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Verification of equivalence with reference method for measurements of PM_{10} concentrations using low-cost devices

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Abstract

This study presents an assessment of the equivalence of measurements of particulate matter PM_{10} concentrations using a low-cost electronic device as compared to the reference method. Data for the study were collected in accordance with the guidelines for research equivalence of the two devices operating in parallel. On this basis, a model correcting raw measurement results was developed. The best results were obtained for the model having the form of a second degree polynomial and taking into account air temperature. Corrected measurement results were used in the equivalence testing procedure. As a result, confirmation of equivalence was obtained for the vast majority of data sets generated from original measurements. This confirms the usefulness of the device as a tool for monitoring air quality.

Introduction

Correct measurement of concentrations of environmental pollutants is currently one of the biggest challenges of air monitoring. Measurements of particulate matter concentrations in ambient air play a special role. Increasing numbers of measurements contributes to a significant increase in the precision of emission forecasts but results in a need to reduce costs. The usual proper method for this type of measurement is the gravimetric method. It is quite expensive, requires cooperation with a specialized laboratory, and results may not come until several weeks after the measurement, while the measurements themselves are carried out with accuracy for one day at a time. This method cannot therefore be used to create a relatively cheap network monitoring concentrations of pollutants, for which the measurements need to be available in real time. Alternatives to the gravimetric method include low-cost measuring devices that use a variety of methods to assess air quality. They eliminate all disadvantages of the reference method, but they also introduce their own problems (Owczarek & Rogulski, 2018; Owczarek, Rogulski & Badyda, 2018; Rogulski & Badyda, 2018; Sówka et al., 2018; Szulczyński & Gębicki, 2018).

Using an alternative to the gravimetric method yields different results. It is therefore necessary to assess whether the results obtained are sufficiently close to those expected. Are they random and what are the errors? Depending on these considerations, the results obtained with the alternative method may be considered equivalent to those from the reference method. The study of the equivalence of methods for assessing air pollution monitoring devices is described in "Guide to the Demonstration" (GDE) (Dorozhovets, 2007a; Dorozhovets, 2007b; EC Working Group, 2010; Gębicki & Szymańska, 2011; PN-EN 12341, 2014).

The methodology for demonstrating device equivalence proposed by GDE has two stages: demonstrating the repeatability of measurements for at least two tested candidate devices, and examining and assessing the sizes of the differences between concentration measurements from the candidate devices versus the reference method. These differences are examined using a tool called measurement uncertainty, the results of which can be understood as the probability of obtaining results that are significantly different from the real ones. In addition, it is required that the equivalence test is carried out repeatedly (at least twice) and under different weather and field conditions, and that the data are collected from devices in close proximity (EC Working Group, 2010).

Aim of the study

The devices used in this study had not yet been tested for equivalence with the reference method for measuring PM_{10} concentrations. It was therefore uncertain whether the results obtained from the devices were correct. There are a few publications on this device, for example (Owczarek & Rogulski, 2018; Owczarek, Rogulski & Badyda, 2018) but the scope of the collected data did not allow a full equivalence test; there was too short a period of measurement and the placement of devices did not fully comply with the guidelines for equivalence testing.

This study aims to demonstrate the equivalence of PM_{10} measurements made using low-cost sensors compared to the reference method, and the usefulness of these sensors for measurements outside the State Environmental Monitoring system. These results will allow one to apply for a certificate of compliance of equivalence with the reference method for these devices.

The additional purpose of the test is the construction of a uniform function correcting the raw measurements of the analyzed devices to comparable values. This function could be placed in device controllers and could correct the received measurements on an ongoing basis.

Therefore, two questions were asked:

1. Is it possible to construct an effective corrective function and what is its form?
2. Is the device equivalent to the reference method after implementing this function?

Answers to these questions will have a significant impact on the further development of the tested device.

Measurement data

This study concerns measuring devices containing low-cost PM sensors using the optical method. The sensors suck outside air into a chamber, illuminate it with laser light, and then assess the concentrations of pollutants in the air by counting the number of reflections. Each sensor of this type, depending on the type of laser used and wavelength of the reflected light, can analyze the content of various pollutants in the air. This study focused on concentrations of particulate matter PM_{10}, i.e. dust with a diameter of no more than 10 μm.

