriptions of the device (https://www.aqmd.gov/docs/default-source/aq-spec/resources-page/plantowerpms5003-manual_v2-3.pdf)

Response: we have that file, which contains specifications of PMS5003. However, some important information about the sensor is not published there. For example, there are two modes (SM and AE) and the Manufacturer did not publish what is the difference. But we agree that our description of PMS5003 is not good, and we have changed it.

~~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.~~ It uses red semiconductor laser, photodetector at 90^0 scattering angle (Kuula et al., 2020) and 32-bit processor (Cypress CY8C4245, 48 MHz). According to (Tanzer et al., 2019) PMS5003 is a nephelometer, not the particle counter.

- The discussion of RH impacts is very cursory and given how big the issue is, it would be important to see how results of the sensors agree or disagree as a function of RH. Here it would be critical to discuss that the gravimetric measurements are done at a given RH but how does this RH compare to the sensor measurements.

Response: according to requirements of the standard EN 12341:2014, all filters were conditioned in our gravimetric laboratory at relative humidity between 45% and 50%, and temperature between 19 and 21 ^{0}C. Without dryers on all devices, we can not control these parameters outside. However, this campaign provided us with enough measurements to draw some important conclusions about humidity influence on measurements from OPS. This text is added in section 2:

According to requirements of the standard, all filters were conditioned at relative humidity between 45% and 50%, and temperature between 19 and 21 ^{0}C.

This text is added in section 3.3:

Figure 8 shows the long-term (13.5 months) comparison of MAQS and BAM-1020 with time resolution of 1 hour, together with measured values of ambient air humidity. By averaging all this data we can estimate the influence of humidity on the MAQS sensor: if we sort the measurements by humidity, subset of points where humidity is below 50% has average bias of

14.3%, for humidity range 50%-70%, bias is 16.5%; for humidity range 70%-85% bias is 31.6% and for humidity rang 85%-100% bias is 37.3%. If we subtract bias of least humidity subset from bias of highest humidity subset, we can estimate that humidity influence adds up to 23% on PM2.5 readings from MAQS sensor, which is similar result to the analysis of humidity influence on our 11-D with dryer installed. While this influence can not be neglected, it is still relatively modest. Reason for this is the composition of particles, where we have mostly fine particles below 300 nm, for which hygroscopic growth is less pronounced (Kosmopoulos 2020).

Can you comment that you are running the sensors close to their technical specs (95% RH) also at times you actually do run the Grimm D11 outside of specs as the Grimm specs say temperatures above 4 degC (although you also seem to heat the inlet) this is not very clear.

Response: primary aim of this research is the evaluation of instruments in realistic scenario. That includes wide range of all operating parameters. During the campaign we followed all recommendations of manufacturers of devices, especially Grimm, to ensure that instruments are running normally. Rigorous quality assurance procedure was used. All measurements below LLOD, outside of the specifications and with error or warning codes in the logs were discarded.

The introduction needs serious revision. Particulate matter and aerosol is not the same thing. Please eliminate all discussion of aerosol as aerosol is the particles and the gases

Response: we accept this. Instead of "aerosol", "particulate matter" or other appropriate term will be used consistently through the entire manuscript.

The introduction is very narrowly focused and does not discuss things like the use of TEOM in networks. Also some of the statements should clearly be supported by references

Response: we agree that network of TEOMs is an interesting topic, but it is not related to this work. TEOM is not an optical scattering device.

You are very non quantitative and non rigorous in the text and very imprecise., This needs substantial improvement. E.g. L81: what is meant by extremely high? L85 you know exactly what your lower size limit is, so please state it, L 166 what is mean by a "good" correlation?

Response: L81 "extremely high" refers to measured value of PM2.5 concentration up to 504.9 $\mu\mathrm{g}/\mathrm{m}^3$. That is extremely high concentration of $\mathrm{PM}_{2.5}$. For example, in US AQI categorization, values of $\mathrm{PM}_{2.5}$ over 500 $\mu\mathrm{g}/\mathrm{m}^3$ are beyond air quality index scale. We accept objection about lower size limit in L88, so instead

~~It can detect particles with diameters from few nm up to 1 μm.~~

we have

It can detect particles with diameters from 10 nm up to 1 μm.

R^2 coefficients from 0.90 to 0.99 represent very good correlation in this context (comparison of optical devices to the reference gravimetric method).

The statistical discussion totally lack rigor. L 262 "the correlations for hourly, daily and monthly average values of PM2.5 are 0.919, 0.980 and 0.998, respectively" what does this mean? Followed by "with absolute values overestimated by 20% on average " how was this obtained? Where is the data? This is not obvious from Fig 8 at all?

Response: Fig 8 is redrawn completely, and now you can see these average bias values on the figure as a function of air humidity. The underlying datasets for this publication are available at

https://doi.org/10.5281/zenodo.3897379

Here are the text changes:

~~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.~~

Based on 13.5 months of continuous comparison of MAQS and BAM-1020, hourly average values give R^2 coefficient 0.919 and MAE 16.7 μg/m^3. Daily average values produce R^2 coefficient 0.980 and MAE 12.2 μg/m^3, while the monthly average values give $R^2 = 0.998$ and MAE $= 11.4\,\mu$g/m^3 (Figure 8).

- the abstract should not read like an experiential section with study dates etc. These details should not go there instead it should contain quantitative results form the paper.

Response: we accept that, here is the new 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 (Mobile Air Quality System) 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).~~ This interval is divided into period 1 - strong pollution and period 2 - mild pollution. 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~~ particles, 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.

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 (Mobile Air Quality System) 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).~~ This interval is divided into period 1 - strong pollution and period 2 - mild pollution. 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 $\mu g/m^3$, with daily average values exceeding 500 $\mu g/m^3$. During period 2, the average concentration of $PM_{2.5}$ was 20 $\mu g/m^3$. 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~~ particles, 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~~ particulate matter 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~~ particulate matter is a dominant pollutant among other components of pollution. Therefore, developing a strategy for reliable quantification of

~~aerosols~~ particulate matter in ambient air is necessary. The traditional and most accurate approach to measuring the particulate matter concentration in the air is the reference method, 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 fixed, semi-automatic stations equipped with ~~BAM~~ beta attenuation monitors (BAMs). 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).

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 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 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 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:

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

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. 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~~ particles, 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~~ particles (Cai et al., 2019). Burning chamber is used in some investigations as well (Wang et al., 2015). However, equation (1) is strongly non-linear in terms of refractive index, and in most practical cases corrections for different ~~aerosol~~ particles' 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~~ particles (Jayaratne et al., 2018; Granados-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.

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 ~~aerosol~~ particulate matter pollution, range of concentrations and meteorological factors. In (Mukherjee et al., 2017) OPC-N2, PMS7003 and 11-R were compared against BAM-1020 during 12 weeks in the Cuyama Valley, California, USA. Grimm 11-R performed well, while both OPC-N2 and PMS7003 (which is a miniaturized version of PMS5003) produced mediocre performance with heavy low bias. PurpleAir (PMS5003) was tested in (Tryner et al., 2020) using laboratory and field tests. High bias of PMS5003 was observed. In (Magi et al., 2020) PurpleAir (PMS5003) was analyzed for 16 months in Charlotte, North Carolina, USA against BAM-1022, high bias of PMS5003 that increases with humidity was reported. High mean bias of PurpleAir (PMS5003) was reported in (Kosmopoulos et al., 2020) as well.

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 $PM_{2.5}$ was very high: 167.3 μg/m^3. In contrast to that, monthly average values for March and April 2020 were 21.6 μg/m^3 and 19.6 μg/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 ~~aerosol~~ particles' diameters, with a span of more than three orders of magnitude. This will give detailed insights on the mass distribution of ~~aerosols~~ particles.

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 for measurements of PM concentrations was a Dadolab Gemini air sampler (Figure 1). It is a single device with two completely independent 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. According to requirements of the standard, all filters were conditioned at relative humidity between 45% and 50%, and temperature between 19 and 21 ^{0}C.

