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CHANGES IN WATER CHEMISTRY AS A RESULT OF RAINFALL RUNOFF FROM THE ROOFS OF VARIOUS COATINGS. PART I

KSZTAŁTOWANIE SIĘ CHEMIZMU WÓD OPADOWYCH W WYNIKU SPŁYWU Z DACHÓW O RÓŻNYCH POKRYCIACH. CZĘŚĆ I

Abstract: The aim of the study was to determine the level of calcium, magnesium, sodium, phosphorus and potassium in the waters flowing from the roofs of houses with varying degrees of coverage on the background of their contents in rain waters. On the basis of the Minister of Environment Decree of 24 July 2006 on conditions to be met for the introduction of sewage into the water or ground and on substances particularly harmful to the aquatic environment, runoff from paved areas shall be treated as sewage, and runoff from roofs of buildings are treated as pure water and can be discharged into the environment without a permit. However, literature data indicate the possibility of a significant enrichment of rainwater at the time of contact with the roof covering. The study included 24 roofs of houses or small trade buildings. As background to the research used rainwater collected in two randomly selected locations within the area of research. The study was conducted in areas with low human impact, in order to best capture the effect of the type of roofing material on the formation of water chemistry. Research area was located in the Luslawice in the Tarnow county in Malopolska province. The study included the most common roofs in the surveyed area: cement tile, ceramic tile, bituminous, unpainted galvanized metal, copper and asbestos cement. The tested water samples to determine the content of calcium, phosphorus, magnesium, sodium and potassium. In addition, it was determined the pH value of water and electrolytic conductivity. The results of this study indicate that the water runs off the roofs of respondents in each case contained a greater quantity of the analyzed elements in comparison with rainwater. For example, while the average content of magnesium in the water flowing from the tile cement was almost ten times higher than in rain water, in the case of tile ceramic was almost five times more water from the bituminous coverings contained about three times more magnesium, and water from the galvanized metal contained about six times more as compared with rainwater. Also in the case of most other elements enrichment factors found in the waters cover the cement (tile, and asbestos cement) then galvanized and coated while the lowest were recorded in the enrichment of waters from the roofs of the covering of ceramic tiles. Also noted an increase in the conductivity values in waters from the roofs of the coverings of cement, galvanized steel and ceramic tiles. All runoff from the roofs were of generally higher pH value compared with rainwater, the biggest reaction - about 8 found in the water with cement tiles, slightly lower, about 7.0 in water from roofs with ceramic tiles and sheet copper. pH of the water in the coated sheet was lower than in water, rainwater and oscillating within 5.5. Rainwater pH was 5.94.

Keywords: rainwater tanks, water pollution, runoff from roofs, sodium, potassium, calcium, magnesium, phosphorus

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Quality of rainwater is greatly important for developing chemistry of surface and underground water. In the areas of low anthropopressure, pollutants in the atmospheric air, originating from long atmospheric transport, may be a main factor worsening quality of water environment [1]. The main sources of rainwater pollution are gases and dusts coming from industries and fuel burning. Development of urbanization and increasing coefficient of area anthropisation leads to increasing impermeable surface, which reveal a very low water retention capacity. Water management in these areas causes problems because of fast and intensive flow of storm waters and water deficiency during rainless periods. In urbanized areas torrential rains occur much more frequently, which still aggravates the problem. Draining of water collected from hardened surface immediately to the receivers seem unjustified from the perspective of rational management of water resources. According to the Decree of the Minister of the Natural Environment of 24 July, 2006 on conditions which must be fulfilled at sewage entry to waters and on substances particularly harmful for water environment [2], water from roofs is treated as clean water and no water-legal permit is required for its entry into the environment. However, many authors have pointed out a possibility of excessive pollution of this water, even reaching the level exceeding the norms stated in legal regulations of many countries [3-6]. A method for alleviating the results of diminishing retentive capacities of built up areas is using installations collecting water flowing from the roofs and using it for sanitary purposes or irrigation during drought. It allows for rationalization of water management and considerably diminishes surface runoff in effect leading to increased water retention. Systems of collecting and storage of water flowing from roofs are often introduced in developed countries all over the world. Countries such as Germany, Denmark, India, Japan or Australia are leaders in constructing small installations for rainwater collection from roofs in individual households [7, 8]. National legislative bodies in many of these countries make laws requiring that all new buildings should have rainwater collecting systems. These regulations aim at limiting the requirements for treated water and collection and storage of rainwater instead of discharging it straight to storm drainage system [9, 10]. Also in Poland a possibility of utilization of water collected from roofings has been increasingly often taken into consideration. However, inadequate quality of rainwater may disqualify it from further use [7, 10]. Simons et al [9] point to a possible hazard of bacteriological contamination of water flowing from roofs, which may lead to diseases and infection of people using the water. Therefore, it is important to determine the quality of water, which is collected from a determined type of roof coverings. Numerous authors point out to considerable changes of water chemistry during its contact with roofing and rain pipes [1, 7, 9, 10].

