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CONCENTRATION CHANGES OF PM₁₀ UNDER LIQUID PRECIPITATION CONDITIONS

ZMIANY STĘŻENIA PM₁₀ W WARUNKACH WYSTĘPOWANIA OPADÓW CIEKŁYCH

Abstract: This study reports the results of field research into variability of the scavenging coefficient (A) of suspended dust comprising particles with aerodynamic diameters less than 10 μm . Registration of PM₁₀ over 7 years in conditions of the occurrence of rainfall (convective light showers, large-scale precipitation and storms) was undertaken in an undeveloped rural area. The analysis involved 806 observations taken at constant time intervals of 0.5 hour. The measurements of the concentration of PM₁₀ were performed by means of a reference method accompanied by concurrent registration of basic meteorological parameters. It was found that, for PM₁₀, the scavenging efficiency is considerably influenced by rainfall intensity R and the type of precipitation. In the case of convective precipitation, data on A are only partially related to “classical approach” of rain scavenging. Within the range of comparable values of rainfall intensity, the type of wet deposition (except for storms) does not influence the effectiveness of scavenging PM₁₀ from the ground-level zone. The large number of observations conducted in real-time conditions yielded a proposal of simple regression model, which can be deemed suitable for the description of variability A ($D_{\text{PM}10}$), but only to a limited extent for large-scale precipitation. The collected results can be applied in air pollution dispersion models and deposition and were found to be generally representative for areas with similar climatic characteristics.

Keywords: aerosol, wet deposition, washout, non-urban area

Introduction

The scavenging of particles of atmospheric aerosol during the phenomenon when particles collide and merge with rain drops involves both in-cloud and below-cloud processes [1, 2]. Scavenging constitutes a process which leads to the removal of pollutants from the atmosphere and plays a principal role in the transmission of pollution from the atmosphere to the ground [3]. Hence, it is one of the most important processes that ensure that a balance is maintained between the sources and outflow of aerosol particles [4].

Wet below-cloud scavenging involves all the phenomena by means of which particles are removed from the air through a number of various types of precipitation: rain, snow, fog and ice. According to [5], from the point of view of human health and the quality of the atmosphere, below-cloud scavenging seems to be more important due to the fact that particles of all types and sizes are deposited and transmitted to the ground-level zone in this

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process. This statement is supported by the fact that the PMs which form an immediate hazard to human health are usually deposited as a result of below-cloud scavenging and the principal mechanism involves the collision of solid particles with rain droplets [6].

The process of wet aerosol scavenging is very intricate since it is affected by a number of external factors, which include: size of droplets, distribution of particle size, chemical composition of water, rainfall intensity, temperature of environment and even chemical and physical properties of droplets and aerosol and the area in which collision between aerosol and droplets occurs [7]. The process of understanding wet particulate matter scavenging is at a stage when more insight is being gained into it step by step. The application of schemes of wet deposition plays an important role in the modelling of long-range transport of air pollution as well as in modelling the transport of chemical compounds.

Below-cloud scavenging of aerosol particles is usually described on the basis of the concept of collisions between rain droplets and particulate matter. Nevertheless, the mechanisms of below-cloud scavenging, including the effect of inertia, Brownian diffusion, thermophoresis, diffusiophoresis and electro-scavenging, have been thoroughly recognized and described [2, 4, 8-14]. One paper [15] in particular contains formulae that enable the researcher to assess the effectiveness of the particular mechanisms in the processes of below-cloud scavenging of aerosols.

The actual effects of washout of particles accumulated in the troposphere during episodes of precipitation are usually determined by means of the scavenging coefficient Λ [s^{-1}] [16], which is considered to be the most important parameter characterizing below-cloud scavenging [17, 18]. For a particle with a given size, the scavenging coefficient is the function of the boundary velocity of drops and the effectiveness of the collision between the droplets and particles of the atmospheric aerosol [8]. However, it was noted that due to the large number of parameters which play a role in the below-cloud scavenging processes (such as the above: effectiveness of the collisions, critical droplet velocity and distribution of rain droplets and aerosol particle sizes), the scavenging coefficient has a large degree of variability [19]. Therefore, the correct parametrization of its properties plays a relevant role in both climate models and models concerning the distribution of pollution [20].

The effectiveness of wet deposition in removing aerosol particles from the troposphere, which is expressed in terms of the scavenging coefficient, has been the subject of numerous research papers, in particular theoretical ones, *eg* [2, 7-9, 13, 21-26]. The experimental research into below-cloud scavenging is realized via a number of different aspects. These processes are dealt with both in a complex manner with a distinction between the effectiveness of scavenging of solid particles corresponding to the specific types of precipitation, and in a detailed manner, when the effectiveness of the scavenging of specific particles is investigated in regard to the type of precipitation which carries them. Experimental research often focuses on measurements in the direct vicinity of anthropogenic sources of emissions, urban areas as well as remote ones (marine). Consequently, some of the results pertain to local conditions while, on the other hand, local emission of pollutants and the structure of clouds have a considerable effect on the wet deposition [3]. Besides, the changes in the concentration of aerosols in the troposphere after the occurrence of precipitation resulting from the extensive effect of horizontal air masses, could also occur in areas both in the vicinity of and distant from the source of pollution [8]. With small exceptions [27, 28], experimental research is limited to continuous, yet short-term observations [29-31] or regards several instances with a small span in time and

pertains to one type of precipitation [32-34]. With the exception of [30, 31, 34], authors do not consider the case of coarse mode of PM₁₀ scavenging.