Grimm 11-D is a high-end optical particle sizer, with sophisticated construction and ability to count individual particles 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. 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 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.~~ It uses red semiconductor laser, photodetector at 90^0 scattering angle (Kuula et al., 2020) and 32-bit processor (Cypress CY8C4245, 48 MHz). According to (Tanzer et al., 2019) PMS5003 is a nephelometer, not the particle counter. 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, since the manufacturer recommends AE mode for ambient air measurements, without further explanation. Figure 2 shows results from our laboratory test using the incense scents as the source of PM. Based on these results, the relationships between SM and AE modes are

$$\mathrm{SM}_{\mathrm{PM2.5}} = \begin{cases} \mathrm{AE}_{\mathrm{PM2.5}} \text{ for } \mathrm{AE}_{\mathrm{PM2.5}} \leq 30 \\ \text{nonlinear for } 30 < \mathrm{AE}_{\mathrm{PM2.5}} \leq 50 \\ 1.5 \times \mathrm{AE}_{\mathrm{PM2.5}} \text{ for } \mathrm{AE}_{\mathrm{PM2.5}} > 50 \end{cases}$$
$$\mathrm{SM}_{\mathrm{PM10}} = \begin{cases} \mathrm{AE}_{\mathrm{PM10}} \text{ for } \mathrm{AE}_{\mathrm{PM10}} \leq 43 \\ \text{nonlinear for } 43 < \mathrm{AE}_{\mathrm{PM10}} \leq 77 \\ 1.5 \times \mathrm{AE}_{\mathrm{PM10}} \text{ for } \mathrm{AE}_{\mathrm{PM10}} > 77 \end{cases} \quad (5)$$

Based 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 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.

Grimm 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 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 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) 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~~ particles 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 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~~ particle 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 DMA use very high operating voltages). The air was sampled from outside using a conductive tube of shortest possible length, to avoid ~~aerosol~~ particle 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

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 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 actual field performance. ~~using standard methodology (3σ), and the results are: 5 μg/m^3 for PMS5003, 2 μg/m^3 for OPC-N2 and 1 μg/m^3 for 11-D. These values are comparable to those reported by (Bulot et al., 2019)~~ Standard deviation (σ) was calculated for periods with near-zero ambient PM concentration and average value of 3σ is estimated LLoD. For PMS5003 our final estimation is 5 μg/m^3. The same value is an estimation of (Magi et al., 2020), calculated by averaging segmented regressions and (Bulot et al., 2019) by combining results from several previous studies. This method applied on OPC-N2 yields LLoD of 2 μg/m^3 and 1 μg/m^3 for 11-D. For reference gravimetric system LLoD was calculated using the blank filters, which were treated exactly the same way as real samples (except the sampling of particulate matter), and the calculated value of LLoD is 0.7 μg/m^3. All measurements below LLoD were discarded during the quality assurance phase.

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 μg/m^3, with minimum daily average value 1.3 μg/m^3 and maximum value 504.9 μg/m^3. In the same period, the average PM_{10} con-

centration was 95.5 μg/m^3, with minimum value 3.6 μg/m^3 and maximum value 549.0 μg/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 (Figure 5). 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 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. Mean absolute error (MAE) was 13.4 μg/m^3 for PM$_{2.5}$ and 10.8 μg/m^3 for PM$_{10}$. 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. MAE for this sensor was 29.4 μg/m^3 for PM$_{2.5}$ and 34.8 μg/m^3 for PM$_{10}$. 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. MAE was 35.9 μg/m^3 for PM$_{2.5}$ and 55.2 μg/m^3 for PM$_{10}$. 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 μm-10 μm] is noisy and inaccurate (Kuula et al., 2020). Further investigation of this behavior would be useful.

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~~ particle 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 6 shows. 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 μg/m^3). The average concentration of PM$_{2.5}$ was 19.7 μg/m^3 with a minimum daily average value of 7.1 μg/m^3 and a maximum value of 39.3 μg/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 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 with dryer, 11-D shows level of performance comparable to BAM, at least those reported by Hafkenscheid and Vonk (2014). MAE was 3.0 μg/m^3 for PM$_{2.5}$ and 4.1 μg/m^3 for PM$_{10}$.

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. MAE was 13.8 $\mu g/m^3$ for $PM_{2.5}$ and 15.8 $\mu g/m^3$ for PM_{10}.

MAQS sensor demonstrated mediocre performance, with R^2 values of 0.730 for $PM_{2.5}$ and 0.718 for PM_{10}. On average, 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). MAE was 7.1 $\mu g/m^3$ for $PM_{2.5}$ and 8.2 $\mu g/m^3$ for PM_{10}.

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 (i.e. appear larger) during measurements due to hygroscopic growth. This can be described using Hänel equation:

$$f_\zeta(RH) = \left(\frac{1 - RH}{1 - RH_{\text{ref}}}\right)^{-\gamma}, \tag{6}$$

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

$$f_\zeta(RH) = \frac{\zeta(RH)}{\zeta(RH_{\text{ref}})}. \tag{7}$$

It is important to note that the coefficient γ, which is an indicator of the hygroscopicity of ~~aerosol~~ particles, depends on the type of ~~aerosol~~ particles (and changes whenever composition of ambient ~~aerosols~~ particles 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, 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 $\mu g/m^3$ 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 $\mu g/m^3$ while 11-D (with dryer) measured 1.4% smaller value. This comparison indicates that the effect of the dryer could be around 23%. A similar analysis for PM_{10} concentrations gives an estimate of about 20% for the dryer effect.

Grimm 11-D has a very useful feature: internal temperature and humidity sensor. Figure 7 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 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).

Figure 8 shows the long-term (13.5 months) comparison of MAQS and BAM-1020 with time resolution of 1 hour, together with measured values of ambient air humidity. By averaging all this data we can estimate the influence of humidity on the MAQS sensor: if we sort the measurements by humidity, subset of points where humidity is below 50% has average bias of 14.3%, for humidity range 50%-70%, bias is 16.5%; for humidity range 70%-85% bias is 31.6% and for humidity rang 85%-100% bias is 37.3%. If we subtract bias of least humidity subset from bias of highest humidity subset, we can estimate that humidity influence adds up to 23% on PM2.5 readings from MAQS sensor, which is similar result to the analysis of humidity influence on our 11-D with dryer installed. While this influence can not be neglected, it is still relatively modest. Reason for this is the composition of particles, where we have mostly fine particles below 300 nm, for which hygroscopic growth is less pronounced (Kosmopoulos 2020).

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 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 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~~ particles, both for number concentrations and mass distribution.

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.

3.5 OPS histograms and Aralkum Desert dust

All tested OPS have data bins, with different number of channels, as described in section 2. Figure 11 shows histograms that compare data bins from 11-D, OPC-N2 and MAQS on 1/18/2020 (strong pollution) and 4/16/2020 (mild pollution). It should be noted that we compare here data bins from devices with different specifications and category. As expected, 11-D has ability to count particles below 300 nm, which appear in greatest numbers. Counting efficiency of OPC-N2 is investigated in laboratory conditions using PSL particles in (Sousan et al., 2016a), and the results were good for particles larger than 0.8 μm while for particles with diameter of 0.5 μm OPC-N2 the device showed lower detection efficiency (detection limit of OPC-N2 is 0.38 μm). In our realistic scenario, dominant contribution to the mass comes from particles much smaller than 0.8 μm (Figures 9 and 10) which is not favorable to OPC-N2.

Contrary to OPC-N2, PMS5003 has problems with coarse particle, as indicated in laboratory test (Kuula et al., 2020). If the fraction of coarse particles is small and steady, PMS5003 performs much better. Ambient conditions in Bosnia-Herzegovina are such most of the time, since the primary source of PM is combustion of coal and biomass. That could explain why PMS5003 performs better than OPC-N2 most of the time. However, different conditions were observed on 3/27/2000 when the dust from Aralkum Desert covered part of Europe, including our test location. During this episode, OPC-N2 performed much better than PMS5003, which wasn't able to determine large fraction of coarse particles correctly (Figure 11). Similar observation about PMS5003 was reported by (Kosmopoulos et al., 2020), when Sahara dust covered Greece.

3.6 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 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 ($R^2 = 0.970$, MAE $= 4.7\,\mu$g/m^3 for hourly average values and $R^2 = 0.994$, MAE $= 2.9\,\mu$g/m^3 for daily average values) confirming our assumption that these two locations share the 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.~~

Based on 13.5 months of continuous comparison of MAQS and BAM-1020, hourly average values give R^2 coefficient 0.919 and MAE 16.7 μg/m^3. Daily average values produce R^2 coefficient 0.980 and MAE 12.2 μg/m^3, while the monthly average values give $R^2 = 0.998$ and MAE $= 11.4\,\mu$g/m^3 (Figure 8).

