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RELATIONSHIP BETWEEN THE CONCENTRATIONS OF $PM_{2.5}$ INDOORS OBTAINED BY USING THE OPTICAL AND GRAVIMETRIC METHODS: PRELIMINARY ANALYSIS

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Abstract
The paper presents preliminary analysis of measurements of the mass concentrations of $PM_{2.5}$ in the indoor environment, obtained with the use of two optical instruments: (Grimm device and low-cost sensor) and a sampler based on the gravimetric method (SKC). It was found that the measurement of $PM_{2.5}$ using an optical device with active sampling underestimates the actual mass concentration of this mode ($PM_{2.5}$), while measurement using an optical device with passive sampling of air overestimates the concentration of $PM_{2.5}$. It has been shown that the physical relationship between the mass concentrations of airborne particles obtained with an optical sensor (C_{sensor}) and concentrations obtained with the gravimetric method (C_{grav}) is not linear. However, for practical reasons, the correct ("true") concentration levels of $PM_{2.5}$ in an indoor environment can be estimated by converting sensor data according to a simple linear equation, i.e., $C_{grav} = a\ C_{sensor}$. The coefficient a for the sensor used was estimated at 0.45.

Keywords: **$PM_{2.5}$; Mass concentration; Gravimetric method; Optical method; Low-cost sensors; Indoor air.**

1. INTRODUCTION

It is well known that fine particulate matter ($PM_{2.5}$) is directly associated with air quality and strongly influences the health status of exposed people. However, due to the nature and sources of $PM_{2.5}$, concentrations of fine particles can vary considerably over a relatively short distance. For example, the analysis of the difference between the concentration of $PM_{2.5}$ on the roadside and the corresponding urban background in Zabrze in Upper Silesia in 2005 showed an average increase in mass concentration above 10 $\mu g\ m^{-3}$ [1]. Fixed monitoring stations may, therefore, give a poor indication of human exposure to $PM_{2.5}$. For example, it has been found that in California, existing monitoring infrastructure cannot adequately characterize spatial and temporal variability in urban $PM_{2.5}$ concentrations, nor human exposure levels [2]. Generally, the inadequacy of ground-based measurements limits environmental health analysis across many regions. Moreover, many people spend most of their time indoors. Because the ratio of indoor/outdoor particulate concentrations often vary, ambient $PM_{2.5}$ concentrations measured at air quality monitoring stations may give a false estimation of personal exposure.

A significant increase in the density of monitoring networks (including the monitoring of indoor air) can be obtained by applying portable low-cost sensors in air quality control. Technological progress in the production of low-cost sensors for air-quality monitoring over the last decade has become possible because of rapid advancements in the fields of material science, digital electronics and wireless communication. A variety of low-cost sensors for measuring air pollution

are now available on the market. However, it is necessary for these devices to be evaluated and their performance to be understood in order to properly interpret the results and reduce confusion when low-cost sensor measurements are not in agreement with measurements from regulatory-grade instrumentation [3]. When analyzing the strong and weak points of low-cost sensors, it should firstly be noted that cheap sensor networks can produce a continuous stream of information about the level and dynamics of changes in $PM_{2.5}$ concentrations. Unfortunately, in this regard, the results obtained by different researchers have led to different conclusions. For example, Yoo et al. [4] found that low-cost portable sensors are only likely to be beneficial for long-term air-pollution monitoring if enough of them are used and they are of adequate quality. On the other hand, Cavaliere et al. [5] field-tested a passive method for long-term, integrated $PM_{2.5}$ mass and specifications, and overall, their findings indicated a good level of accuracy and precision in terms of $PM_{2.5}$ in urban areas. Interesting results obtained by Castell et al. [6] showed better agreement at sites with low traffic than at high traffic sites. Jayaratne et al. [7] suggest that the low-cost $PM_{2.5}$ sensors should be calibrated individually for each sources in the environment of their intended use.