The aim of the investigations was determining the influence of various roofing kinds on chemistry of rainwater in the areas of low level of anthropopressure. The other goal of the research was an assessment of the quality of water flowing from roofs from the viewpoint of its possible use.

Materials and methods

Water for the analyses was collected in September and October 2009 from roofs of detached houses or farm buildings located in Luslawice village in the Tarnow county in the Malopolska province. Water was collected on the second and third day after heavy rains. Rainfall total during investigated periods exceeded 30 mm. Water was collected from the southern, eastern or northern side in order to limit the effect of organisms growing on the northern side of the roofs. The investigated area is situated in the place with small antropopressure, so that the effect of roofing on water chemistry change could best registered. The investigations focused on four roofs covered with cement roof-tiles, 3 roofs covered with ceramic roof-tiles, 3 roofs with bitumen roofing, 3 roofs covered with galvanized sheets, 3 with coated sheet, 3 roofs with copperplate and 3 roofs covered with corrugated eternit. Selected roofs were constructed at an angle of about 45^o and covered detached houses or farm buildings, except one copperplate roof covering a church in Zakliczyn nad Dunajcem village.

The possibly little used roofs were selected, with no visible traces of corrosion. The exception were roofs covered with cement roof-tiles and eternit, which were built in the seventies of the 20^{th} century. The background was rainwater collected directly to polyethylene containers in two randomly chosen places. Water was collected during a rainy period, after long lasting rains in order to eliminate the influence of dusts settled on roofs on water chemistry. Collected water was filtered, stabilized using nitric(V) acid and the contents of analysed metals were assessed by means of atomic emission spectrometry in inductively coupled argon plasma on Jobin Yvon 284 apparatus. The quality of analysis was verified using internal reference material. pH value was determined in raw water in the place of water sampling.

Results and discussion

Obtained results of research evidence that rainwater may considerably change its composition during flow from roofs. The kind of roofing and chemical composition of rainwater to the greatest extent affect changes of chemical properties of water flowing from roofs. Despite a very short water contact with roof, there is an intensive leaching of components from the materials forming the roof cover. There is plenty of information in scientific literature about the quality of runoffs from roofs concerning its utilization. Particularly pollution with sulphur and nitrogen compounds which affect rainwater pH and in result the rate at which elements become washed out from roof coverings [11]. In order to recommend a specific form of utilization of water flowing from anthropogenically transformed areas, one should monitor the chemistry of these waters.

Average pH value of rainwater was 5.86 and the reaction is typical for atmospheric precipitations in the areas with low anthropopressure. At such reaction no effect of acidifying air pollutants or alkaline dusts is noticed. A change of reaction of water flowing from the roof is connected with washing out alkaline ions from the roof surface. A statistically significant change in water reaction was registered for water flowing from roofs covered with cement roof-tiles and copperplate. pH value of these waters were respectively 7.26 and 7.19. Reaction of the other samples remained on a lower level and no statistically significant differences in comparison with rainwater were revealed. Insignificant differences in pH were noticed among the water samples from the same roof type (Fig. 1). Farreny et al [12] found similar dependencies of the reaction and the kind of roofing. The authors stated higher pH in the water from roof with concrete roofing in comparison with roofs covered with ceramic roof-tiles or metal sheet. Water flowing from copperplate roof was characterized by higher pH than the rainwater [14]. Goebel et al [13] stated increased pH values in water flowing from cement roofs, copperplate, zinc coated sheet and also from ceramic roofs. The authors did not state any differences in pH value in water collected from various roofs. Reaction of rainwater reported by these authors was on the level of 5.0 and after flowing from the roofs was about 5.7. Water reaction is an important parameter of water quality from the point of view of its utilisation. Increase in water pH may be a positive phenomenon at its use for irrigation. The contents of analyzed macroelements in rainwater was comparable with the data presented by Kruszyk [15], *ie* 0.202 mg Mg · dm^{-3}; 1.757 mg Ca · dm^{-3}; 0.289 mg Na · dm^{-3}; 0.097 mg K · dm^{-3} and 0.0322 mg P · dm^{-3}. Honorio et al [5] reported a chemical composition of rainfall in Brazil containing several times more sodium and potassium but less magnesium and calcium than the analysed precipitations. Arsene et al [16], who analyzed macroelement concentrations in rainwater in northern Romania found over twice bigger concentrations of magnesium, calcium and potassium and over 6 times higher content of sodium than in the analyzed samples. In the rainwater in northern China Lu et al [17] assessed over ten times more calcium, over 4 times more magnesium and over twice more sodium, over 10 times more potassium. Rainwater in Canada by Ontario Lake, in the areas of low anthropopressure contained comparable with the presented samples amounts of the analyzed elements [14]. Rainwater in the investigated area was characterized by a much smaller contents of the analyzed elements than rainwater in the anthropogenically transformed areas and comparable to clean regions of the world.