This leads us to the conclusion that time averaging reduces a lot the influence of variation of ~~aerosol~~ PM 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 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~~ particles, insert a lot of noise, and the benefit is questionable. Depending on ambient conditions, self-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~~ PM changes, inevitably injecting noise into the results.

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

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-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 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~~ particles, both in number and mass concentrations. Particles with diameters around 100 nm (and sometimes below) represent 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

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.

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 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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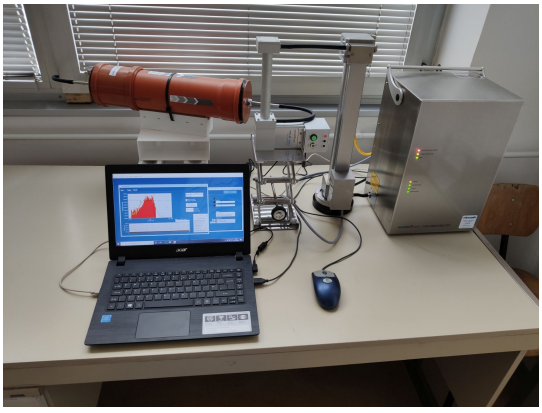
(a) Air sampler and Stevenson screen.

(b) Devices under test.



(c) SMPS with dryer.

Figure 1. Experimental setup: a) colocated air sampler and Stevenson screen, b) devices under test inside of Stevenson screen: 11-D with dryer, OPC-N2 with SPI adapter and (white-orange) enclosure, MAQS (white enclosure with grey front panel), and c) indoors SMPS with dryer.

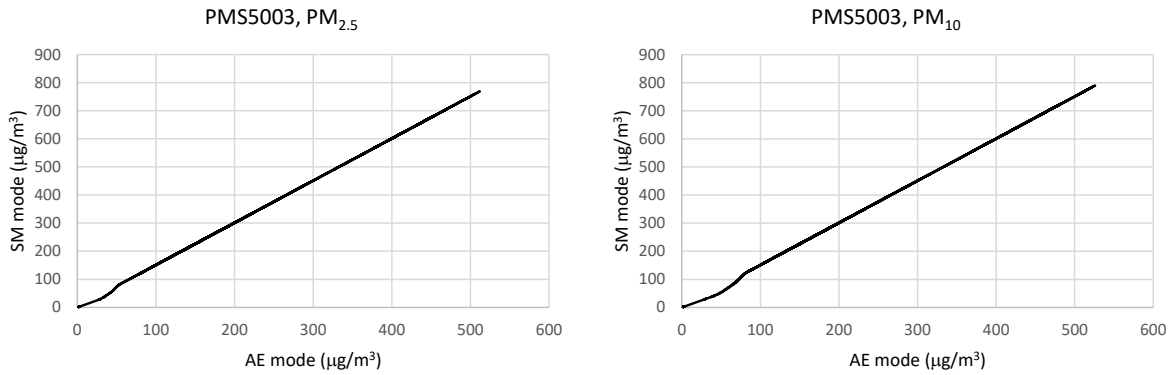


Figure 2. AE and SM modes of PMS5003 sensor.

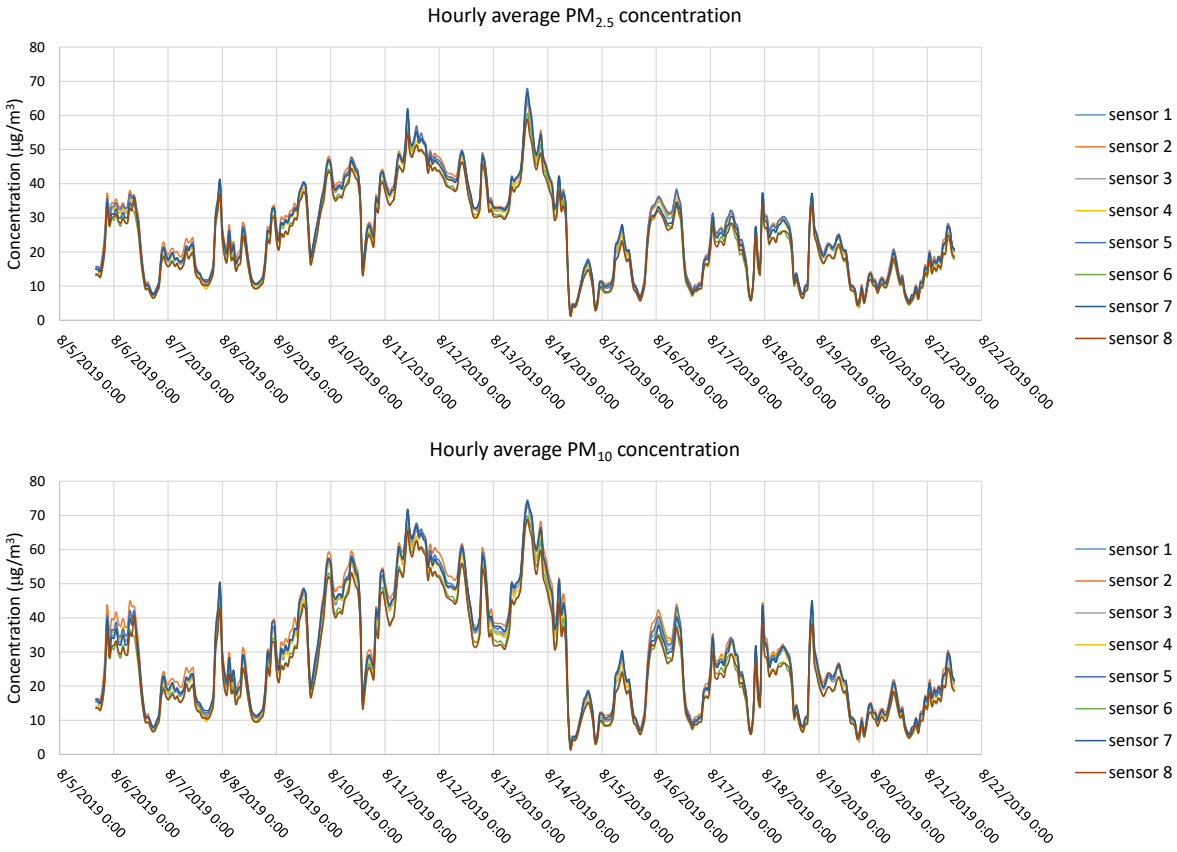


Figure 3. Preliminary test of 8 MAQS sensors, outdoor measurements.