This paper reports the results of research into the variability of the scavenging coefficient for PM₁₀ in the ground-level zone in the atmospheric conditions that lead to the development of rainfall. The principal objective in this work is associated with analysis of the variability of the scavenging coefficient depending on: type of precipitation, rainfall intensity and the conditions such as movement of air masses, corresponding to the instances of various types of precipitation. The paper undertakes to verify hypothesis that the weather conditions during liquid hydrometeor type deposition in specific conditions do not have an effect on the variability of concentration of particulate matter.

Materials and methods

The testing was performed over a period of 7 successive years (2007-2013). In order to minimize the effect of anthropogenic sources, the concentration of PM₁₀ was measured in an undeveloped area, *ie* in the vicinity of a village (Kotorz Maly, Poland, 50°43'37"N; 18°03'22"E, 1,025 inhabitants). The measurement point was located in an open, yet shielded meadow area protected by the surrounding wood - 9 km from the border of a provincial town (Opole, 122,000 inhabitants) and 1.5 km from the nearest compact rural building development. The measurement campaign involved the observation of the concentration of PM₁₀ occurring as a result of incidents of three types of precipitation (convective light showers without thunder, storms and large-scale precipitation).

Meteorological parameters and PM₁₀ sampling procedure

A portable weather station was used to determine weather conditions. Portable stations are used for registration of weather conditions in tests on the effect of rainfall on aerosanitary conditions [35]. This weather station was installed 12 m from the PM aspirator. The sensors, which determined relative humidity (*RH*), temperature (*T*), atmospheric pressure (*Ap*), wind speed (*Ws*), wind direction (*Wd*) and rainfall (*R*), were installed at a height of 2 m above the ground. The standard measurement uncertainty was equal to: 0.5% for *RH*, 0.5°C for *T*, 0.06 hPa for *Ap*, 0.06 m s⁻¹ for *Ws* and 1° for *Wd*. The weather station is equipped with a bucket rain gauge with limited capacity of water (2 cm³). Within a short period of observation, the rain detector is not sensitive to precipitation of very low intensity. Consequently, the results do not include data on precipitation with the intensity below 0.2 [mm h⁻¹].

The procedure by which the measurement of the concentration of PM₁₀ was performed conformed with the European standard [36]. The reference method, which is often relied on [1, 4], was also applied in this case. The aspiration of the PM₁₀ in the air was carried out by a MicroPNS HVS16 sequential dust sampler. Similarly to the case of the sensors in the weather station, the aspiration header was installed 2 m above ground level. The flow rate was 68 m³ h⁻¹. The PM separators applied Whatman GF/A fibreglass air filters with a diameter of 150 mm. Prior to and after aspiration, the filters were seasoned over 24 hours in conditions of constant temperature and humidity and subsequently their weight was determined by a differential scale (RADWAG XA 52/2X). The aspiration at a constant time interval of 0.5 h was conducted directly before and during the occurrence of precipitation. The expanded concentration measurement uncertainty (*U*) did not exceed

3.2%. The time interval guaranteed the PM collection to a degree that was sufficient to determine the mass of the captured particulate matter, even in conditions when its concentration in the air was low [37] and ensured that the effect of synoptic processes and activity of the sources of PM emission on the variability of aerosols was limited [38]. The initial testing ($n = 25$, time interval of registration - 10 seconds, time of a single registration - 1800 seconds) using DustTrak 8520 Aerosol Monitor - TSI®, was conducted in variable weather conditions; however, with the exception of rain, it did not yield considerable differences in the results of PM₁₀ concentration over 10 and 1800 seconds in the investigated area.

Scavenging coefficient calculation

The scavenging coefficient of the particles with a given diameter D_p can be expressed with the relation [11, 39]:

$$\frac{dn_M(D_p)}{dt} = -\Lambda(D_p)m_M(D_p) \quad (1)$$

where $\Lambda(D_p)$ is the coefficient of scavenging particles with diameter D_p .

Hence, $\Lambda(D_p)$ denotes the relative change of the aerosol mass in a specific time for particles with diameter D_p , resulting from the below-cloud scavenging with the rain droplets. Consequently, using the relation in (2), the scavenging coefficient can be derived on the basis of measurements, knowing the initial concentration c_0 at time t_0 and concentration c_1 at time t_1 ($t_1 - t_0$ determines the duration of the precipitation) [40]:

$$\Lambda(D_p) = -\frac{1}{t_1 - t_0} \ln \left[\frac{c_1(D_p)}{c_0(D_p)} \right] \quad (2)$$

The scavenging coefficient is relative to the aerodynamic diameter of the PM (D_p); however, due to the applied measurement methodology, the entire fraction of PM with the diameter below 10 μm was identified. In addition, it was assumed that the scavenging as a result of rainfall forms a mechanism of suspended particle removal from the ground-level zone [27, 41]. Concern assumption may seem controversial, since the formula (2) can be applied only if rain scavenging is the only sink in a spatially homogeneous system [27]. Such conditions are not certainly during convective rainfall. However, the assumption allows the comparison of the effectiveness of the removal of aerosols during rainfall events of different origin, as well as an estimate of the role of convection, advection and turbulence in the dust removing process.

Statistical analyses

An initial analysis with the application of the Kolmogorov-Smirnov test indicates that the registered values of the specific meteorological parameters (with the exception of RH and T for storms) and the calculated values of the scavenging coefficient are not characterized with the normal distribution. Consequently, statistical analyses which verify the initial hypotheses were limited to the application of non-parametrical tests. The analysis applied all instances which are characterized by varied values in terms of the concentration of PM₁₀ as well as the type and intensity of the rainfall.