Table 1

Selected statistical parameters

Type of roof	Parameter [$mg \cdot dm^{-3}$]	min [$mg \cdot dm^{-3}$]	max [$mg \cdot dm^{-3}$]	Relative standard deviation [%]	Type of roof	min	max	Relative standard deviation [%]	Type of roof	min	max	Relative standard deviation [%]
Rain water	Mg	0.133	0.270	39.3	Cement tiles	0.857	1.635	19.0	Ceramic tiles	0.399	0.740	27.4
	Ca	1.620	1.890	8.88		15.50	20.40	10.0		2.390	7.780	42.4
	Na	0.176	0.401	45.1		0.637	1.497	42.4		0.268	0.552	30.4
	P	0.030	0.034	9.32		0.031	0.040	8.55		0.027	0.030	4.06
	K	0.063	0.131	40.5		0.421	1.115	39.1		0.106	0.310	40.9
	pH	5.80	5.94	1.37		6.85	8.19	7.30		5.93	7.52	9.93
Bitumic roofing	Mg	0.211	0.447	31.4	Galvanized steel	0.314	0.780	37.2	Cooper steel	0.300	0.890	57.1
	Ca	1.605	6.720	54.8		1.630	4.720	41.8		1.777	3.177	32.6
	Na	0.371	0.755	38.8		0.341	1.185	55.2		0.386	1.217	59.8
	P	0.013	0.025	33.2		0.113	0.157	16.1		0.063	0.064	0.86
	K	0.354	0.422	9.11		0.135	0.361	39.5		0.626	0.714	9.65
		5.93	6.870	6.59		6.00	7.43	9.52		7.13	7.26	1.04
Coated plate	Mg	0.535	0.877	22.0	Asbestos	0.275	1.905	70.6				
	Ca	3.112	4.742	18.9		10.20	25.70	38.5				
	Na	0.219	0.925	52.2		0.423	1.440	48.8				
	P	0.031	0.046	14.7		0.331	0.495	17.6				
	K	0.170	0.345	31.8		0.320	0.501	19.6				
		5.34	6.25	8.27		5.95	6.61	5.00				

