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PM_{10} concentration reduction due to the wet scavenging in the Ciuc Basin, Romania

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Abstract. The PM_{10} concentration reduction caused by large-scale precipitation in the Ciuc Basin was studied under no-wind conditions. The PM_{10} concentration changing before, during, and after the rainfall was followed up from 2008 to 2019. After the rainfall episode, the PM_{10} concentrations were lower in the cold and warm periods with 2.8 $\mu g/m^{3,}$ and 2 $\mu g/m^3$ respectively. The highest PM_{10} concentration reduction was detected in the cold season, by the moderate and light rain intensity, after 6 hrs of continuous rain (35.61%, 32.46%), and the average PM_{10} concentration reduction in the cold and warm periods was 22.3% and 16.1% respectively.

Keywords: particulate matter, atmospheric purification, rainfall, meteorological condition

1. Introduction

Particulate matter (PM_{10}) with aero-diameter less than 10 µm harms human health, causing various respiratory and cardiovascular diseases and premature death around the world [1]–[3]. The precipitation washout mechanisms perform the primary atmospheric purification. The unfavourable meteorological parameters, such as inversion and fog, have a negative influence on air pollution concentration [4], [5]. On the other hand, the favourable meteorological conditions via dilution and elimination have a reduction effect [6]. The atmospheric purification may happen through dry and wet scavenging [7]. Wet scavenging can take place in two different ways: in the cloud and below cloud [8]–[10]. The effectiveness of washout is strongly related to the seasonal PM_{10} variation between the seasons: relatively high PM_{10} concentration in the cold period and a significantly lower level in the warm period [11]–[13].

In the background of the PM_{10}, washout effects are responsible for several mechanisms such as Brownian diffusion, thermophoresis, diffusion, inertia, and electric washing [4], [8], [10], [14]. The lifting condensation level variation also plays an essential role in particulate matter variation. Many studies show the effect of precipitation washout by comparing the PM_{10} concentration with the precipitation and non-precipitation periods [9], [15], [16]. The air pollutant (PM_{10}) reduction efficiency by precipitation scavenging depends on the precipitation quantity and duration [8].

This research paper presents the PM_{10} concentration reduction efficiency in the Ciuc Basin caused by rainfall washout under windless conditions, taking into account rainfall intensity, duration, and meteorological parameters.

2. Materials and methods

2.1. Meteorological data and PM_{10} sampling procedure

The study was conducted in the Ciuc Basin area and covered 11 years between 2008 and 2019. The sampling site is an inter-Carpathian, closed-type depression, located in the central part of the Eastern Carpathians, where the fog and inversion phenomena are very frequently present mainly in the cold period [17]–[19]. The hourly PM_{10} and meteorological dates, such as air temperature, relative humidity, precipitation quantity, and wind speed, were provided by the National Environmental Protection Agency Harghita. The monitoring station is situated in Jigodin, with the following coordinates: latitude: 46.33 °N, longitude: 25.81 °E, and altitude: 697 m.

In order to determine the purification effect of precipitation, the hourly PM_{10} concentration was followed up and compared in the case of rainfall and no precipitation period. For an in-depth assessment to determine the atmospheric purification by wet scavenging, different conditions have been set: 1. no-wind condition (< 1 m/s), 2. three rain intensity levels (light: 0.2–0.4 mm h^{-1}, moderate: 0.4–3.9 mm h^{-1}, and heavy rains: > 3.9 mm h^{-1}), and 3. rain duration from 1 to 6 hours were analysed. The analysis was carried out separately for the cold (Oct–Mar) and warm period (Apr–Sep).

2.2 Procedure for determining the removal efficiency coefficient ΔC

The effectiveness of PM_{10} scavenging by precipitation was calculated based on *Equation 1*, where the percentage change (ΔC) was obtained from the concentration variation before (C_0) and after (C_t) episodes of rain.

$$\Delta C = \frac{C_t - C_0}{C_0} * 100 \quad (1)$$

Based on air temperature and relative humidity, the lifting condensation level (LCL) was calculated. Regarding the lifting condensation level (LCL), it was calculated according to *Equation 2*:

$$LCL = 20 + \frac{T}{5}(100 - RH), \quad (2)$$

where: *LCL* – lifting condensation level, *T* – air temperature (ºC), and *RH* – relative humidity (%).