All statistical operations were undertaken by means of the STATISTICA 12® program.

Results and discussion

Characteristics of meteorological conditions

Table 1 presents descriptive statistics of the meteorological parameters which describe the conditions in which the experiment was undertaken. The measurement campaign lasting for over 7 years yielded the results of 806 instances accompanied by the potential changes in the PM₁₀ concentration. Thus, this study contains a significant basis for further consideration.

The mean annual concentration of PM₁₀ in the examined area was equal to 26.3 $\mu\text{g m}^{-3}$. The majority, *ie* around 77%, of the observations involved frontal precipitation. Almost 20% of the measurements yielded the results of short-term convective precipitation with various intensities. The study was complemented by 25 observations involving changes in PM₁₀ concentration during storms. Convective light showers and showers without electrical charges reaching the earth were principally encountered during the summer season (corresponding to the period of higher emission from natural sources), while large-scale showers took place over the entire year and were usually associated with the transition of weather to cold fronts (in 76% of cases).

Table 1
Meteorological parameters characterizing the conditions during the observations

Type of precipitation	No of 0.5 hour observ.	Descriptive statistics	<i>T</i> [°C]	<i>RH</i> [%]	<i>R</i> [mm h ⁻¹]	<i>Ws</i> [m s ⁻¹]	PM ₁₀ <i>c</i> ₀ [$\mu\text{g m}^{-3}$]	PM ₁₀ <i>c</i> ₁ [$\mu\text{g m}^{-3}$]
Convective	161	avg	19.0	80.7	1.60	3.10	16.9	13.9
		med	18.9	84.0	0.80	2.30	16.2	13.6
		<i>SD</i>	3.62	12.0	1.71	2.91	7.80	6.60
		min	3.63	62.0	0.20	0.00	5.40	2.70
		max	28.9	94.0	7.40	16.3	42.0	38.0
		<i>C_V</i> [%]	19.1	15.2	109.6	93.0	46.0	47.7
Convective - thunderstorm	25	avg	19.7	80.0	9.40	5.60	27.0	10.4
		med	19.7	81.0	6.40	4.80	29.0	9.20
		<i>SD</i>	3.15	0.11	8.29	4.12	5.80	4.80
		min	14.6	51.0	1.60	0.00	10.0	2.00
		max	24.3	95.0	37.0	16.8	36.0	18.4
		<i>C_V</i> [%]	16.0	13.9	87.8	74.0	21.5	46.1
Frontal (large scale or stratiform)	620	avg	9.30	87.9	0.90	5.00	16.4	14.5
		med	9.10	90.0	0.50	2.70	16.0	14.0
		<i>SD</i>	4.17	0.08	0.98	6.30	8.10	7.40
		min	0.00	68.0	0.20	0.00	4.80	1.10
		max	27.2	99.0	13.0	58.8	59.0	49.0
		<i>C_V</i> [%]	45.1	9.48	112.1	125.5	49.5	51.4

Obviously, with regard to a single occurrence, frontal precipitation was characterized by a considerably longer duration than the remaining types of rainfall. Consequently, this is reflected in the considerable difference in the number of observations taken every half an hour. In accordance with the classification that is commonly applied [42], the largest number of registered cases (48%) corresponds to light showers with precipitation in the range $R \leq 0.5 \text{ mm h}^{-1}$. Light showers were not noted during storm occurrences. For large-scale and convective showers not accompanied by thunder, the proportion of light rainfall was equal to 52 and 40%, respectively. Intermediate rainfall, *ie*, in the range from

0.6 to 2 mm h⁻¹ for frontal rain, was registered 247 times, while for convective rainfall the figures were 58 times and 5 times for storms. In addition, 76 instances of precipitation with rainfall levels from 2.1 to 5 mm h⁻¹ were registered. For the case of large-scale precipitation, their proportion was equal to 7%, while storms and convective precipitation were registered in 16 and 17% of cases, respectively. Heavy rainfall (> 5 mm h⁻¹) was most often encountered during storms (64% of cases) and incidentally during convective light and large-scale precipitation, with the proportions of 7 and 0.6%, respectively.

During the experiments, the relative humidity varied but was at a comparable level for all types of precipitation. With respect to both types of the observed convective precipitation, the variability in the air temperature was also small. The largest variation in this parameter is noted for frontal precipitation during the whole season. Beside the intensity of hydrometeors, the velocities of the horizontal air masses are also highly variable. However, 18% of all instances were found not to be accompanied by wind. Horizontal movement of air masses was displaced from the north (45%) and south directions (33%), *ie* from areas with considerable environmental quality and low air pollution [43, 44]. In merely 13% of noted instances (in the case of air movement from the west and north-west), the influx of air masses originated from areas with high pollution, *ie* from the area of Opole with a considerable level of PM pollution.

Scavenging coefficient λ . Variability analysis

Figure 1 contains a collective interpretation with an illustration of the values of λ calculated for the specific types of precipitation.

The variability in the concentration of aerosols over two successive time intervals in actual conditions is related to a number of factors, *eg* turbulence in the boundary layer, chemical processes in the liquid phase, as well as potential emission and transport of pollution from remote areas [27]. These processes are reflected in both positive and negative values of the scavenging coefficient. Over the course of the studies, the occurrence of negative values of λ was incidentally noted for light rains, which confirms the observations made by Laakso et al [27]. Detailed analysis of the cases reported there indicates that light rains accompanied the cold season, where horizontal air masses were displaced from the NW and W directions (*ie* from areas with high pollution). Consequently, one is right to note that in the conditions of the actual field measurements it is impossible to totally isolate the test spots from all factors which influence (even to a very small degree) the relative variable.