Measurements of PM_{10} concentrations were conducted in Nowy Sącz between February and July 2018. We used a measuring device containing two low-cost PM sensors located a few meters from the measuring station belonging to VIEP. The intakes of the measuring device belonging to VIEP and the low-cost sensor devices were at the same height. The device using low-cost sensors generated measurements of PM_{10} concentrations every minute. These measurements were then aggregated to hourly averages and later daily averages. After removing unreliable observations from the sample using the Grubbs test, 129 observations were obtained from which two measurement campaigns were distinguished: winter, consisting of 47 measurements from February 1st to March 28th, and spring-summer, including 50 measurements from May 11th to June 30th. The results are presented in Figures 1 and 2 (Grubbs, 1950; ECS, 2013; GIOŚ, 2019).

Based on Figures 1 and 2, clear differences in the concentrations obtained by the reference method and the candidate method can be stated. These differences are particularly pronounced on days with low average daily air temperature. It is therefore necessary to correct measurements obtained from the tested devices in order to obtain comparable results. Many different functional correction models were tested using various independent variable vectors. The coefficient of determination and residual variance were used as measures to assess the quality of models. The best results of such correction were obtained with the model using a second- degree polynomial based on the indications of the candidate device and average air temperature (Boggs & Rogers, 1990; Myers, 1990; Leng et al., 2007; Green, Fuller & Baker, 2009; Czechowski, 2013).

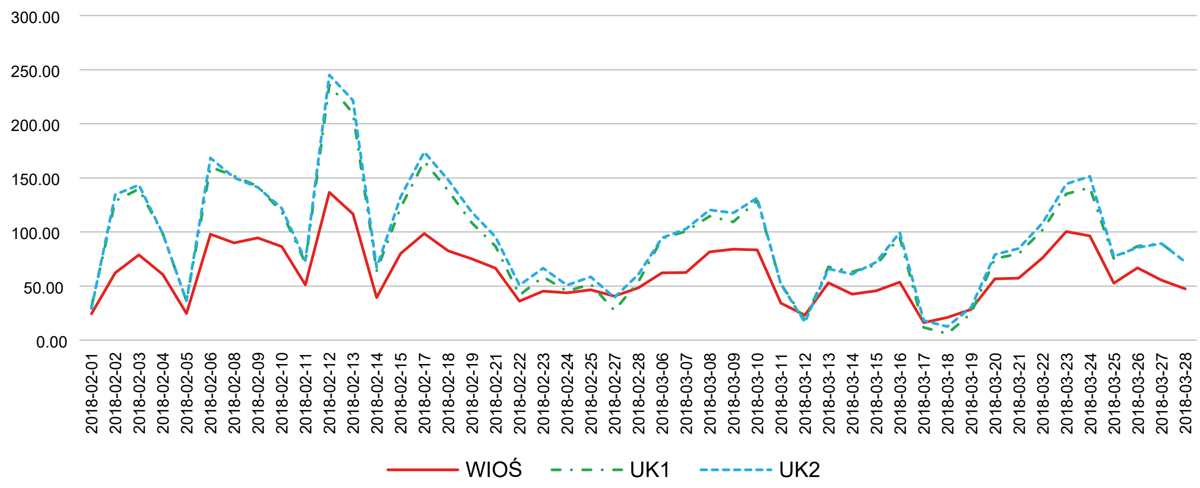

Figure 1. PM_{10} concentrations (in µg/m^3) from the reference method (VIEP) and tested devices (UK1 and UK2) in the winter campaign