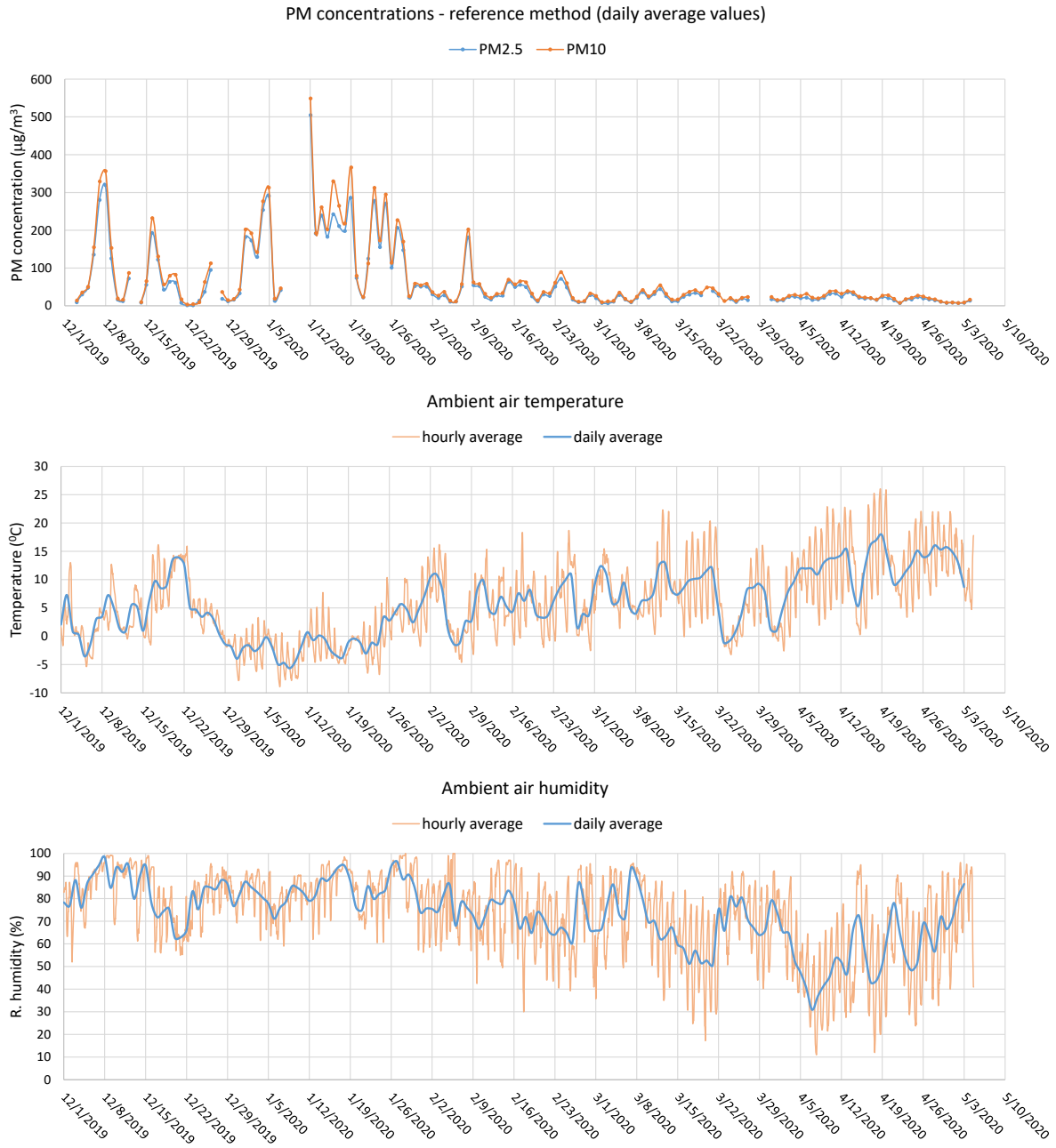


Figure 4. Reference PM concentrations with ambient air temperature and humidity.

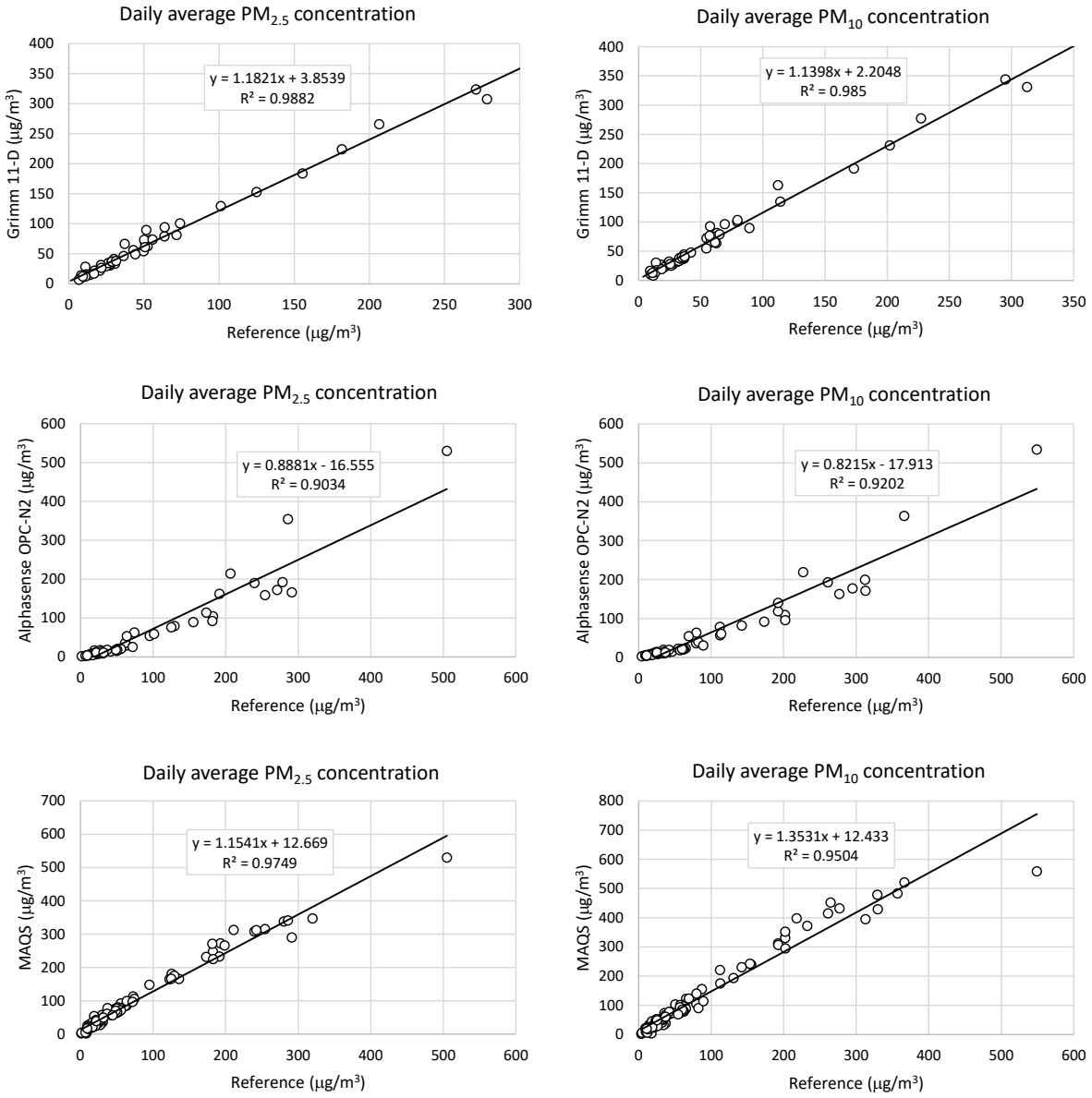


Figure 3. OPS performance during the period of strong pollution (12/2/2019–3/12/2020).