The intensity of atmospheric aerosol displacement is most often derived as a mean on the basis of the measurement data. One can note that, with the exception of storms, the values of λ are similar for convective and large-scale precipitation. Nevertheless, the analysis conducted by means of the Kruskal-Wallis (ANOVA) test rejects this claim completely (p -value = 0.0023). One can risk a statement that the higher fall in the PM₁₀ concentration in the ground-level zone after convective precipitation (with a median of $\lambda = 8.06 \times 10^{-5} \text{ s}^{-1}$) than after large-scale precipitation (median of $\lambda = 5.61 \cdot 10^{-5} \text{ s}^{-1}$) results not only from the effect of the precipitation and its intensity, but also from the transport of aerosol with the upwelling filaments. The most efficient scavenging is specific to the precipitation that accompanies storms, where the value of the calculated median $\lambda = 5.33 \cdot 10^{-4} \text{ s}^{-1}$ is nearly an order of magnitude higher than for other types of precipitation. This situation can, however, be explained by means of the difference in the rainfall,

although a number of researchers, such as Nicolson et al [45] and Chate and Pranesha [33], do not report this kind of relation. The considerably higher value of Λ during thunderstorms is undoubtedly associated with the ionization of the air and particle charging [13], as well as the effect of phoretic forces [33], which are considered as processes which aid in the scavenging process. It can be stated, that especially during convective rainfall, the aerosol concentration changes due to external factors can dominate the effects caused by the interaction with hydrometeors.

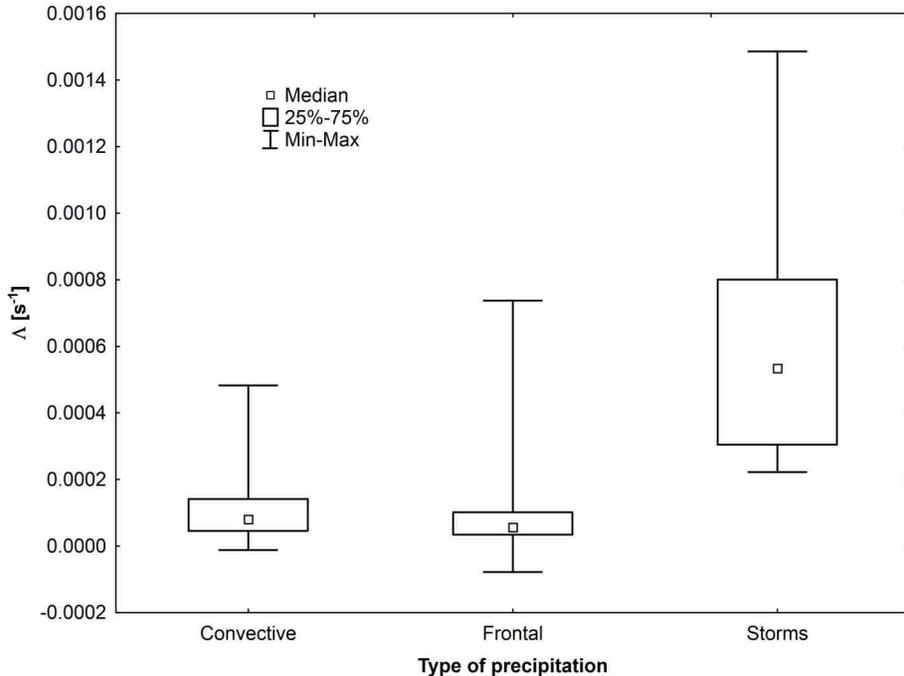


Fig. 1. Range of the calculated scavenging coefficient $\Lambda_{(PM_{10})}$ [s^{-1}] for all types of precipitation

Table 2 summarizes the effects of the Spearman correlation between the registered parameters which characterize the weather conditions and the calculated value of the scavenging coefficient PM₁₀ for the investigated types of precipitation.

Table 2

Results of Spearman correlation between Λ and others parameters for different types of precipitation

Type of precipitation	Temp. [°C]	RH [%]	R [mm h ⁻¹]	Ws [m h ⁻¹]
convective	0.081	-0.055	0.922*	-0.278**
thunderstorm	-0.169	0.373**	0.998*	0.026
frontal	0.191**	-0.030	0.718*	-0.156

*significant at $p < 0.001$; **significant at $p < 0.01$

By reference to the results to the Guilford scale [46], one can see that the wind speed, relative humidity and tropospheric air temperature do not affect the value of the scavenging

coefficient derived for PM_{10} . Despite statistically relevant correlations found between the values of RH and A for the case of storms, as well as Ws and A for convective precipitation, nevertheless, the relations which were established are low and characterized by low values of the A coefficient. The initial results indicate the high dependence between the scavenging coefficient and rainfall intensity (for large-scale precipitation) and the very high relation for convective precipitation.

Figure 2 presents collective graphical interpretation of the relations between A and R . The detailed data (for rainfall intensity in the range from 0.2 to 2.0 $mm\ h^{-1}$) for the examined types of precipitation are presented at Figure 3.