The technology used in aerosol sensors is based on analysis of scattered light on aerosol particles in a small volume of air within the sensors' working space. As a result of the measurements taken, number of concentrations of aerosol particles in individual fractions, including $PM_{2.5}$, can be obtained, which are then automatically converted into the mass concentration (assuming that the counted particles are balls), with a unit density (1 g cm^{-3}). However, when many aerosol particles of different density have various shapes (other than spherical), this can lead to serious errors. The second source of errors is the fact that classification of particles as individual fractions according to their size is realized in sensors on the basis of optical diameter, while $PM_{2.5}$, PM_5 and PM_{10} fractions are defined according to their aerodynamic diameter. Meanwhile, the relationship between optical diameter and aerodynamic diameter can change significantly when the particle morphology and its chemical composition change (especially chemical composition of the particle surface layer). For example, airborne particles containing mainly elemental carbon (e.g., soot) in their surface layer primarily absorb light, while particles with a surface consisting mainly of sulphates scatter light. This is important in field studies. For example, Pastuszka et al. [8] found that although airborne particles in four cities in southern Poland contain high amounts of elemental carbon, oxides and sulfates play an important role in promoting light reflectance onto aerosol particles during winter. Diffraction of light also depends on particle size. The relationship between mass concentrations indicated by sensors and mass concentrations obtained by the gravimetric method is, therefore, very complex and depends on aerosol characteristics, which, in turn, depend on a number of parameters, such as characteristics of the largest particle-emission sources, topography and meteorological conditions etc. It should be mentioned that calibration alone does not guarantee good results because a change in even one meteorological factor (for example, wind velocity) can rapidly change the optical characteristics of airborne particles in a studied area. When wind undergoes a change in direction, it can transport to the sampling point particles emitted from different sources, changing the average properties of studied aerosol [9, 10]. A similar effect can be observed when wind increases speed, transporting to the sampling point particles emitted from more distant sources. For example, in one study, XPS analysis of aerosol samples, as well as microscopic investigation of individual particles, showed that significant quantities of sulfur, oxygen and sodium were transported to the downtown area of Katowice from distant sources located in the eastern and southeastern sector [11]. Another important finding is hygroscopic growth of fine particles containing water-soluble material and transported from distant sources [12]. This phenomenon can strongly influence the optical properties of atmospheric aerosol [13]. The average optical properties of $PM_{2.5}$ can be also changed significantly near the busy roads [14].

For these reasons, it is very difficult to find a general algorithm that converts the concentrations obtained by sensors into concentrations that would be obtained by gravimetric methods. Anyway, studies of airborne-particle sensors have been developing very intensively. One of the latest reviews of current research into use of low-cost sensors for air quality assessment was held in Utrecht, the Netherlands, some years ago [15]. At the seminar, it was concluded that it would be a serious mistake not to take into account low-cost sensor networks, which can be an extremely valuable supplement to official national monitoring networks, provided they are synchronized using appropriate calibration modeling. A similar conclusion can be found by reading an overview of

this topic and possible future applications of low-cost sensors, published recently by the World Meteorological Organization [16].

So far, research carried out at various scientific centers around the world has focused on the determination of equations based on experimental data, comparing concentrations of PM_{10}, PM_5 and $PM_{2.5}$ ascertained by the gravimetric method with concentrations obtained from low-cost sensors [2, 6, 17–20]. Available results generally indicate that a number of these sensors could potentially be useful tools for characterizing $PM_{2.5}$ levels in particular, in ambient environments (if the data is interpreted and understood correctly) [3].

Although certainly atmospheric aerosol-concentration values obtained from sensor networks could be converted (using a special algorithm) into the concentration levels that would have been obtained using gravimetric devices, the scope of this study has been limited to indoor air. The influence of meteorological factors on optical properties of aerosol particles does not, therefore, need to be taken into account. However, the optical properties of fine particles indoors also vary depending on the characteristics of the indoor environment because of different possible emission sources, such as small, non-effective stoves, resuspension from carpets and cigarette smoking etc. [21, 22], as well as varying levels of airborne particles penetrating indoor air from outdoors [23].

The aim of this work was to find the relationships between concentration of fine airborne particles obtained by gravimetric method using active sampler and two optical methods using both active and passive sampling devices. In the next step it is discussed the possibility of converting indoor $PM_{2.5}$ concentrations obtained with low-cost sensors into "actual" equivalent mass concentrations that would have been obtained using the gravimetric method.