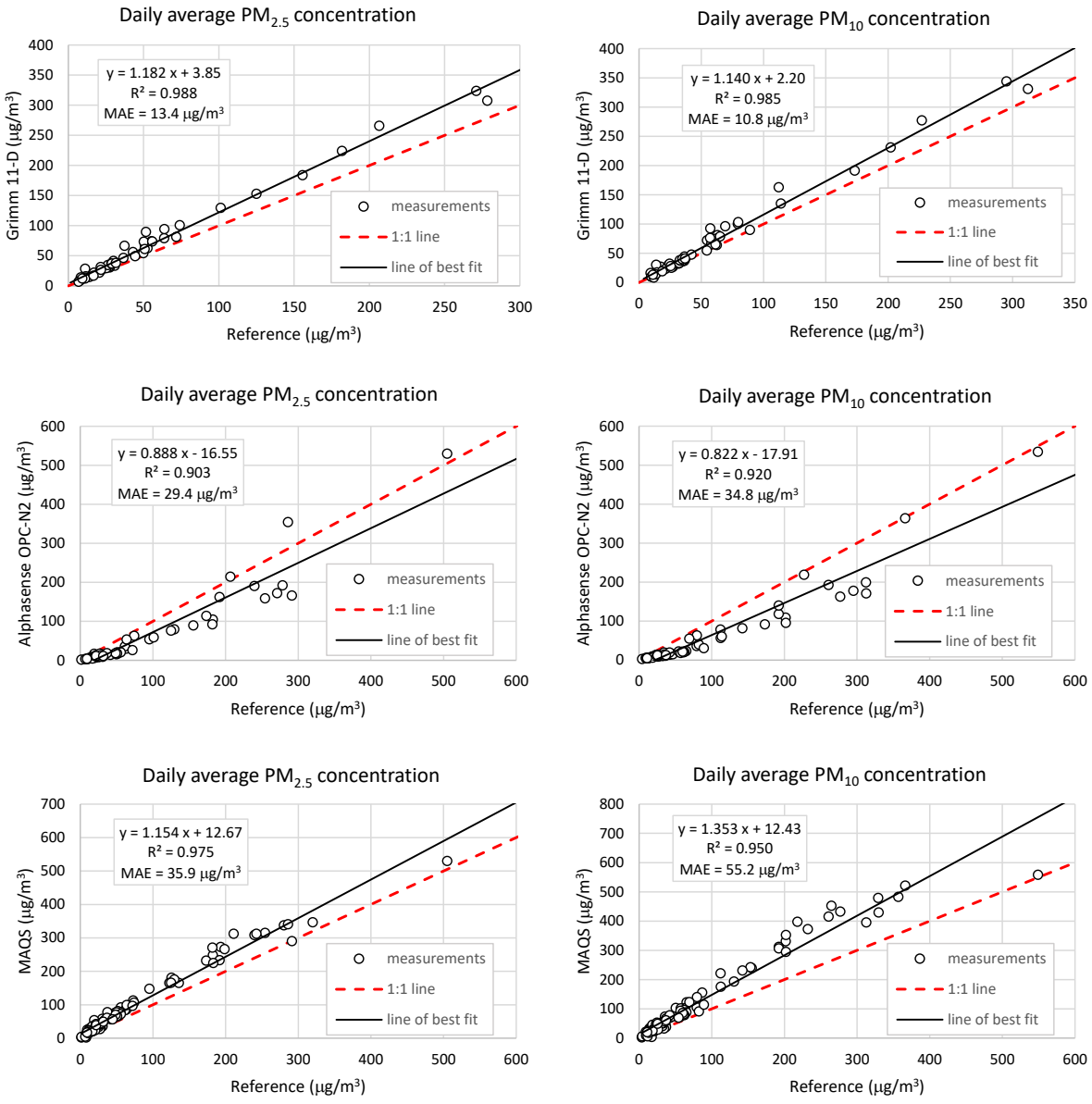


Figure 5. OPS performance during the period of strong pollution (12/2/2019–3/12/2020).

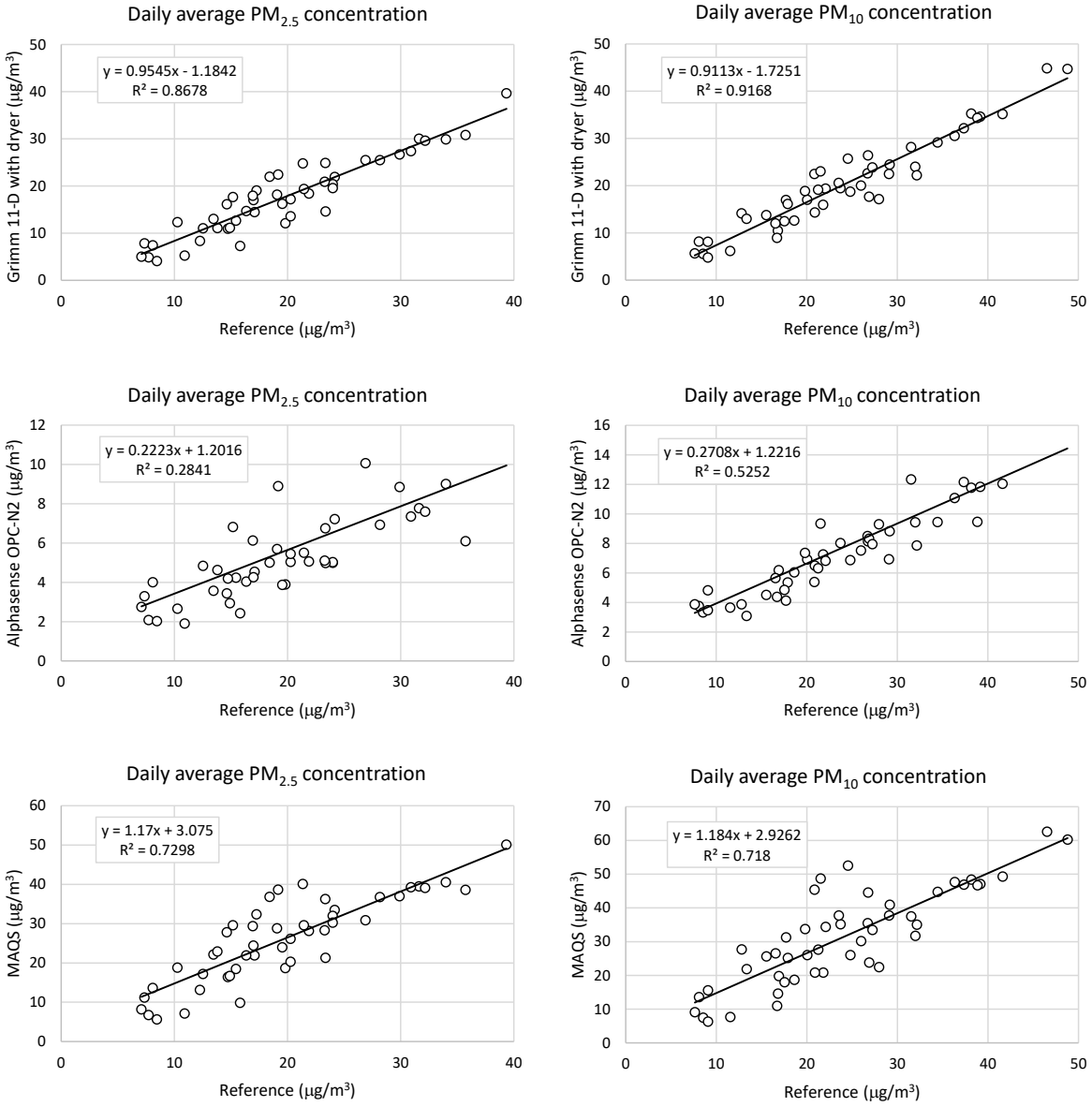


Figure 4. OPS performance during the period of mild pollution (3/13/2020–5/4/2020).

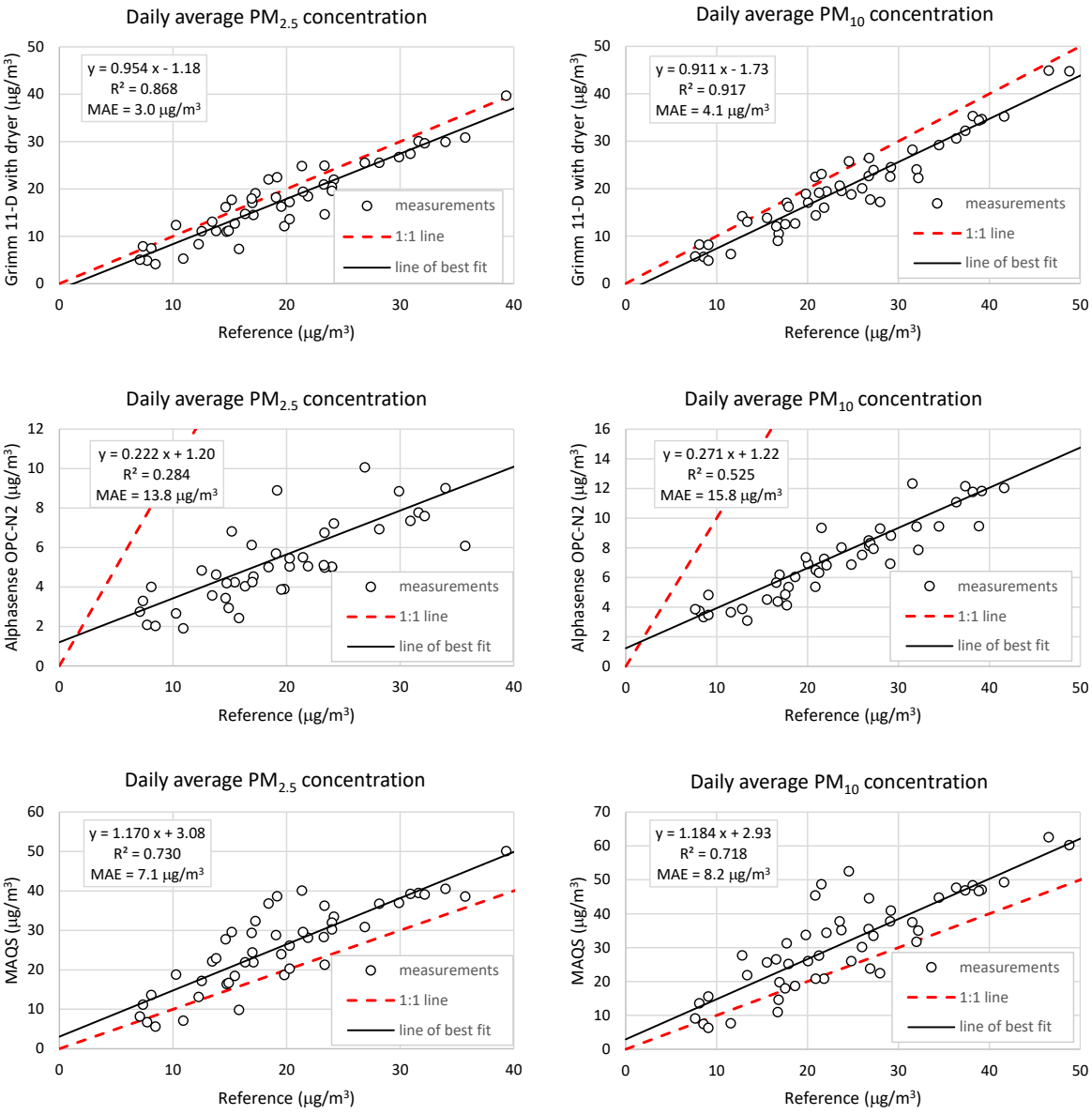


Figure 6. OPS performance during the period of mild pollution (3/13/2020–5/4/2020).