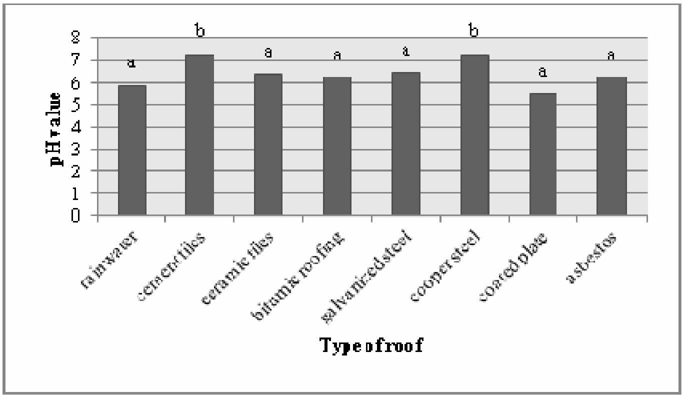

Fig. 1. pH value of collected water

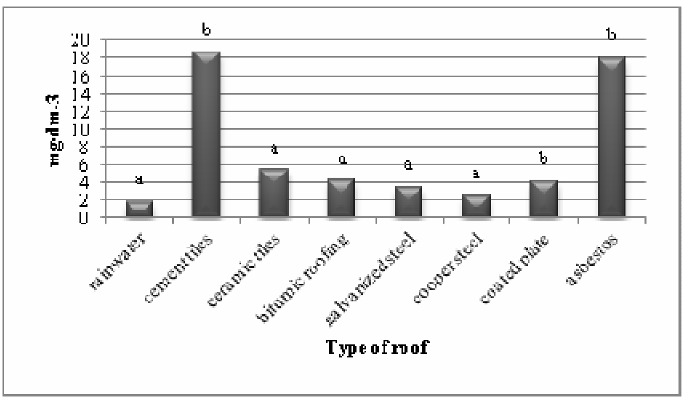

Fig. 2. Calcium content in collected water

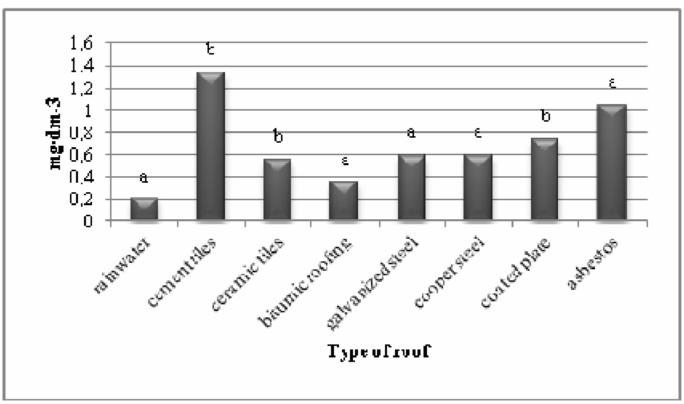

Fig. 3. Magnesium content in collected water

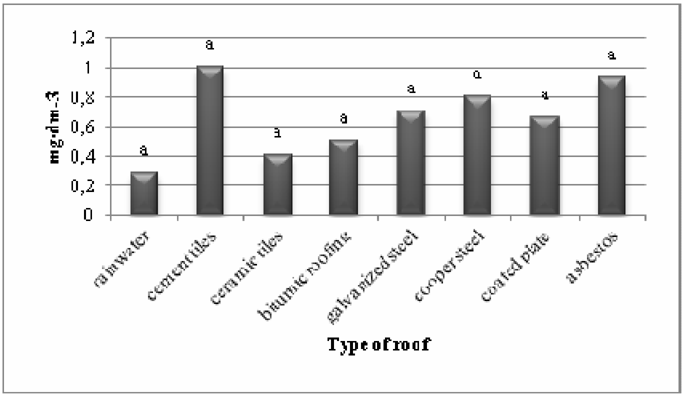

Fig. 4. Sodium content in collected water

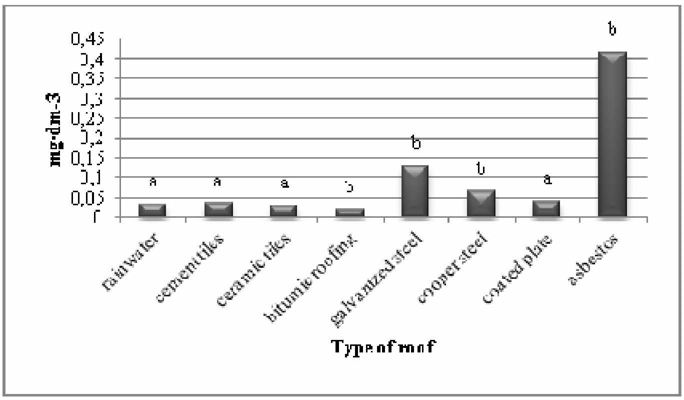

Fig. 5. Phosphorus content in collected water

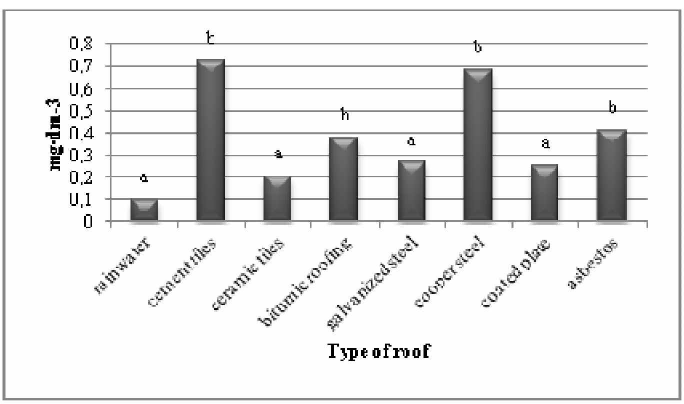

Fig. 6. Potassium content in collected water

Calcium concentration in the analyzed waters ranged from 1.6 to 25.7 mg · dm^{-3} (Table 1). The biggest amounts of this element were assessed in water collected from the roof covered with cement roof-tiles (Fig. 2). It was over 10 times more in comparison with rainwater. A similar amount of calcium was registered in the rainwater collected from eternit. This element concentration in waters from the other roof types was 1-3 times higher in comparison with the rainwater. Relative standard deviation of this element concentration for the same roof type was 18.9÷59.8%. Calcium concentration in the rainwater was small and water enrichment in this element was stated for all types of roofing. An average annual outflow of calcium from the investigated roofs may be computed on the basis of precipitation amounts and roof parameters. It is almost 7 g from cement roof-tiles and eternit roofing, whereas from the other types of roof between 0.3 and 1.5 g. Many authors point to water enrichment in calcium in result of water contact with roofing irrespective of the kind of cover. Water flowing from the copperplate roof contained between 1 and 80 times more calcium in comparison with rainwater [14]. Many authors point out to an increased calcium concentration in water collected from roofs. On the other hand, Goebel et al [13] reported that water flowing from roofs with various covering contained about 20% more calcium, irrespective of the roofing. However, in the research of the same authors rainwater contained about 5 times more calcium than the data presented in this paper. If used for irrigation it is advantageous, but higher calcium concentration worsens usable quality of water when used for washing or sanitary purposes [7].

Magnesium content in water collected from the analyzed roofs considerably differed depending on the roof covering (Fig. 3). The biggest quantities of this element, on average 1.34 mg · dm^{-3}, were assessed in water collected from roofs covered with cement roof-tiles. Enrichment coefficient of water flowing from the roofs covered with cement roof-tiles was 6.34. Water running down the eternit covered roof contained 1.04 mg · dm^{-3}, which was about 5 times more in comparison with water collected directly from the rainfall. Magnesium concentrations in water from the other roofs were lower and ranged from 