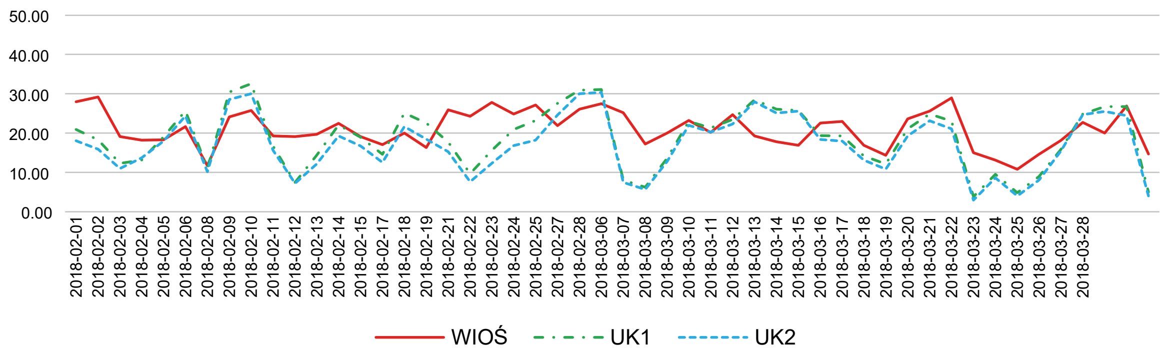

Figure 2. PM_{10} concentrations (in µg/m^3) obtained by the reference method (VIEP) and with tested devices (UK1 and UK2) in the spring-summer campaign

Ultimately, the correction model took the form:

$$y_{Ki} = 14.337 + 0.53 \cdot y_i - 0.0002 \cdot y_i^2 + 0.027 \cdot T_i \quad (1)$$

where: y_i – measurement values of the tested device on the i-th day, T_i – average temperature on that day.

The correction function could be implemented in the device driver using low-cost sensors, thanks to which it will be possible to use the obtained results without further processing. The adjustment of measurements from the electronic device after correction to the reference data is presented in Figures 3 and 4.

Methodology

After correction, a satisfactory concentration adjustment was obtained (Figure 3 and Figure 4).

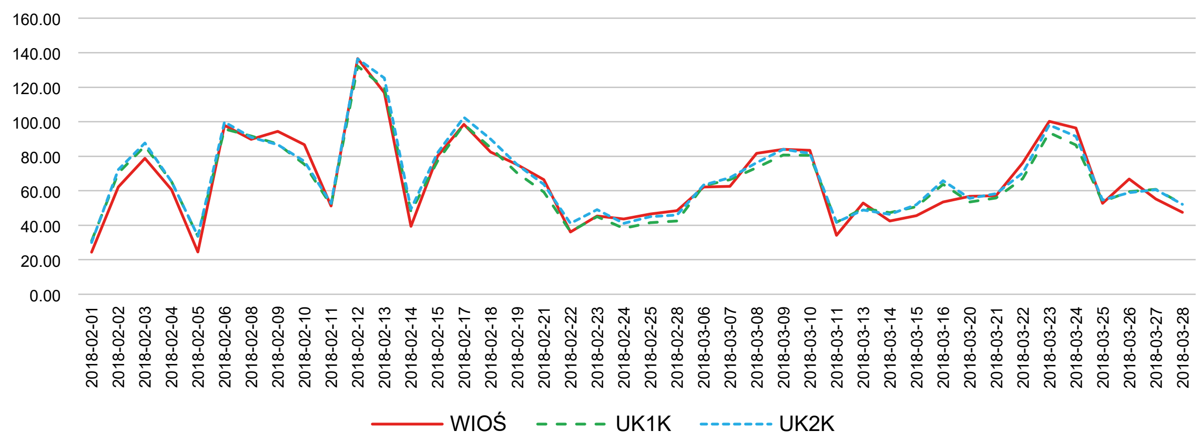

Figure 3. PM_{10} concentrations (in µg/m^3) obtained by the reference method (VIEP) and with tested devices (UK1 and UK2) after correcting the results with a second degree polynomial in the winter campaign