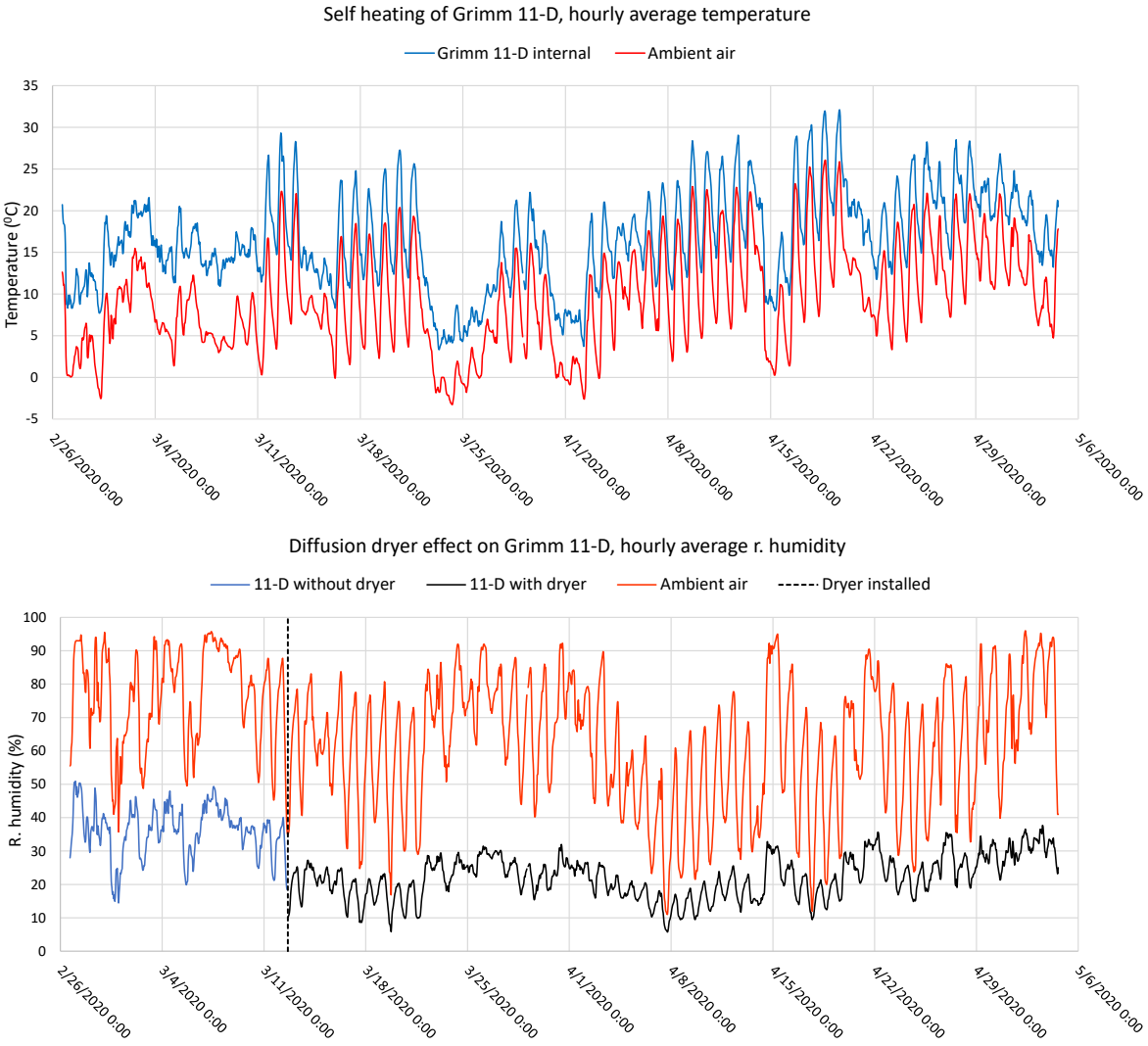


Figure 7. Self-heating and diffusion dryer effect of Grimm 11-D. Dryer was installed on 3/12/2020 15:30.