0.35 mg · dm^{-3} from bitumen roofing to 0.74 mg · dm^{-3} in water collected from roofs covered with coated sheet. In all cases water enrichment in this element was noted. Relative standard deviation of this element content in waters from various roof types fluctuated from 19÷70.6% (Table 1). Kelly et al [14] stated enrichment coefficient in water running down copperplate roof on a 1-4 level. Assuming average annual rainfall in Malopolska province on the level of 650 mm and roof angle 45 degrees, the total amount of magnesium leached from 1 m^2 of roof surface covered with ceramic roof-tiles is about 400 mg, whereas from eternit roof slightly over 300. For the other roofs the value fluctuated between 100 and 200 mg.

Sodium content in the analyzed waters ranged from 0.176 to 1.497 mg Na · dm^{-3} (Table 1). The smallest quantities of this element were assessed in rainfall water. Water enrichment in this element was observed for all kinds of roofs. Increase in sodium content in water flowing from the roof was observed on the two to 3-fold level (Fig. 4). The greatest amounts were assessed in water from roofs covered with cement roof-tiles and copperplate, whereas the least for coated sheet roofing. Changes in sodium concentrations in the analyzed waters were not statistically significant. A considerable changeability of this element concentrations in water was observed, ranging between 30.4% in water from ceramic roof-tiles and 59% for eternit roofing. Goebel et al [13] states sodium concentration in water running down the roofs with various covering in the range between 0.22 and

22 mg Na · dm^{-3} , however no dependencies were stated between this element quantity in water and the kind of roofing.

Average phosphorus content in rainwater was 32.2 mg · dm^{-3} (Fig. 5). Among all samples, the greatest quantities of phosphorus, 0.417 mg · dm^{-3}, were assessed in water collected from the eternit covered roof and from copperplate roof where its amount exceeded 0.130 · dm^{-3}. A very low changeability of this element concentration was registered for roofs of the same type. It ranged from 0.86% in water from copperplate covered roof to 33% for bitumen roofing (Table 1). Phosphorus concentrations in water running down roofs with various covering, similar to presented in this paper, were reported by Farreny et al [12]. A slight enrichment in this element was noticed in water flowing from the roof covered with cement roof-tiles, copperplate or coated sheet. A decrease in this element quantity in relation to rainwater was registered for the roof covered with ceramic roof-tiles and bitumen material. This element amount leached from 1 m^2 of roof was estimated on the level of 150 mg for the eternit roof, 39 mg for coated sheet roof and 12 mg for copperplate roof. The amount of phosphorus from the other roofs had no importance.