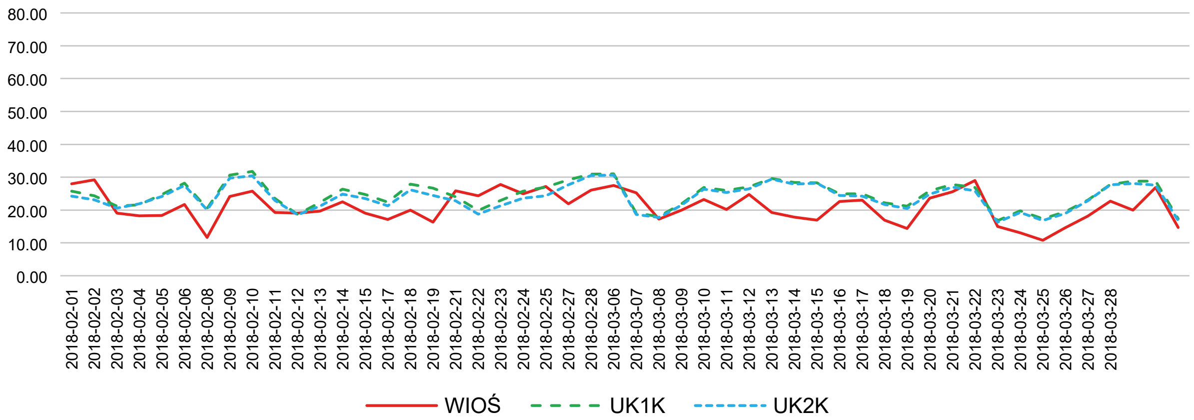

Figure 4. PM_{10} concentrations (in μg/m^3) obtained by the reference method (VIEP) and with tested devices (UK1 and UK2) after correcting the results with a second degree polynomial in the spring-summer campaign

This allowed for an equivalence procedure. First, the repeatability of results obtained by both tested devices (after correction) was examined. For this purpose, the concept of uncertainty between rehearsals (*between-sampler/instrument uncertainty*) was used, as described by the formula:

$$u_{BS}^2 = \sum_{i=1}^{n} \frac{(y_{1,i} - y_{2,n})^2}{2n} \tag{2}$$

The uncertainty of measurements is satisfactory if it does not exceed u_{BS} = 2.5 μg/m^3 and should be tested for all observations, and separately for observations above 30 μg/m^3 (high concentrations of PM_{10}). In both cases, the values of the calculated uncertainty does not exceed the limit value.

Uncertainty for all observations is u_{BSo} = 1.527 while for high concentrations u_{BS30} = 1.977. On this basis, it can be concluded that the devices work and give similar results, recorded PM_{10} concentrations are reproducible, and differences in observed measurements are small.

The reference method was then compared with the candidate devices. The comparison is made for all collected data, broken down into measurement campaigns, and separately for observations with concentration values greater than or equal to 30 μg/m^3. It is also assumed that each of the mentioned sets should include at least 40 observations.

The basic measure used to compare candidate devices with the reference method is the total uncertainty of measurements. It contains estimates of all sources of measurement errors occurring in the equivalence testing process and can be expressed by the formula:

$$u_{CM}^2(y_i) = \frac{RSS}{n-2} - u^2(x_i) + [a + (b-1) \cdot x_i]^2 \tag{3}$$

where:

$u^2(x_i)$ – the measurement uncertainty of the reference method, most often 0.67 μg^2/m^6;

$[a + (b-1) \cdot x_i]^2$ – the measurement uncertainty arising from the deviation of the linear regression

$$y = a + b \cdot x \tag{4}$$

between the results of the reference and candidate methods from the identity function (it is assumed that in this model a is statistically insignificantly different from 0, while the directional factor b is statistically insignificantly different from 1);

RSS/(n–2) – the rest variance of the linear model.

Based on the total uncertainty (3), the relative total measurement uncertainty is constructed:

$$w_{CM}^2(y_i) = \frac{u_{CR}^2(y_i)}{y_i} \tag{5}$$

and extended measurement uncertainty:

$$W_{CM} = k \cdot w_{CM} \tag{6}$$

assuming y_i and 50 for PM_{10}, and k equal to the critical value in the t distribution for the corresponding number of degrees of freedom (GUM, 1999; Dorozhovets, 2007a, 2007b; EC Working Group, 2010; Working Group, 2013).

The candidate method may be considered correct if the value of the expanded uncertainty W_{CM} does not exceed the assumed level of allowable uncertainty for devices measuring PM_{10} set at 25%.