3. Results and discussions

3.1. Statistical description and Spearman's correlation

Summary statistics data are presented in *Table 1* including data regarding the selected air pollutant (PM_{10}), meteorological parameters, and the precipitation, examined during the 11-year experiment. Rainfall intensity (light, moderate, heavy) and duration (1 to 6 hours) have an essential role in the PM_{10} concentration reduction from the troposphere. The analysis of these parameters showed that the main form of wet deposition was carried out by rainfall with low intensity (0.2–0.4 mm h^{-1}) in cold and warm periods (68.82%, 55.92%). The lowest occurrence was observed for rainfall with high intensity (0.4%, 3.4%). During the cold period of observation, the average PM_{10} concentration was 1.32 times higher than the annually acceptable limit (20 µg/m^3). Due to the different emission sources and the unfavourable meteorological conditions, a significant difference was found between the cold and warm period PM_{10} concentration, which was 26.42 µg/m^3 and 10.97 µg/m^3 respectively. The LCL has an important effect on the PM_{10} concentration evolution – almost two-fold differences were found between the average cold and the warm season LCL highs (344 m, 626 m).

Table 1. PM_{10} and meteorological parameters characterization

Precipitation period	Number of rain episode		PM_{10}	T (°C)	RH (%)	LCL (m)	Ws (m s^{-1})
Cold season (Oct–Mar)		**Avg.**	26.42	0.47	83.13	344	0.71
	L (6176)	**Med**	16.23	0.56	86	269	0.3
	M (2762)	**Min**	0.10	-27.6	15	0	0
	H (36)	**Max**	251.82	28.37	100	1,900	50
Warm season (Apr–Sep)		**Avg.**	10.97	15.28	73.71	626	1.28
	L (4407)	**Med**	9	15.1	77	524	0.5
	M 3199)	**Min**	0.1	5.01	12	0	0
	H (274)	**Max**	102.1	35.75	100	2,273	50

Note: T – temperature, RH – relative humidity, LCL – Lifting Condensation Level, Ws – wind speed, Avg. – average, Med. – median, Min – minimum, Max – maximum, L – light 0.2–0.4 mm h^{-1}, M – moderate 0.4–3.9 mm h^{-1}, H – heavy rains > 3.9 mm h^{-1}

During large-scale rain events in the cold and warm seasons, the average air temperature was around 0.47 °C and 15.25 °C respectively. The cold season is characterized by higher relative humidity and lower wind speed than the warm season with 83.13%, 73.71% and 0.71 m s^{-1}, 1.28 m s^{-1} respectively.

3.2. PM_{10} concentration with and without precipitation

Concentrations of PM_{10} were lower in the case of rainfalls than in the non-precipitation period, and a noticeable difference in average PM_{10} concentrations was observed in the cold and warm period: 2.8 μg/m^3 and 2 μg/m^3 respectively (*Figure 1*).

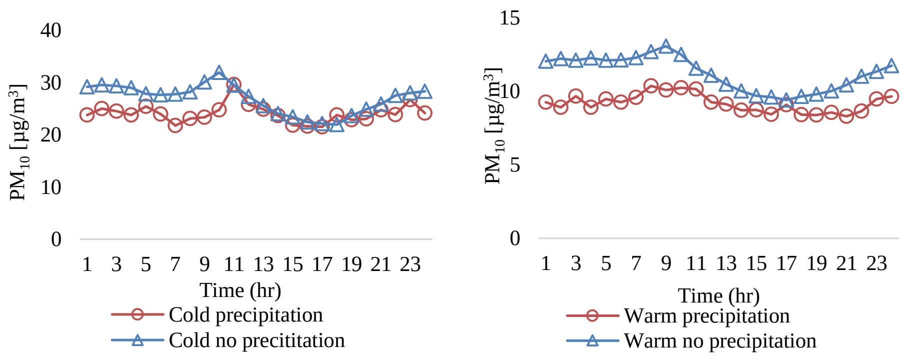
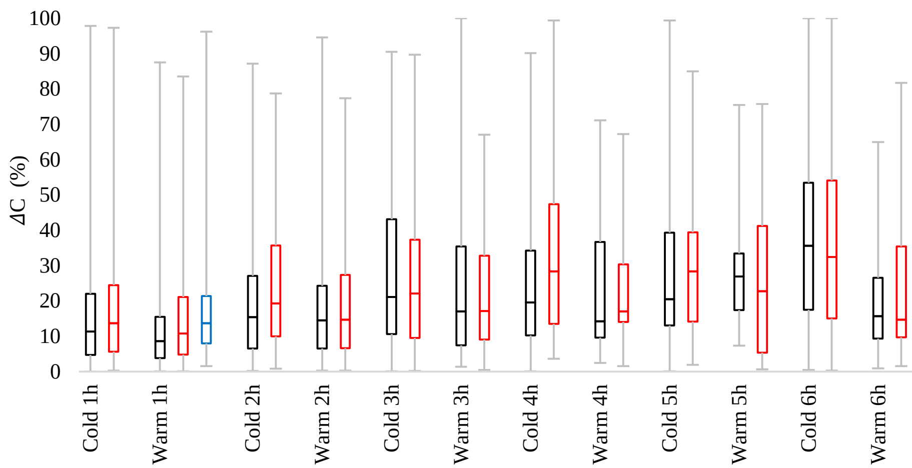