Average potassium content in water flowing from various roofing fluctuated from 1.57 to 15.7 mg K · dm^{-3} (Table 1). Its average amount in rainfall water was 1.76 mg · dm^{-3}. A considerable increase in this element amount was observed in water collected from the roof covered with cement roof tiles and asbestos (Fig. 6). For the other roofing the element amount was slightly bigger than in rainwater. Statistically significant water enrichment in this element was observed for cement tile and bitumen roofing, copperplate and eternit. Water flowing from copperplate roof of Royal Military College of Canada was characterized by higher concentrations of this element in comparison with rainwater. Coefficient of enrichment in potassium for water flowing down the copperplate roofs ranged from 2 to 10 [14].

Development of chemistry of rainwater flowing from the roofs with various coverings is strongly connected with physicochemical and chemical properties of rainwaters, but also with the area pollution rate. The most important factor increasing the amount of dissolved substances is water pH, which affects the intensity of element leaching from the roof surface. Also chemical composition of rainwater is important, as is influences the intensity of chemical erosion of elements from the roof surface, but also provides a background for water drained from hardened surfaces. Many authors also point out to dry deposition of pollutants which in the areas under considerable anthropopressure is an important component of a total amount of pollutants drained to the environment [4]. Pollutants originating from dry deposition settle on roof surface over a long period and during rainfall are drained together with water runoff. Therefore, the first stage of the runoff may be characterized by a high content of chemical elements. It may pose a hazard to proper use of water collected from the roof. Presented investigations determined the effect of roof surface on developing the chemistry of drained waters, therefore water was collected during a rainfall period, after the second and third day of intensive rainfall. Introduction of the water from the roof collecting system is connected with retention of pollutants deposited on the roof surface during drought period, which most probably will increase total element quantity in the collected water. Determining the specificity of water gathered from individual roof types in order to create a universal way of handling water is difficult because both the quantity of elements washed out from the roof surface and the amount of air pollutants absorbed by the roof surface will increase with the roof ageing [18]. It is associated with increasing the surface and porosity of roofing due to corrosion. Another important issue is the roof surface settling by living organisms, which on one hand absorb pollutants from the air during drought periods and on the other release metabolites accelerating corrosion. It is most apparent in case of roofs covered with cement materials. They are usually overgrown with moss, which definitely changes water composition. They increase its biogen content and limit heavy metal pollution. Therefore, it may be possible that a considerable part of elements in water gathered from cement roofs originate from dry deposition temporarily captured on the roof. So, in case of water collected from cement roofing there is a hazard of algae overgrowing the containers in which the water may be stored. In case of water gathered from roofs covered with coated sheet or copperplate there is a hazard of excessive water enrichment in zinc, nickel or copper, or in hydrocarbons in case of bitumen roofs. It may limit the use of water for certain purposes. Despite rainwater enrichment in the analyzed elements, no worsening of its properties was registered from the viewpoint of possible utilization. Macroelements in water do not pose any hazard to the environment, but may reduce water applications. The idea of creating rainwater collecting system assumes setting up artificial retention in the urbanized and transformed areas. At an average precipitation total in Poland, 72 m^3 of water may be collected from a building with an area of 120 m^2. Its reuse for irrigation allows for one hand to decrease surface runoff of rainwater and on the other the irrigated areas will act as wastewater treatment plants. By using water for washing or sanitary purposes we contribute to saving treated water of high quality. Water from roofs collected for ponds will allow to increase biodiversity in the urbanized areas and improve retention capacities of the whole area. The most frequent way of rainwater recycling in the world is using the water for watering lawns and toilet flushing, whereas only 8% is used for car washing.