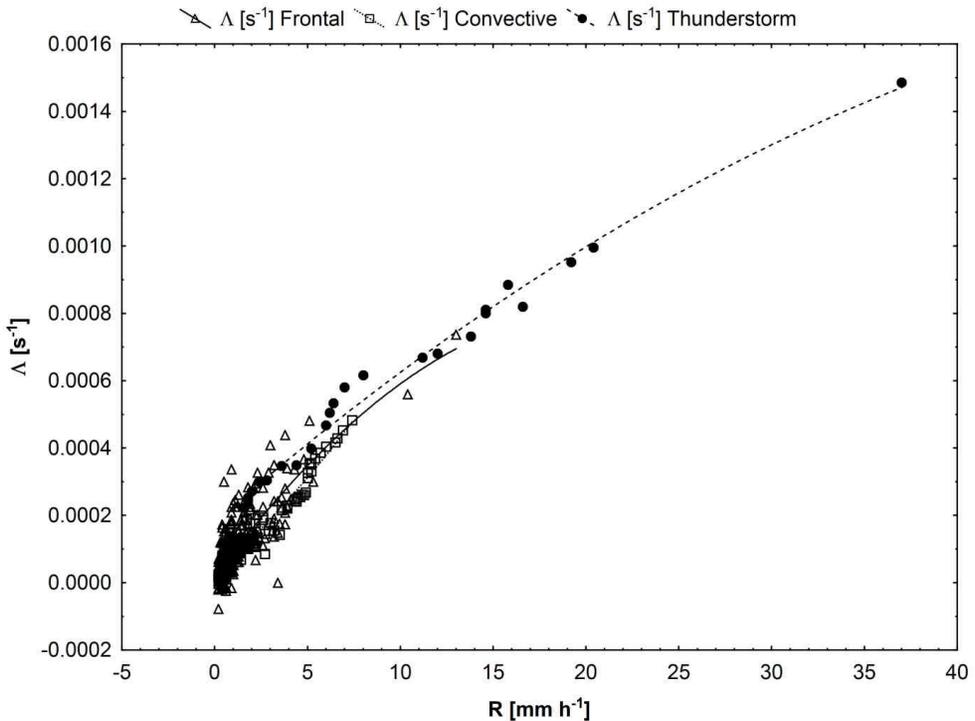


Fig. 2. Collective results of $A = f(R)$ for various precipitation type and for all ranges of rainfall intensity

By comparing convective precipitation with frontal rainfall, one can see the considerable scatter of A for the specific ranges of rainfall intensity for the case of large-scale precipitation. For rainfall intensity in the range from 0.2 to 0.5 $mm\ h^{-1}$, considerable divergences were noted in the value of the scavenging coefficient. While the values of A for frontal and convective rains are comparable, the coefficient of variability C_v for large-scale precipitation is almost two times higher. The results of basic statistical treatment of the comparative analysis for rainfall intensities of 0.2, 0.3, 0.4 and 0.5 $mm\ h^{-1}$ indicate that in all cases the value of the variability of coefficient A assumes considerably higher values for large-scale precipitation, equal to 99.9-41.3, 44.7-16.0, 69.0-36.6 and 72.3-35.9%, respectively. Concurrently, for frontal precipitation, the medians of A assume

15-20% higher values than the results obtained during convective rainfall. The wide divergences in the scavenging coefficient for frontal rainfall are difficult to explain. One might assume that the methodology of the conducted research is responsible for such conclusions. Around 75% of the registered variations in PM₁₀ concentrations noted in the intervals of 30 minutes coincide with episodes of continuous precipitation, sometimes exceeding 6 hours in duration (eg 12 independent observations during a single instance of rainfall).

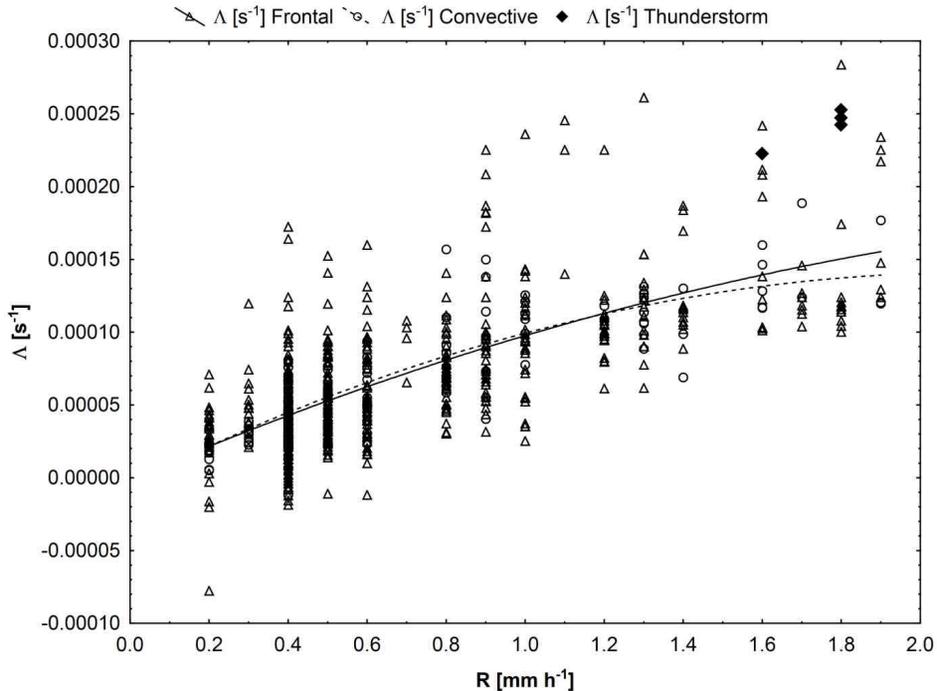


Fig. 3. Graphical interpretation of $A = f(R)$ for three types of precipitation. Graph for R from 0.2 to 2.0 mm h⁻¹ only