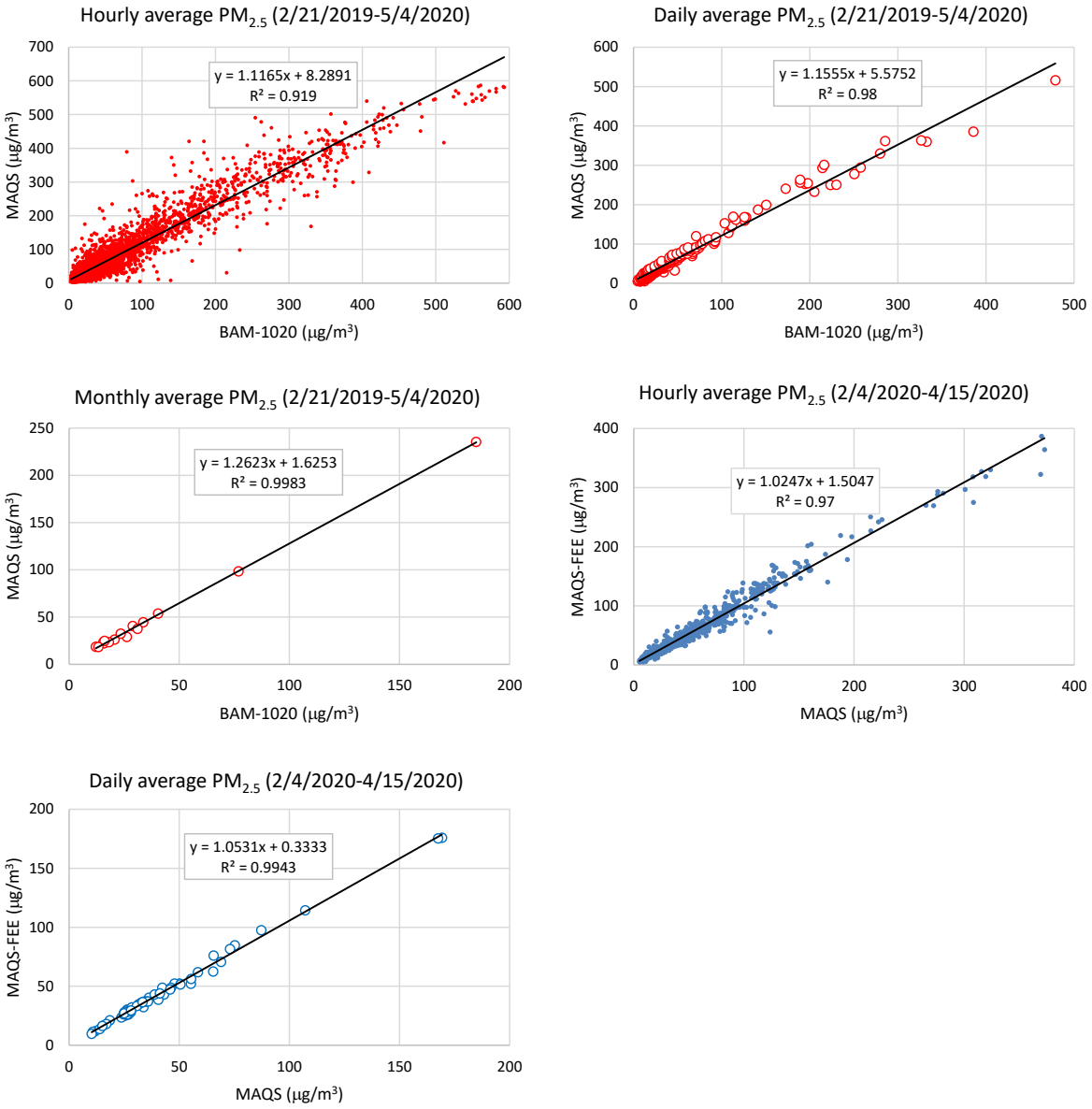


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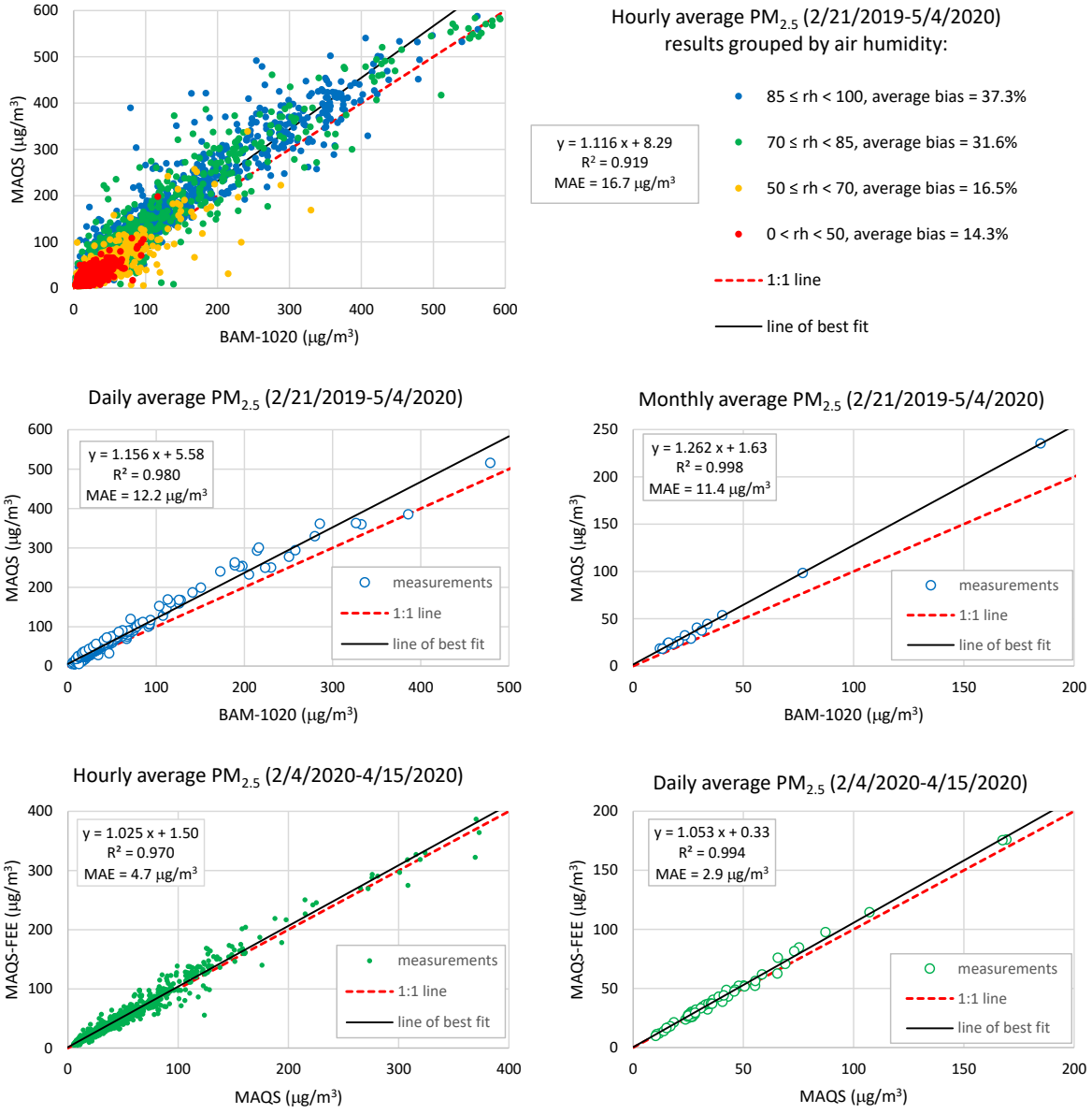
Figure 8. Long-term comparisons of MAQS sensor with BAM operated by US EPA at the nearby location: hourly, daily, monthly average values and comparison of hourly and daily average values of two MAQS sensors: first one (MAQS) at our main facility and second one (MAQS-FEE) at Faculty of Electrical Engineering in the immediate vicinity of BAM-1020.



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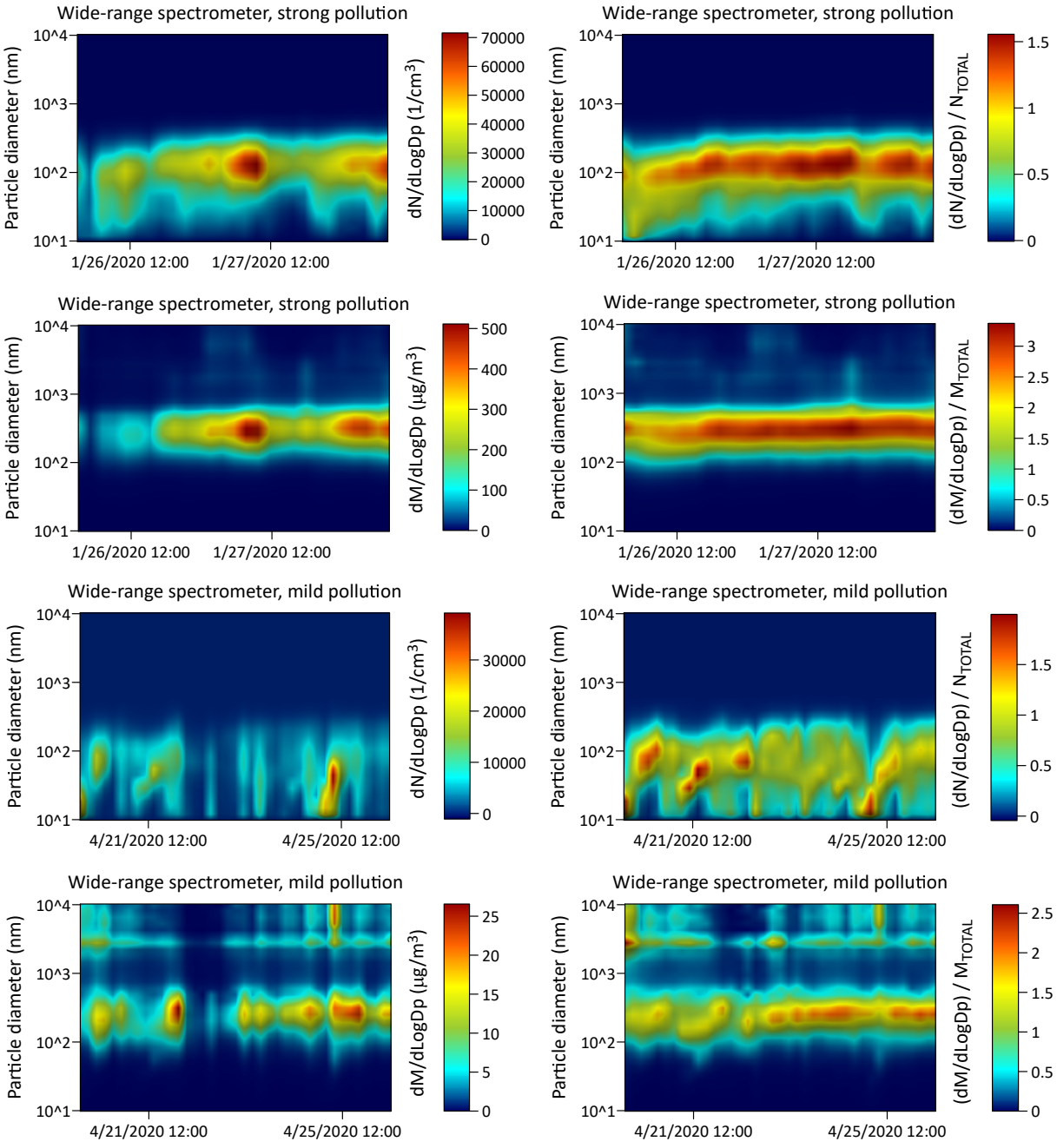

Figure 9. Wide-range spectrometer (SMPS+11D), hourly average values. Relative density 1.65 was applied on SMPS to calculate mass of particles.