2. METHODS

The research was carried out in two apartments in two cities in Upper Silesia, Poland: Katowice and the area around Sosnowiec, where optical and gravimetric measurements were carried out simultaneously. The apartments studied had only natural ventilation. When measurements were being taken, windows remained closed. The temperature indoors ranged from 22 to 24°C, and relative humidity ranged from 62 to 66%. The equipment used in the study consisted of an SKC sampler (US), an optical Grimm instrument (Germany) and a low-cost laser aerosol sensor manufactured by Shenzhen More-Suns Electronics Co., Ltd. (China).

The SKC sampler consists of a filter head connected to a battery-powered pump, equipped with an electronic device that ensures constant pump performance during 24-hour measurements. The measurement head designed for collecting $PM_{2.5}$ particles uses an inertial impaction (with a cut-size diameter equal to 2.5 μm) to separate coarse particles (unlike other heads, where cyclones are used as coarse-particle selectors).

The Grimm instrument uses light-scattering technology to count individual particles, with a semiconductor laser as the light source. The dispersed signal from each particle passing through the laser beam is focused at an angle of about 90° in the mirror and reflected (transmitted) onto the receiving diode. The diode signal, after appropriate amplification, passes to the multichannel signal classifier. The pulse-height analyzer then classifies the signal transmitted in each channel. These values can be displayed and stored in the data memory card, to be sent to the computer for further analysis. Air is sucked into the device using an internal pump with a flow rate of 1.2×10^{-3} m^3 min^{-1} (1.2 liters/minute). The pump also provides necessary clean, protective air, which is filtered and passes through the casing air regulator, back to the optical chamber. This is to prevent dust contamination in the optical laser unit. This particle-free air flow is also used for the zero-reference test during auto-calibration.

Small, generally passive aerosol sensors were used in this study, equipped with a miniature laser. Counting and classification of airborne particles present in the measurement space are based solely on the optical analysis. The manufacturer does not provide any construction details, but determination of numerical concentrations in individual particle fractions and conversion into mass concentrations are based on the assumption that all particles are balls with a density of 1g m^{-3}.

During measurement, $PM_{2.5}$ concentrations (indicated by the sensor and Grimm instrument) were read every hour. Average daily values of $PM_{2.5}$ concentrations were then compared with $PM_{2.5}$ mass concentrations obtained via the gravimetric method using the SKC sampler.

However, at the beginning, concentrations of $PM_{2.5}$ outside one of the apartments in Katowice were measured using a low-cost laser aerosol sensor. These data were compared with the concentration values of

airborne fine particles obtained simultaneously at a monitoring station located very close to the apartment (at the Institute for Ecology of Industrial Areas).

3. RESULTS AND DISCUSSION

Figure 1 shows the relationship between the concentration of $PM_{2.5}$, obtained in the outdoor air in Katowice with the use of the low-cost optical sensor and monitoring data. It can be seen that although this study was only conducted in the summer (June to August), the correlation between the sensor and the monitoring data is rather weak ($r^2 = 0.60$). It should be noted that Fig. 1 shows only the statistical relationship. The physical dependence must take into account the straight line starting point (0;0), which means that the computed correlation would be even much weaker. This result agrees with a number of previous literature data and support the general thesis that it is not so easy to use the concentration values obtained by a low-cost sensor to find the real mass concentration of $PM_{2.5}$. Therefore, this study is oriented into finding such relationship for indoor air where the number of various factors influencing this relationship is strongly limited.