The long-term precipitation and lack of other potential sources of PM emission in the immediate surroundings result in the effective scavenging process, which leads to the minimum differences in the registered concentrations of C_0 and C_1 , which has an obvious effect on the decrease in the value of A . However, an analysis of the singular observations of instances of large-scale precipitation ($n = 93$), lasting only 30 minutes, also indicates the considerably higher value of C_V than for convective precipitation. The principal reason for this could be associated with the variable structure of the precipitation [4, 25]. As far as large-scale precipitation is concerned, in the investigated range of the intensity of wet deposition, one could distinguish drizzle (small, densely packed droplets) as well as deposition of denser, yet more lightly falling, raindrops. The results for light rain seem to confirm the statement by Zhang et al [25] that the densely deposited rainfall with the fine droplet structure tends to wash out particulate matter from the atmosphere better due to the

droplets' larger developed surface. Another factor could be associated with the effect of processes of atmospheric dispersion and the transport of PM a result of the movement of air masses. The smaller values of λ for PM₁₀ were noted as accompanying the increase of wind speed, in particular from the west and north-west (*ie* from areas with the intensive effect of anthropogenic emission sources).

Analysis of the scavenging coefficient performed with the use of the Mann-Whitney test for specific rainfall intensities changing every 0.1 mm h⁻¹ indicates that for convective precipitation it is necessary to reject the statement that there are no differences between the values of λ (p -value < 0.001). For the case of frontal precipitation, the hypothesis regarding the equality of the λ median is valid only for the ranges of R : 0.3 and 0.5, 0.4 and 0.5 as well as 0.5 and 0.6 mm h⁻¹ (p -value 0.59, 0.42 and 0.30, respectively). The same test performed for the ranges corresponding to the standard classification of rainfall intensity (light, intermediate, heavy) indicates that for the investigated types of precipitation we can find relevant differences in the λ value when they are considered individually.

The effect of this 7-year campaign took the form of mean, minimum and maximum values of the scavenging coefficient for each type and range of rainfall intensity. Table 3 contains the collective results of the study.

Table 3
Scavenging coefficient λ [s⁻¹] for three types of tested precipitation and four ranges of rainfall intensity

Precipitation type	Convective	Frontal	Thunderstorm
$R \leq 0.5 \text{ mm h}^{-1}$			
avg	$4.65 \cdot 10^{-5}$	$7.32 \cdot 10^{-5}$	no data
min	$-1.19 \cdot 10^{-5}$	$-7.76 \cdot 10^{-6}$	no data
max	$8.22 \cdot 10^{-5}$	$1.72 \cdot 10^{-4}$	no data
sd	$2.04 \cdot 10^{-5}$	$2.79 \cdot 10^{-5}$	no data
$R = 0.6-2.0 \text{ mm h}^{-1}$			
avg	$8.70 \cdot 10^{-5}$	$8.69 \cdot 10^{-5}$	$1.47 \cdot 10^{-4}$
min	$2.33 \cdot 10^{-5}$	$9.97 \cdot 10^{-6}$	$2.22 \cdot 10^{-4}$
max	$1.89 \cdot 10^{-4}$	$3.37 \cdot 10^{-4}$	$2.72 \cdot 10^{-4}$
sd	$5.70 \cdot 10^{-5}$	$3.20 \cdot 10^{-5}$	$7.70 \cdot 10^{-5}$
$R = 2.1-5.0 \text{ mm h}^{-1}$			
avg	$1.05 \cdot 10^{-4}$	$2.09 \cdot 10^{-4}$	$2.40 \cdot 10^{-4}$
min	$8.56 \cdot 10^{-5}$	$6.70 \cdot 10^{-5}$	$2.99 \cdot 10^{-4}$
max	$3.27 \cdot 10^{-4}$	$4.38 \cdot 10^{-4}$	$3.99 \cdot 10^{-4}$
sd	$6.13 \cdot 10^{-5}$	$5.80 \cdot 10^{-5}$	$6.50 \cdot 10^{-5}$
$R > 5.0 \text{ mm h}^{-1}$			
avg	$1.94 \cdot 10^{-4}$	$6.19 \cdot 10^{-4}$	$7.69 \cdot 10^{-4}$
min	$3.31 \cdot 10^{-4}$	$2.99 \cdot 10^{-4}$	$4.68 \cdot 10^{-4}$
max	$4.83 \cdot 10^{-4}$	$7.37 \cdot 10^{-4}$	$1.49 \cdot 10^{-3}$
sd	$6.78 \cdot 10^{-5}$	$1.82 \cdot 10^{-4}$	$2.55 \cdot 10^{-4}$

The conclusion from the research, confirmed by the results of Spearman's correlation, indicates that the scavenging coefficient λ for large particles is considerably related to the intensity and type of precipitation. Similar results were found in the research reported by Gonzalez and Aristizabal [47]. The relatively high values of Standard Deviation (and C_V), namely for convective rainfall and thunderstorm events shown in Table 3, indicate that aerosol concentration changes accompanied by air mass changes often dominate over the effects of the interaction with hydrometeors. This confirms the important role of the

convective movements of the air, vertical diffusion and turbulence, which are very strong close to the Earth surface.