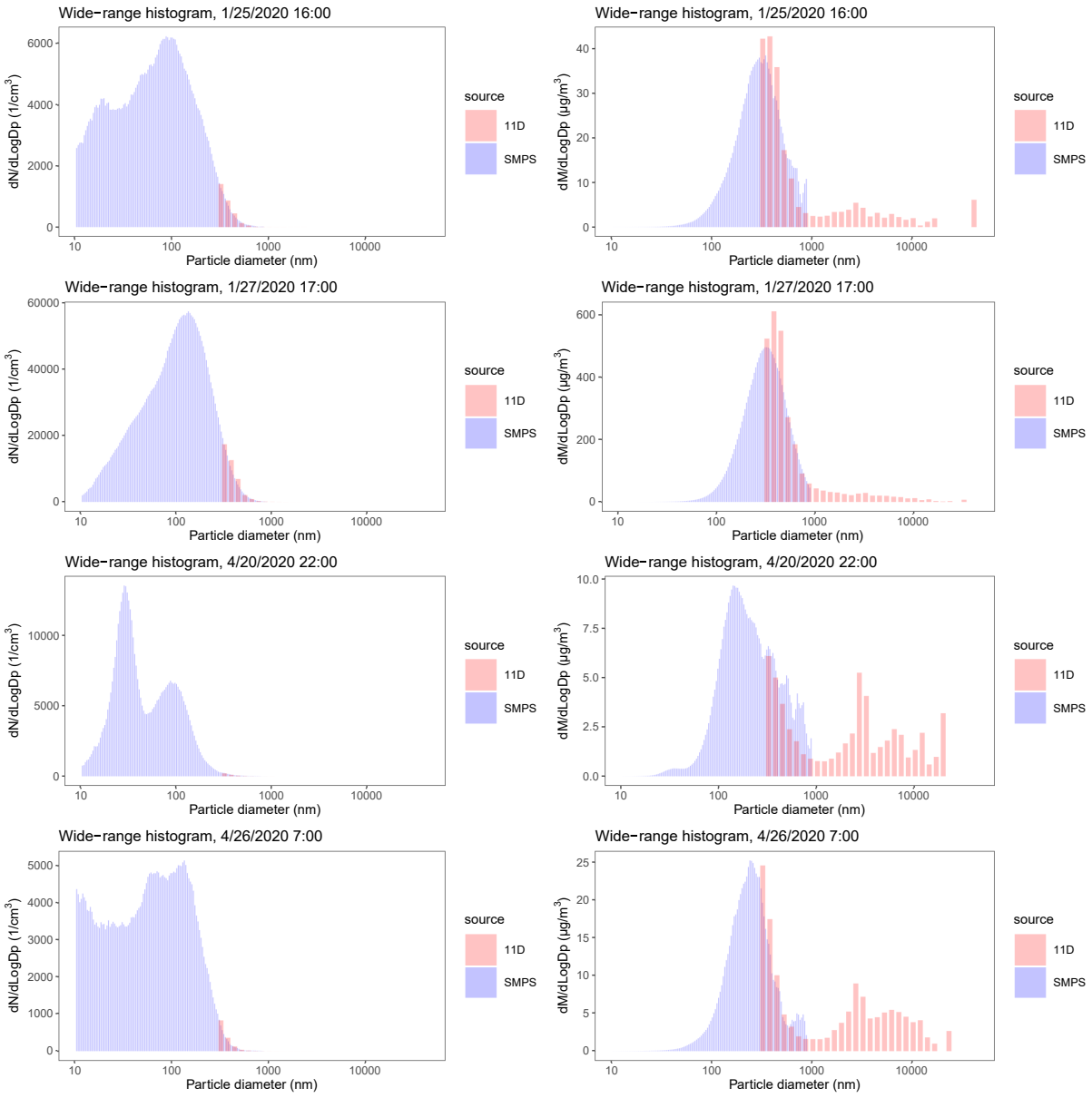

Figure 10. Wide-range histograms, hourly average values. Relative density 1.65 was applied on SMPS to calculate mass of particles.

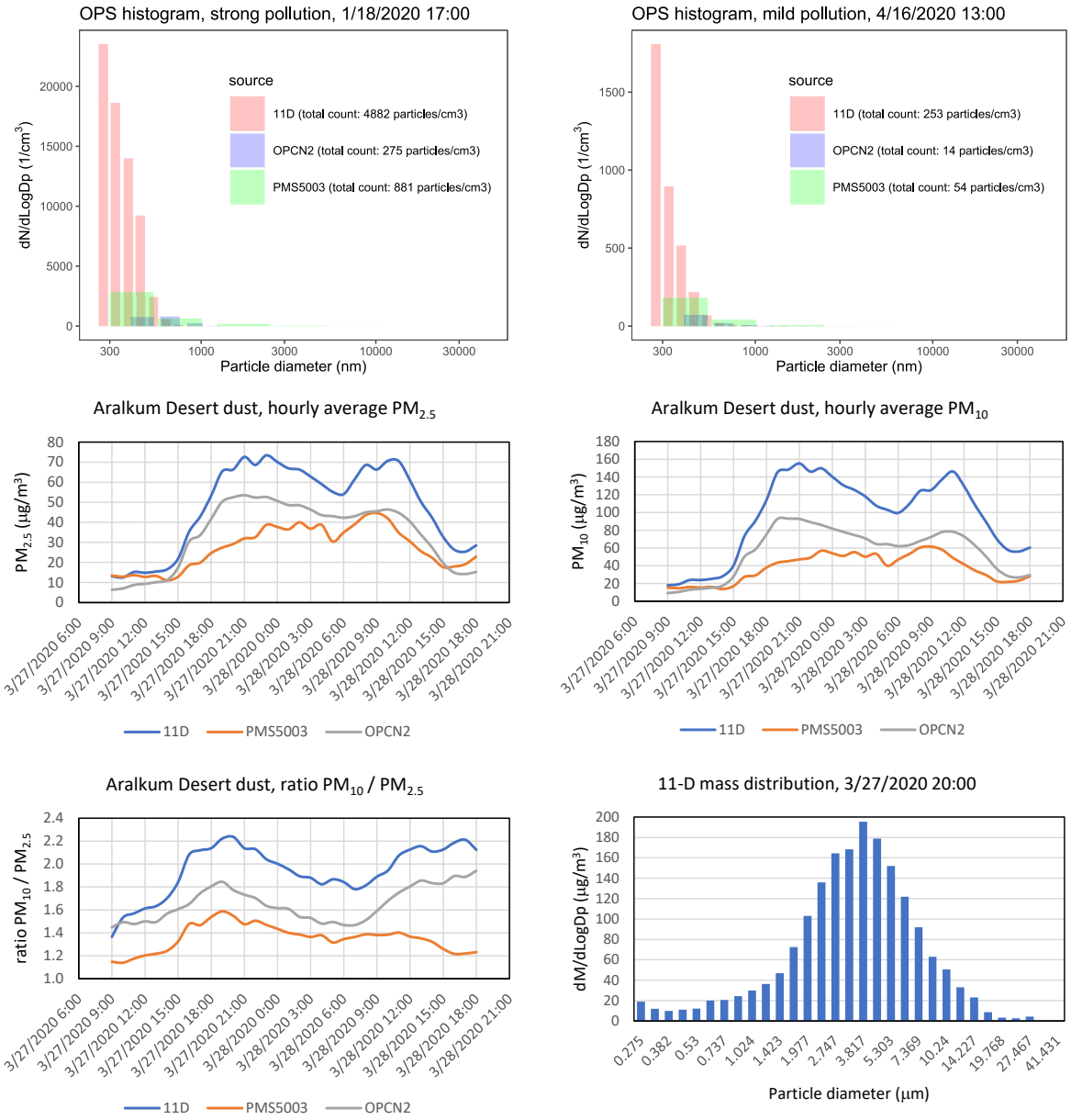

Figure 11. OPS histograms and Aralkum Desert dust episode, hourly average values.