Comparison of airborne-particle concentrations obtained using the two optical instruments (Grimm device and low-cost sensor) and the gravimetric method (SKC sampler) are presented in Table 1. Preliminary analysis of the obtained data can be made using indicators of deviation from the gravimetric concentrations (Δ), defined as follows [24]:

$$\Delta_{sensor} = \frac{|C_{graw} - C_{sensor}|}{C_{graw}} \quad (1)$$

$$\Delta_{Grimm} = \frac{|C_{graw} - C_{Grimm}|}{C_{graw}} \quad (2)$$

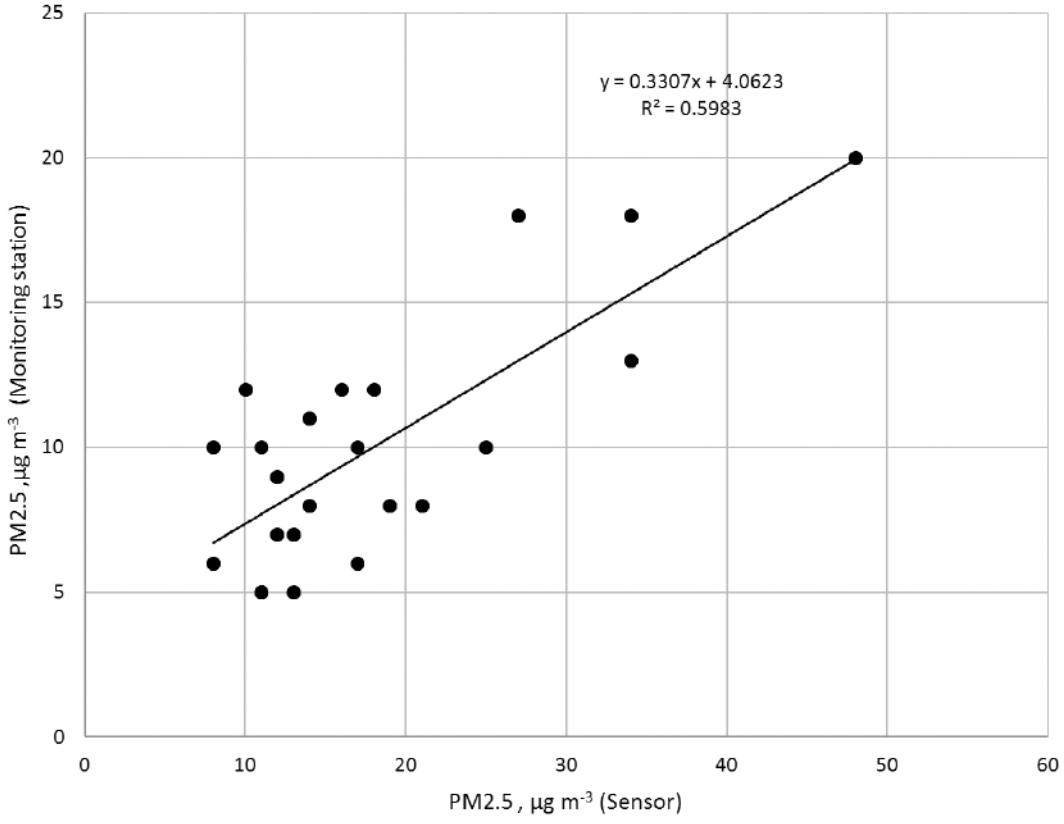

Figure 1.
Statistical dependence between the concentrations of $PM_{2.5}$ in the atmospheric air in Katowice, measured with a low-cost sensor, and data obtained simultaneously from a nearby air monitoring station

Table 1.
Concentration of $PM_{2.5}$ obtained by using the Grimm instrument, the low-cost sensor and the SKC sampler (gravimetric method). Source: [23]

Day	Concentration of $PM_{2.5}$, μg m^{-3}			Δ_{Grimm} %	Δ_{Sensor} %
	Grimm	Sensor	Grav. (SKC)		
27.10.2017	4.0	15.0	12.4	67.7	21.0
8.11.2017	8.3	30.0	17.3	52.0	73.4
22.11.2017	12.3	39.0	18.3	32.8	113.1
17.12.2017	37.1	51.2	27.5	34.9	86.2
4.01.2018	5.1	13.0	10.4	51.0	25.0
7.01.2018	23.9	57.2	24.1	0.8	137.3
14.01.2018	2.8	11.5	4.2	33.31	173.8
20.01.2018	17.5	48.5	23.0	23.9	110.9
28.01.2018	41.1	117.6	46.6	11.8	152.4
3.03.2018	21.5	64.3	31.0	30.6	107.4
Arithmetic mean	17.4	44.7	21.4	33.9	100.1