If the limit value is exceeded by the uncertainty (6), it is possible to use a calibration function built on the basis of a linear regression function (2) of the form:

$$y_{CAL} = \frac{y - a}{b} \tag{7}$$

to correct the concentration values obtained from the candidate method. After its application, the total measurement uncertainty can take the form depending on the significance of regression parameters (2):

$$u_{CM}^2(y_i) = \frac{RSS}{n-2} - u^2(x_i) + \left[c + (d-1)\cdot x_i\right]^2 + \\ + \left[u^2(a) + x_i^2 \cdot u^2(b)\right] \quad (8)$$

where $u(a)$ and $u(b)$ are standard errors of estimation of parameters a and b for function (2) and c and d are parameters of the new regression function calculated after calibration.

If the value of the expanded uncertainty W_{CM} still does not meet the criterion of 25% of the allowable uncertainty, the candidate method cannot be considered equivalent to the reference method.

Obtained results

In accordance with the "Guide to …" (EC Working Group, 2010) methodology, the values of extended measurement uncertainty were calculated for all data groups, i.e. for each candidate device and for all measurements, broken down into measurement campaigns, and for observations with values greater than 30. The calculations were repeated in all cases where it was necessary to use a calibration function. The results are shown in Table 1.

The analysis shows that device U1 successfully passed the equivalence test for all generated data sets. The values of expanded uncertainty were between 0.195 and 0.241 and were definitely lower than the allowable value of 0.25.

In the case of device U2, the tests carried out for all data and for the winter campaign gave positive results (expanded uncertainty values 0.21 and 0.22, respectively). In the case of the spring-summer campaign, the equivalence test result was negative. The value of expanded uncertainty (0.283) for uncalibrated data slightly exceeds the allowable value. Unfortunately, the use of the calibration function not only did not improve uncertainty, but rather significantly worsened it. A similar situation occurred for the data set containing observations over 30 µg/m^3 for device U2. The original uncertainty value (0.262) and the value after calibration (0.274) slightly exceed the limit-value.

It can be assumed that both negative equivalence test results were caused by imperfections of the corrective function (1). It will be necessary to further improve it using more data.

Conclusions

For the analyzed low-cost devices it is necessary to apply a correction function. This study showed that the function can be based on a second-degree polynomial using PM_{10} concentrations and temperature values. This function has the form:

$$y_{Ki} = 14.337 + 0.53\cdot y_i - 0.0002\cdot y_i^2 + 0.027\cdot T_i$$

The correction function should be integrated into the device controller so that the device results will more closely match the reference values.

The tested devices passed the equivalence test with the reference method in most of the tested data configurations, which should be considered satisfactory. It can be assumed that the PM_{10} concentration values obtained from mobile devices after correction well approximate the concentration values obtained by the reference method. The values of expanded uncertainty were from 0.195 to 0.241. Only in the case of a campaign covering warm days did the uncertainty expand to a negative value of 0.614 for the U2 device. Thus, the study showed that it is possible to apply for a certificate of equivalence for the tested devices.

It is necessary to continue research on devices containing low-cost optical sensors in order to

Table 1. Results of equivalence tests for low-cost measuring devices for all data groups

Device	Feature	All	Campaign 1	Campaign 2	Greater than 30
U1	Expanded uncertainty	0.195	0.218	0.201	0.241
	Calibration function	is not necessary	is not necessary	is not necessary	is not necessary
	Expanded uncertainty after calibration	–	–	–	–
	Result of the equivalence test	**Passed**	**Passed**	**Passed**	**Passed**
U2	Expanded uncertainty	0.210	0.220	0.283	0.262
	Calibration function	is not necessary	is not necessary	$y_{CAL} = 1.443y - 13.303$	$y_{CAL} = y - 4.307$
	Expanded uncertainty after calibration	–	–	0.614	0.274
	Result of the equivalence test	**Passed**	**Passed**	Not passed	Not passed

improve them. It is also necessary to carry out equivalence tests in other locations to verify equivalence for them.

The data obtained in this way should also be used to further improve the internal correction function so that the measurements obtained will be equivalent to those of the reference method under all conditions.