Figure 1. PM_{10} hourly variations under conditions of precipitation and non-precipitation: in the cold and in the warm period

The pattern of hourly PM_{10} concentration variation under conditions of precipitation and non-precipitation was quite similar to each other. Quantitatively, the reduction effect of precipitation scavenging in the cold period was higher than in the warm period. Still, the percentage reduction was 11.69% and 22.06%, resp., thanks to the unequal PM_{10} concentration during the cold and warm season. The hourly PM_{10} concentration in the warm period increased due to a relatively more substantial direct effect of vehicle emissions despite the rainfall.

3.3 Effectiveness of PM_{10} scavenging

Precipitation cases with a duration of one hour in the cold and warm season had the highest frequency with 48.56 % and 76.11%, resp., and a duration of 2 hours was next with 11.51% and 9.72% resp. The PM_{10} concentration reduction due to rain is presented in *Figure 2*. The graphical results indicate that the removal efficiency is growing with the rainfall intensity and duration in the cold period. This increasing trend in the warm period was valid for a rain duration of 1 to 3 hours.



Note: the box plot colour symbolizes rain intensity: black – light (L), 0.2–0.4 mmh^{-1}; red – moderate (M), 0.5–3.9 mmh^{-1}; blue – heavy (H), > 4 mmh^{-1}.

Figure 2. Effectiveness of PM_{10} scavenging in the function of the duration and intensity of large-scale precipitation

The constant values of the ratios ΔC6h to ΔC1h (1.6 and 1.4) and ΔC3h to ΔC1h (1.3 and 1.2) are observed for the cold and warm seasons respectively. The average PM_{10} concentration reduction in the cold and warm periods was

22.3% and 16.1% respectively. In all studied cases, the highest PM_{10} concentration reduction was detected in the cold season in the case of low and moderate rain intensity, after 6 hrs of continuous rain (35.61%, 32.46%). Following the observation, it became evident that the PM_{10} concentration reduction from the atmosphere in the case of the light (< 1 mm) rainfall was smaller thanks to the fact that the impact of pollution exceeded the washout effect. In the cold season, the PM_{10} concentration reduction by wet scavenging from the air was 11.58% higher for the case of heavy rains than for the light ones. In the warm period, this reduction was lower, with 9.09%.

Using the Pearson correlation, the statistical data confirms the significant relationship between PM_{10} concentration reduction and rainfall duration, with a considerable degree of relation ($r = 0.93$). The correlation coefficient was higher than the significance level ($P < 0.05$, $r = 0.7$) in the case of moderate and low rainfall: 0.98 and 0.88 respectively. For the Pearson correlation coefficient calculation in the warm period, the rainfall duration between 1 to 3 hours was taken into consideration only when the continuous removal PM_{10} effect was detected. The Pearson correlation coefficient was higher than the significance level ($P < 0.05$, $r = 0.87$), with 0.97 and 0.99 in the case of low and moderate precipitation resp.

4. Conclusions

During the cold period of observation (2008–2019), the average PM_{10} concentration in the Ciuc Basin was 1.32 times higher than the annually acceptable limit (20 $\mu g/m^3$). The dominant rainfall was the precipitation with low intensity, and the most frequent duration for rainfalls was 1 hour. Due to the different emission sources and the meteorological conditions, a significant difference was found between the PM_{10} concentration measured in the cold and warm periods. The LCL had an essential effect on the PM_{10} concentration evolution; almost two-fold differences were found between the average cold and the warm season. Quantitatively, the reduction effect of precipitation scavenging in the cold period was higher than in the warm period.

In all cases studied, the highest PM_{10} concentration reduction was detected in the cold season in the case of the low and moderate rain intensity, after 6 hrs of continues rain. In the cold season, the efficiency of PM_{10} scavenging by rainfall shows a continuously increasing trend based on the rain duration from 1 to 6 hours. This increasing trend in the warm period was true for rain durations of 1 to 3 hours. The Pearson correlation based on the statistical data confirms the significant relationship between PM_{10} concentration reduction and rainfall duration, with a considerable degree of relation.