The calculated results showed a 100% difference (relative deviation) between the indoor $PM_{2.5}$ mass concentrations obtained by using a low-cost sensor and the results obtained by the gravimetric method, while the so-determined deviation of concentrations measured simultaneously with the Grimm instrument was about 34%. These results clearly indicate that low-cost dust sensors can currently only be used for preliminary analysis of air pollution. On the other hand, certain published results show that data obtained from the sensor and gravimetric method are well-correlated (see the Introduction and [25, 26]), although it should be noted that some researchers found only a moderate linearity of sensor responses [27]. The question arises, however, of whether this good correlation has only a random statistical character or not. It is, therefore, important to analyze physical phenomena used in the measurement of particulate matter concentrations with gravimetric and optical instruments.

The fundamental question is whether the ratio of mass concentrations of airborne particles obtained using gravimetric and optical methods (C_{grav}/C_{opt}) is linear or not. If this ratio can be described by the linear function, calibration of the optical method should be very simple. If not, calculation of "real" $PM_{2.5}$ mass concentrations using data obtained from optical devices becomes more complicated. Especially, if this relationship could be described by the exponential function, the ratio would be very sensitive to even small changes in different environmental factors. In this context, it is important to note that light scattering provides an extremely sensitive tool for measuring the concentrations and particle size of aerosols. Unfortunately, one disadvantage of light-scattering instruments is that scattering may be sensitive to small changes in the refractive index, scattering angle, particle size and particle shape, which can lead to confusing or misleading results [28]. Active optical devices (with a pump) have the additional problem of recovery time. It cannot be assumed that each particle contained in the sample flow produces a simple count. In practice, counting losses due to coincidence occur. A less-than-10% loss in particle counts (approximately) is required [29, 30] due to recovery time (t_r) (typical t_r is 20 μs), which refers to the time between successive count events, including the transit time for particles passing thorough the light beam and the pulse-processing time of the multichannel analyzer. With active optical instruments, coincidence errors are particularly important, which arise when two or more particles are in the sensitive volume of the optical instrument at the same time, causing a spurious signal that leads to underestimation of particle-number concentrations and overestimation of particle size. The additional discussion on this subject can be found in the supplementary materials.

The sensitive volume is the region from which signals are generated. It is defined by the incident and scattered beams and the size of the aerosol stream. The ratio of the observed count N_O to the true count N_t for an instrument with a sensitive volume of v_s is given by Hinds [28]:

$$\frac{N_O}{N_t} = \exp\left(-N_t v_s\right) \tag{3}$$

Assuming that, in this case, in active optical instruments equipped with an air pump, mass concentra-