Similarly effective scavenging was found to be specific to the examined ranges of rainfall intensity: convective precipitation without thunder and stratiform rainfall. Presenting the results in a more friendly form, one can see that for the results taken at intervals of 30 minutes and rainfall $R \leq 0.5 \text{ mm h}^{-1}$, the total concentration of PM₁₀ in the ground-level zone decreases by 8%. These values were two times higher for rainfall intensity in the range $0.6\text{-}2.0 \text{ mm h}^{-1}$. Heavy rains lasting 30 minutes result in the decrease of aerosol levels by 51 and 59% for convective and large-scale precipitation respectively. For the case of thunderstorms, the decrease of PM₁₀ is considerably greater. For rainfall intensity from $0.6\text{-}2.0 \text{ mm h}^{-1}$ it is equal to 17%, while in the range $2.1\text{-}5.0 \text{ mm h}^{-1}$, the results are as much as 46%, and over 73% for heavy rainfall.

Table 4 presents the results of the Mann-Whitney test and illustrates the results of the calculated values of scavenging coefficient depending on the rainfall intensity and type (at the level of test relevance $\alpha = 0.05$). The results strongly confirm the observations made in this study.

Table 4

The results of Mann-Whitney test of A comparison. p -value for three types of precipitation

Rain rate [mm h^{-1}]	≤ 0.5		<i>0.6-2.0</i>
	Convective	Frontal	Storms
Convective		0.25	<i>0.0002</i>
Large scale	0.73		<i>0.0002</i>
Storms	no data to compare	no data to compare	
Rain rate [mm h^{-1}]	$2.1\text{-}5.0$		<i>> 5.0</i>
	Convective	Frontal	Storms
Convective		0.21	<i>< 0.0001</i>
Frontal	0.77		<i>0.03</i>
Storms	0.003	0.01	
Bold values showed realization of Mann-Whitney test. Italics fonts showed test results for $0.6\text{-}2.0$ and $> 5.0 \text{ mm h}^{-1}$			

Linear regression model

The high values of Spearman's correlation coefficient (A , R) were found for all types of precipitation. However, analysis aimed at the development of regression model was conducted only for large-scale rainfall. From considered, it is unique type of precipitation that meets the criterion occurrence of spatially homogeneous system in which external influences are small in comparison to the interaction between aerosol and hydrometeor.

Classical analysis was undertaken, under the assumption of the most important foundations, including:

- adoption of stability of the function between the examined phenomena in the model,
- linear characteristics of the model in respect to parameters of expression:

$$Y = \beta_1 \cdot X + \beta_0 + \gamma \quad (3)$$

where β_1 and β_0 are the structural parameters of the model,

- residual value which is the random variable with normal distribution $N(0, \sigma^2)$.

For the case of large-scale precipitation, despite the stability of the function between the examined phenomena, it was impossible to identify a regression model for the entire

range of the measurement data. The analysis of residuals indicated that the condition of the normality distribution was not fulfilled. Nevertheless, for windless weather and intensity of the frontal precipitation in the range $0.2\text{-}2.0\text{ mm h}^{-1}$, all of these conditions were fulfilled. The results of the analysis of 45 instances of rainfall corresponding to the above criteria lead to the conclusion that the regression model provides an explanation of 98% of the variations in the variable Λ . The mean difference between the actual values of the dependent variable and the values predicted by the model was equal to $8 \cdot 10^{-6}\text{ s}^{-1}$ (which is equal to 8.8% of the mean for the dependent variable (with a mean of $9.1 \cdot 10^{-5}$)). The high value of the statistical F (above $2.5 \cdot 10^3$) and the level of probability p ($p < 10^{-4}$) corresponding to it confirm the statistical relevance of the linear model. The value of statistical t (51) used to assess the relevance of the β_1 coefficient and the level of probability $p < 10^{-6}$ corresponding to it confirm that this parameter is considerably different from zero. Increase in the intensity of large-scale precipitation by 1 mm h^{-1} results in an increase of the scavenging coefficient by $1.17 \cdot 10^{-4}\text{ s}^{-1}$. One can see at this point that the model preserves its relevance only on condition that the large-sale precipitation maintains its intensity range and the weather is windless.

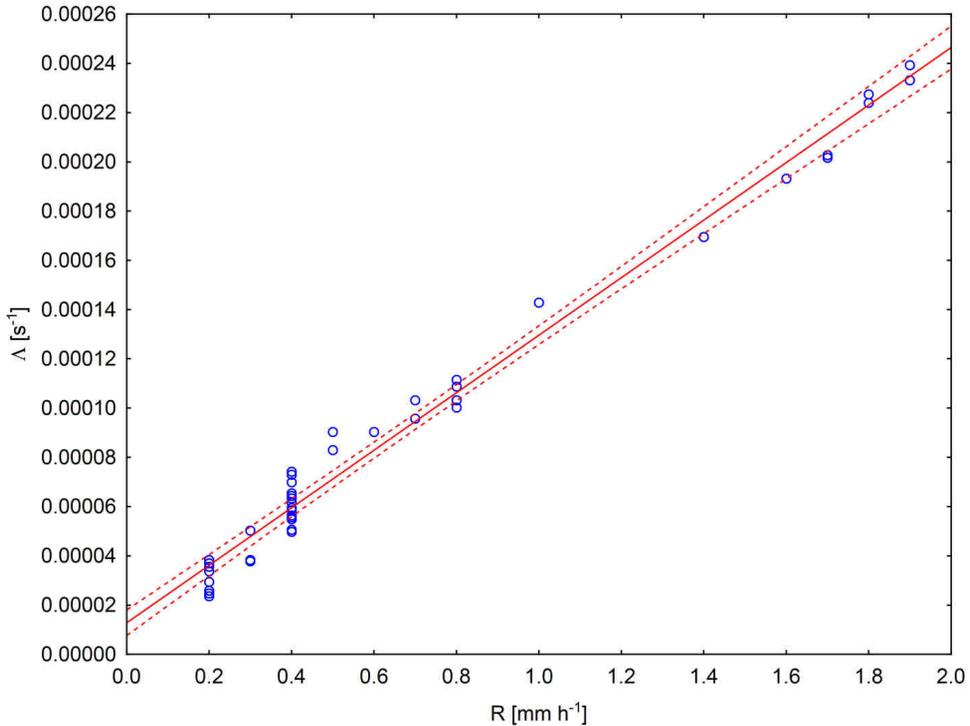


Fig. 4. Large-scale precipitation. Relation $\Lambda = f(R)$. Only for $R: 0.2\text{-}2.0\text{ mm h}^{-1}$ and no-wind conditions