References

1. Boggs, P.T. & Rogers, J.E. (1990) Orthogonal Distance Regression. In: Brown P.J. & Fuller W.A. (Eds). *Statistical Analysis of Measurement Error Models and Applications*. Contemporary Mathematics 112, Providence Rhode Island, pp. 181–194.
2. Czechowski, P.O. (2013) *New methods and models of data measurement quality in air pollution monitoring networks assessment*. Gdynia Maritime University Press (in Polish).
3. Dorozhovets, M. (2007a) Uncertainty of linear orthogonal regression. *Pomiary Automatyka Kontrola PAK* 53, 31, pp. 31–34 (in Polish).
4. Dorozhovets, M. (2007b) Proposals for extending the methods for determining the uncertainty of measurement results according to the GUM Guide. *Pomiary Automatyka Robotyka* 1, pp. 7–15 (in Polish).
5. EC Working Group (2010) *Guide to the demonstration of equivalence of ambient air monitoring methods*. Available from: http://ec.europa.eu/environment/air/quality/legislation/pdf/equivalence.pdf [Accessed: October 15, 2019]
6. ECS (2013) *Ambient Air – Automated measuring systems for the measurement of the concentration of particulate matter (PM10; PM2,5)*. CEN/TS 16450. European Committee for Standardization.
7. Gębicki, J. & Szymańska, K. (2011) Comparison of Tests for Equivalence of Methods for Measuring PM10 Dust in Ambient Air. *Polish Journal of Environmental Studies* 20, 6, pp. 1465–1472.
8. GIOŚ (2019) *Measurement of particulate matter in the air*. [Online] Available from: http://powietrze.gios.gov.pl/pjp/content/show/1000919 [Accessed: October 10, 2019] (in Polish).
9. Green, D.C., Fuller, G.W. & Baker, T. (2009) Development and validation of the volatile correction model for PM10 – An empirical method for adjusting TEOM measurements for their loss of volatile particulate matter. *Atmospheric Environment* 43, 13, pp. 2132–2141.
10. Grubbs, F.E. (1950) Sample criteria for testing outlying observations. *Annals of Mathematical Statistics* 21, 1, pp. 27–58.
11. Working Group (2013) Grupa robocza Komitetu EA ds. Laboratoriów. Wyznaczanie niepewności pomiaru przy wzorcowaniu. Evaluation of the Uncertainty of Measurement in Calibration. EA-4/02 M: 2013 (in Polish).
12. GUM (1999) *Expressing measurement uncertainty. Guide*. Warszawa: Główny Urząd Miar (in Polish).
13. Leng, L., Zhang, T., Kleinman, L. & Zhu, W. (2007) Ordinary least square regression, orthogonal regression, geometric mean regression and their applications in aerosol science. *Journal of Physics: Conference Series* 78, 012084. DOI:10.1088/1742-6596/78/1/012084.
14. Myers, R.H. (1990) *Classical and modern regression with applications*. Duxbury Thomson Learning.
15. Owczarek, T. & Rogulski, M. (2018) Uncertainty of PM_{10} concentration measurement on the example of an optical measuring device. *SHS Web of Conferences* 57, 02008.
16. Owczarek, T., Rogulski, M. & Badyda, A. (2018) Preliminary comparative assessment and elements of equivalence of air pollution measurement results of portable monitoring stations with using stochastic models. *E3S Web of Conferences* 28, 01028.
17. PN-EN 12341 (2014) *Atmospheric air – Standard gravimetric measuring method for determining the mass concentrations of PM10 or PM2.5 fraction of particulate matter* (in Polish).
18. Rogulski, M. & Badyda, A.J. (2018) Application of the Correction Function to Improve the Quality of PM Measurements with Low-Cost Devices. *SHS Web of Conferences* 57, 02009.
19. Sówka, I.M., Chlebowska-Styś, A., Pachurka, Ł. & Rogula-Kozłowska, W. (2018) Seasonal variations of PM2.5 and PM10 concentrations and inhalation exposure from PM – bound metals (As, Cd, Ni): first studies in Poznań (Poland). *Archives of Environmental Protection* 44, 4, pp. 86–95.
20. Szulczyński, A. & Gębicki, J. (2018) The applicability of low-cost PM10 sensors for atmospheric air quality monitoring. *SHS Web of Conferences* 57, 02013.