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References

[1] Adar, S. D., Filigrana, P. A., Clements, N., Peel, J. L. (2014), Ambient coarse particulate matter and human health: A systematic review and meta-analysis. *Curr. Environ. Heal. Reports* 1, 258–274.

[2] Azam, A. G., Zanjani, B. R., Mood, M. B. (2016), Effects of air pollution on human health and practical measures for prevention in Iran. *J. Res. Med. Sci*. 21(65), 1–12.

[3] Blanco-Becerra, L. C., Gáfaro-Rojas, A. I., Rojas-Roa, N. Y. (2015), Influence of precipitation scavenging on the PM2.5/PM10 ratio at the Kennedy locality of Bogotá, Colombia. *Rev. Fac. Ing*. 76, 58–65.

[4] Chate, D. M., Rao, P. S. P., Naik, M. S., Momin, G. A., Safai, P. D., Ali, K. (2003), Scavenging of aerosols and their chemical species by rain. *Atmos. Environ*. 37, 2477–2484.

[5] Feng, J. (2009), A size-resolved model for below-cloud scavenging of aerosols by snowfall. *J. Geophys. Res*. 114, 1–8.

[6] Feng, X., Wang, S. (2012), Influence of different weather events on concentrations of particulate matter with different sizes in Lanzhou, China. *J. Environ. Sci*. 24, 665–674.

[7] Keresztesi, Á., Birsan, M.-V., Nita, I.-A., Bodor, Z., Szép, R. (2019), Assessing the neutralisation, wet deposition and source contributions of the precipitation chemistry over Europe during 2000–2017. *Environ. Sci. Eur*. 31(50), 1–15.

[8] Kim, S., Hong, K., Jun, H., Park, Y., Park, M. (2014), Effect of precipitation on air pollutant concentration in Seoul, Korea. *Asian J. Atmos*. 8, 202–211.

[9] Lim, D., Lee, T., Kim, D. (2012), Quantitative estimation of precipitation scavenging and wind dispersion contributions for PM 10 and NO 2 using long-term air and weather monitoring database during 2000–2009 in Korea. *J. Korean Soc. Atmos*. 28(3), 325–347.

[10] Liu, H., Tian, Y., Xiang, X., Sun, K., Juan, J., Song, J., Cao, Y., Xu, B., Hu, Y. (2017), Air pollution and hospitalization for acute myocardial infarction in China. *Am. J. Cardiol*. 120, 753–758.

[11] Nuhog, A., Bayraktar, H., Turalıog, F. S. (2005), Impacts of some meteorological parameters on SO2 and TSP concentrations in Erzurum, Turkey. *Chemosphere* 59, 1633–1642.

[12] Olszowski, T. (2016), Changes in PM10 concentration due to large-scale rainfall. *Arab. J. Geosci*. 9(160), 1–11.

[13] Santachiara, G., Prodi, F., Belosi, F. (2013), Atmospheric aerosol scavenging processes and the role of thermo- and diffusio-phoretic forces. *Atmos. Res*. 128, 46–56.

[14] Stephan, H., Andre S. H. P. (2011), Influence of meteorology on PM10 trends and variability in Switzerland from 1991 to 2008. *Atmos. Chem*. Phys. 11, 1813–1835.

[15] Szép, R., Keresztes, R., Korodi, A., Tonk, S. (2017), Study of air pollution and atmospheric stability in Ciuc Basin, Romania. *Rev. Chim*. 68, 1763–1767.

[16] Szép, R., Mateescu, E., Niță, I. A., Birsan, M. V., Bodor, Z., Keresztesi, Á. (2018), Effects of the Eastern Carpathians on atmospheric circulations and precipitation chemistry from 2006 to 2016 at four monitoring stations (Eastern Carpathians, Romania). *Atmos. Res*. 214, 311–328.

[17] Tiwari, S., Chate, D. M., Pragya, P., Ali, K., Bisht, D. S. (2012), Variations in mass of the PM10, PM2.5, and PM1 during the monsoon and the winter at New Delhi. *Aerosol Air Qual. Res*. 12, 20–29.

[18] Wu, Y., Liu, J., Zhai, J., Cong, L., Wang, Y., Ma, W., Zhang, Z., Li, C. (2018), Comparison of dry and wet deposition of particulate matter in near-surface waters during summer. *PLoS One* 13, 1–15.

[19] Yoo, J., Lee, Y., Kim, D., Jeong, M., Stockwell, W. R., Kundu, P. K., Oh, S., Shin, D., Lee, S. (2014), New indices for wet scavenging of air pollutants (O3, CO, NO2, SO2, and PM10) by summertime rain. *Atmos. Environ*. 82, 226–237.