Figure 4 contains the chart for stratiform precipitation which illustrates the dependence between Λ and R derived by means of the experiment and described by the linear model ($r = 0.9919$). Additionally, the boundary of the area with 95% confidence in relation to the

regression line was marked. The chart in Figure 4 confirms the adequacy of the linear model for describing the relations between A and R . The graphical illustration confirms the data from the actual experiment. The applicability of this model is further proved by the test of residuals normality distribution. For the adopted level of relevance $\alpha = 0.05$, the p -value for the Kolmogorov-Smirnov test with Lilliefors correction was equal to > 0.2 (p value for Shapiro-Wilk test 0.18). The observed linear relation between the theoretical and experimental quantiles confirms the normal distribution of the random component.

The results of regression analysis for large-scale precipitation:

$$A = \beta_1 R + \beta_0$$

where: $\beta_1 = 1.2 \cdot 10^{-4}$, $SE_{\beta_1} = 0.2 \cdot 10^{-5}$, $\beta_0 = 1.3 \cdot 10^{-5}$, $SE_{\beta_0} = 0.2 \cdot 10^{-5}$, $r^2 = 0.98$, estimation error: $1 \cdot 10^{-5}$, $t = 51$ for $p < 1 \cdot 10^{-5}$.

Considering the large number of tests and the results of statistical analysis, one may assume that, in respect to the scavenging of PM₁₀, the results in the form of equation of simple regression can offer an explanation for over 90% of cases of the variability in the value of A . Such a high proportion leads to the conclusion that such relations are not coincidental and remain relevant also in the general population.

Conclusions

Knowledge of the concentration of PM₁₀ offers a foundation for the air quality assessment performed by institutions which realize continuous monitoring of atmosphere. The presented research and analysis of the results have not only practical application. The results of PM₁₀ concentration measurements taken continuously and at constant time resolutions and additionally supplemented with the results of continuous registration of the characteristics of wet deposition, offer additional insight into the processes of particulate matter washout from the atmosphere. The fulfillment of the condition of constant time intervals of the measurements enables adequate accounting for dynamic processes of particulate matter concentration, relations between PM₁₀ concentration and type, and rainfall intensity and processes of below-cloud scavenging.

As the conclusion from this research, the following statements can be made:

- data on A are only partially related to classical rain scavenging. In the case of convective precipitation (mainly during thunderstorms episodes), the aerosol concentration changes due to external factors (horizontal mass changes, convection, vertical diffusion, turbulence) can dominate the effects caused by the interaction with hydrometeors,
- the structure of the precipitation plays an important role in the effectiveness of scavenging,
- within the range of comparable values of rainfall intensity, the type of wet deposition (except for storms) does not influence the effectiveness of scavenging PM₁₀ from the ground-level zone,
- the results of experimental observations, which can be more useful in describing the variability of particulate matter pollution in the atmosphere than laboratory measurements, can be applied in models of dispersion and deposition of pollutants,
- the results gained from this analysis seem to be representative of areas with similar climate characteristics.

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ZMIANY STĘŻENIA PM_{10} W WARUNKACH WYSTĘPOWANIA OPADÓW CIEKŁYCH

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Abstrakt: W pracy przedstawiono wyniki połowych badań nad zmiennością współczynnika wymywania A pyłu zawieszonego o aerodynamicznej średnicy cząstek poniżej $10 \mu\text{m}$. Siedmioletnie rejestracje zmian stężenia PM_{10} w warunkach występowania trzech typów opadów ciekłych (konwekcyjnych, wielkoskalowych i burz) przeprowadzono na obszarze niezurbanizowanym. Analizie poddano 806 przypadków obserwacji o stałej rozdzielczości czasowej 0,5 h. Pomiary stężenia PM_{10} prowadzono z użyciem metody referencyjnej przy jednoczesnej rejestracji podstawowych parametrów meteorologicznych. Wykazano, że dla PM_{10} efektywność wymywania jest silnie zależna od intensywności opadu R oraz od typu opadu. W przypadku opadów konwekcyjnych dane dotyczące wartości A są tylko częściowo związane z „klasycznym podejściem” wymywania cząstek przez deszcz. W zakresie porównywalnych wartości intensywności opadu typ mokrej depozycji (z wyjątkiem burz) nie wpływa na efektywność oczyszczania troposfery przyziemnej z PM_{10} . Znaczna ilość prowadzonych w warunkach rzeczywistych obserwacji opadów wielkoskalowych pozwoliła na zaproponowanie prostego modelu regresji, który z wysokim prawdopodobieństwem, ale w ograniczonym zakresie, może zostać uznany za odpowiedni do opisu zmienności A (D_{PM10}). Uzyskane wyniki badań mogą znaleźć zastosowanie w modelach dyspersji i depozycji zanieczyszczeń i są reprezentatywne dla obszarów o podobnej charakterystyce klimatu.

Słowa kluczowe: aerozol, mokra depozycja, wymywanie podchmurowe, obszar niezurbanizowany