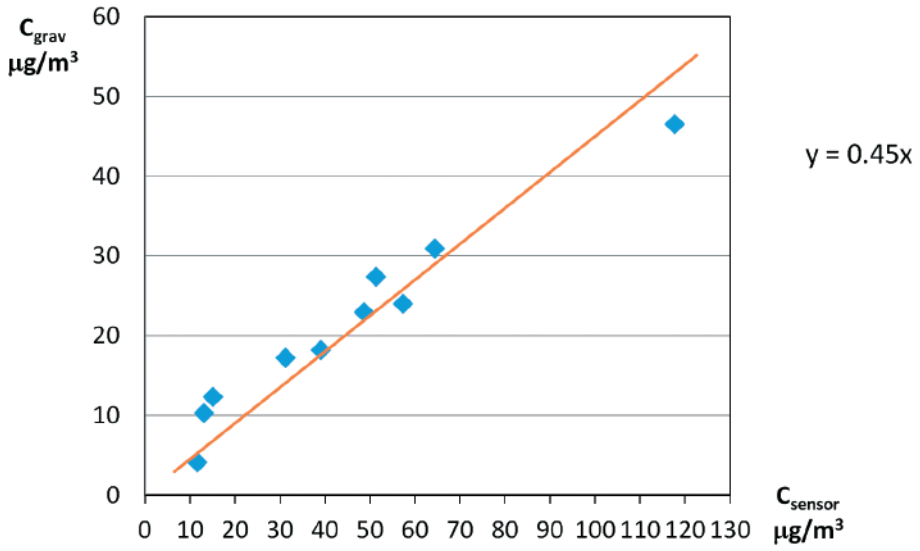

Figure 2.
Mass concentration levels of $PM_{2.5}$ (C_{grav}) corresponding to the concentration values obtained by the sensor (C_{sensor}). Results are calculated according to equation (6) and compared to the experimental data. (Correlation coefficient = 0.9780; Spearman coefficient = 0.9746)

Table 2.
Contribution of airborne particles with a diameter between 1 and 2.5 µm to the mass concentration of $PM_{2.5}$ indicated by the Grimm instrument and the low-cost sensor

Day	Concentration of airborne particles, µg m^{-3}				$\frac{PM2.5-PM1}{PM2.5}$, %	
	PM_1		$PM_{2.5}$			
	Grimm	Sensor	Grimm	Sensor	Sensor	Grimm
27.10.2017	3.4	11.0	4.0	15.0	26.7	15.0
30.10.2017	1.0	3.0	1.4	4.0	25.0	28.6
3.11.2017	2.9	11.1	3.5	15.0	26.0	17.1
8.11.2017	7.6	19.0	8.3	31.1	38.9	8.4
21.11.2017	5.5	15.8	6.1	22.0	31.8	9.8
22.11.2017	11.3	28.0	12.3	39.0	28.2	8.1
17.12.2017	33.4	33.3	37.1	51.2	35.0	10.0
4.01.2018	4.5	9.3	5.1	13.0	28.5	11.8
7.01.2018	21.9	39.2	23.9	57.2	31.5	8.4
14.01.2018	2.6	9.0	2.8	11.5	21.7	7.1
20.01.2018	15.8	34.0	17.5	48.5	29.9	9.7
28.01.2018	38.8	74.5	41.1	117.6	36.6	5.6
3.03.2018	18.1	43.1	21.5	64.3	33.0	5.8
Arithmetic Mean	12.8	25.3	14.2	37.6	30.2	11.2

tion is proportional to the number concentration (C = σN)*, the following relationship between mass concentrations obtained by the optical instrument (observed concentrations) (C_{opt}) and mass concentrations obtained by the gravimetric method ("true" concentrations) (C_{grav}) can be written as follows:

*$C \approx (\rho v_{particle} N)/V_{sampled\ air}$ where ρ and $v_{particle}$ is the averaged particle density and volume, respectively, while N is a number of sampled particles in active optical instruments equipped with an air pump

$$\frac{C_{opt}}{C_{grav}} = \exp(-C_{grav}k) \quad (4)$$

where k is the coefficient containing v_s.

Equation (4) shows that concentrations obtained using the optical instrument should be lower (underestimated) in relation to the concentrations obtained by the gravimetric method. This conclusion is supported by comparing results obtained using the Grimm instrument with results obtained using the gravimetric method (Table 1).

With passive samplers (like low-cost sensors), the number of particles present in the analyzed space is much smaller than in active samplers, so overestimation of particle size will be a much more important factor in the coincidence error than underestimation of particle-number concentrations (see the supplementary material). As a result, the mass concentration of the measured aerosol fraction will be significantly overestimated.

A simple calculation indicates that the averaged ratio of mass concentration of coarser $PM_{2.5}$ particles (i.e., particles with a diameter between 1 and 2.5 μm) to the total $PM_{2.5}$ concentration is 11% for data obtained with the Grimm instrument and 30% for data obtained with the sensor (Table 2).

Given that coarse particles contribute more to mass concentrations than the fine fraction (mass is proportional to d^3), we can expect that the mass concentration of all $PM_{2.5}$ obtained by sensors will be significantly greater than the concentration of $PM_{2.5}$ obtained by the gravimetric method, which is confirmed in Table 1.

The mass optical concentration obtained by the sensor (C_{sensor}) and level of "gravimetric" concentration (C_{grav}) can, therefore, be connected by a more general equation, which is different to (4):

$$C_{sensor} = C_{grav}\, F(C_{grav}, k) \tag{5}$$

where factor/function F depends on C_{grav} and k.