Conclusions

1. Contents of the analyzed macroelements in rainwater were not high and did not point to an anthropogenic enrichment. Obtained results were similar or lower than these elements concentrations in precipitation water in the other regions of the world.
2. Water runoff from roofs significantly modified water chemical properties.
3. Generally the highest water enrichment in the analyzed elements was assessed in samples from roofs covered with eternit and cement roof tiles.
4. The lowest concentrations of the analyzed elements in rainwater were registered for roofs covered with ceramic roof tiles or bitumen roofing.
5. Almost half lower concentrations of phosphorus in comparison with rainwater were assessed in water collected from roof covered with ceramic tiles.
6. Water gathered form roofs was characterized by a higher pH in comparison with rainwater.
7. Considering possible applications of the analyzed water collected from roofs, it may provide a valuable source of water for the purposes which do not require the highest quality water.

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KSZTAŁTOWANIE SIĘ CHEMIZMU WÓD OPADOWYCH W WYNIKU SPŁYWU Z DACHÓW O RÓŻNYCH POKRYCIACH. CZĘŚĆ I

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Abstrakt: Celem pracy było określenie składu chemicznego wód spływających z dachów domów z różnym pokryciem na tle ich zawartości w wodzie deszczowej. W niniejszej pracy przedstawiono zawartość wapnia, magnezu, sodu, fosforu i potasu. Na podstawie Rozporządzenia Ministra Środowiska z dnia 24 lipca 2006 r. w sprawie warunków, jakie należy spełnić przy wprowadzaniu ścieków do wód lub do ziemi oraz w sprawie substancji szczególnie szkodliwych dla środowiska wodnego spływy z terenów utwardzonych traktowane są jako ścieki, natomiast spływy z dachów budynków traktowane są jako wody czyste i można je odprowadzać do środowiska bez pozwolenia wodnoprawnego. Dane literaturowe wskazują jednak na możliwość znacznego wzbogacenia wody deszczowej w czasie kontaktu z pokryciem dachowym. Badaniami objęto 24 dachy domów jednorodzinnych lub małych budynków pełniących funkcje siedzib firm handlowo-usługowych. Jako tło do badań użyto wody opadowej zebranej w dwóch losowo wybranych miejscach na terenie obszaru badań. Badania przeprowadzono na terenach o niskiej antropopresji, aby jak najlepiej uchwycić wpływ rodzaju pokrycia dachowego na kształtowanie się chemizmu wód. Obszar badań zlokalizowany był we wsi Lusławice w powiecie tarnowskim, województwo małopolskie. Badaniami objęto dachy najczęściej występujące na badanym terenie: dachówka cementowa, dachówka ceramiczna, pokrycie bitumiczne, blacha ocynkowana, blacha miedziana, a także eternit. W badanych próbkach wód oznaczono zawartość wapnia, fosforu, magnezu, sodu oraz potasu. Dodatkowo oznaczono wartość pH wody oraz przewodność elektrolityczną. Wyniki przeprowadzonych badań wskazują, że woda spływająca z badanych dachów w każdym przypadku zawierała większe ilości analizowanych pierwiastków w porównaniu z wodą deszczową, przy czym na przykład średnia zawartość magnezu w wodzie spływającej z dachówki cementowej była prawie dziesięciokrotnie większa niż w wodzie deszczowej, w przypadku dachówki ceramicznej było to prawie 5 razy więcej, natomiast woda z pokryć bitumicznych zawierała około 3 razy więcej magnezu, natomiast woda z blach ocynkowanej i powlekanej zawierała około 6 razy więcej tego pierwiastka w porównaniu z wodą deszczową. Także w przypadku pozostałych pierwiastków największe współczynniki wzbogacenia odnotowano w wodach z pokryć cementowych (dachówka i eternit), następnie z blach ocynkowanych i powlekanych, a najmniejsze wzbogacenie odnotowano w wodach z dachów o pokryciach z dachówek ceramicznych. Odnotowano także zwiększenie się wartości przewodności elektrolitycznej w wodach z dachów o pokryciach cementowych, blachy ocynkowanej oraz dachówki ceramicznej. Wszystkie spływy z dachów odznaczały się generalnie większą wartością pH w porównaniu do wody deszczowej; największy odczyn - około 8 - stwierdzono w wodzie z dachówki cementowej, nieco niższy, około 7,0, w wodzie z dachów z dachówki ceramicznej i blachy miedzianej. Wartość pH wody blachy powlekanej była niższa niż w wodzie deszczowej i oscylowała w granicach 5,5. Odczyn deszczówki wynosił 5,94.

Słowa kluczowe: zbiorniki wód opadowych, zanieczyszczenia wody, spływy z dachów, sód, potas, wapń, magnez, fosfor