Equation (5) indicates the basic relationship between $PM_{2.5}$ concentrations measured using sensors and the gravimetric method. As can be seen, this relationship is rather complicated, and notably, it is not linear. However, we believe that for every indoor environment and for a limited range of concentration values, this relationship can be significantly simplified. For our data, the value of F can be calculated for all C_{grav} and C_{sensor} data contained in Table 1. The results are within the range of 1.15 to 4.65, with a mean value of 2.2, which means that the dependence of the approximate model above can be written as follows:

$$C_{sensor} = C_{grav}\, F(C_{grav}, k) \tag{6}$$

where $a = 1/F_{average} \approx 0.45$

To better illustrate this relationship, Figure 2 has been prepared using the values contained in Table 2. It can be seen that equation (6) surprisingly well describes the relationship between C_{sensor} and C_{grav}. The correlation is very strong (0.97) and the relationship: $C_{grav} = 0.45\, C_{sensor}$ can be applied to convert the concentration of $PM_{2.5}$ obtained by the used sensor to concentration levels that could be obtained by the gravimetric method (using SKC sampler) in the indoor environment. Therefore, although the relationship between mass concentrations of airborne particles obtained with the optical sensor (C_{sensor}) and concentrations obtained by the gravimetric method (C_{grav}) is not linear in nature, in an indoor environment, for a relatively limited range of concentrations, a linear function can approximate this relationship very well. Unfortunately, it should be emphasized that such a dependence cannot generally be expected in the case of measurements in atmospheric air due to a number of disturbing factors. For example, Johnson et al. [31] found that although their sensor (PPD42NS) displayed good agreement with a reference during laboratory testing with incense smoke, the sensor had effectively no agreement with reference monitors in any of the measurement environments. Among the various possible reasons for such a result, hygroscopic growth of particles certainly seems to be very important. In circumstances where the relative humidity approaches 100%, there is a possibility of mist or fog droplets, which are detected as particles [32]. Recently, Jayaratne et al. [33] showed that even deliquescent growth of particles and formation of atmospheric fog droplets can lead to significant increases in particle-number concentrations and mass concentrations reported by such sensors.

However, on the basis of existing knowledge, it can be assumed that for selected areas, changes in physical-chemical characteristics of the aerosol will be closely related to meteorological conditions. Hence, the calibration parameters might change over time depending on the meteorological conditions and the location [6]. On the other hand, it seems that development of an appropriate database of the physicochemical parameters of aerosol particles associated with meteorological parameters should enable an appropriate algorithm to be developed:

$$C_{grav} = \mathrm{f}(C_{sensor}, \text{coordinate of the sampling point, meteorological parameters}) \tag{7}$$

This will allow calculation of mass concentrations obtained by sensors for correct mass concentrations that would otherwise have been obtained using standard equipment at monitoring stations. It seems that to achieve this goal, it is necessary to use advanced data-analysis methods based on the so-called internet of things (IoT).

4. CONCLUSIONS

It was found that measurement of $PM_{2.5}$ using an optical instrument with active sampling underestimates actual mass concentrations of this mode, while using an optical device with passive sampling overestimates $PM_{2.5}$ concentrations.

The relationship between mass concentrations of airborne particles obtained with an optical sensor (C_{sensor}) and concentrations obtained with the gravimetric method (C_{grav}) is not linear in nature, which significantly limits use of low-cost optical sensors in practical field measurements.

However, the relationship between C_{sensor} and C_{grav} can be adequately approximated by a linear function in an indoor environment, for a relatively limited range of concentrations. In our study, the correct ("true") concentration levels could be obtained by multiplying individual values indicated by factor a, which was estimated as 0.45.

To convert a mass concentration of atmospheric aerosol measured with low-cost sensors into concentration levels obtained using the gravimetric method, a different approach is needed. It seems that developing an appropriate database of physicochemical parameters of aerosol particles associated with meteorological parameters should, in turn, enable an appropriate algorithm to be developed.

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