

AFTER-REMEDIATION MONITORING OF PAH CONCENTRATION IN GROUNDWATER OF AIRPORT SLIAČ – SOUTHERN REGION (SLOVAKIA)

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Abstract: Old environmental burdens, such as the area of Airport Sliač-South, are a source of multiple contaminants which are able to transfer to the surrounded environment and, therefore, pose a serious risk for the environment itself and human health. Among the group of contaminants polycyclic aromatic hydrocarbons (PAHs) are also classify and determined in the groundwater within the after-remediation monitoring from 2015 to 2020. In total, 7 sampling spots were chosen at the southern area of the airport. Groundwater samples were taken twice a year into prepared sample boxes. The samples were delivered to the State Geological Institute of Dionýz Štúr laboratory in Spišská Nová Ves within 24 hours after collection. Major part of PAHs in all the samples and throughout the period of measurements was represented mostly by naphthalene. The relationship between the concentrations of PAHs and TOC as well as between the concentrations of PAHs and NES was studied. Positive correlation was proved in the relationship between the concentration of PAHs and NES. The result of the after-remediation monitoring of studied locality results into the conclusion that the extent of pollution is decreasing. Concentrations of NES and TOC exceeding the limits (Directive of the Ministry of the Environment SR 1 / 2015-7 for risk analysis) can be still found in the samples of groundwater from the bores within the source locality of environmental burden. Up to 2016, the concentrations of PAHs were exceeding the limits as well. Generally cited guidelines indicate that the groundwater samples manifest poor quality of the groundwater and refer to the importance of after-remediation monitoring of sanitized environmental burdens.

Keywords: polycyclic aromatic hydrocarbons (PAHs), naphthalene, groundwater, non-polar extractable substances (NES), total organic carbon (TOC)

1. INTRODUCTION

Old environmental burdens can be explained as polluted areas posing a serious risk to human health and the environment, and often comprise a wide range of sites that have been contaminated by industrial, military, mining, transportation, or agricultural activities, as well as improper waste management. At present, environmental burdens present significant environmental problems mainly because they are

sources of considerable contamination, mainly by organic pollutants.

Non-polar extractable substances (NES) belong to significant pollutants not only in the Slovak Republic, but also in other parts of the world (Onwurah et al., 2007). The NESs originate mainly in the exploitation, transport and washing of oils, the processing of oil and coal, the production, storing and use of oil and coal products and it may also be produced by plants, animals, and microorganisms (Bozek et al., 2011).

PAHs are present in many fuel mixtures and have long been on the US Environmental Protection Agency (EPA) list of priority pollutant (Samanta et al., 2002). PAHs are highly toxic, mutagenic and carcinogenic substances present in all spheres of the environment, including the groundwater (Wu et al., 2008). These hydrophobic hydrocarbons feed long term pollutions in aquifers (Bouzid et al., 2017) and interact more strongly with particulate matter, therefore they are more persistently bound to sediments (Zhou et al., 2000); (Zhou et al., 1996). They are very mobile, and a possible grave threat to the environment (Krzebietke et al., 2020); and they have a detrimental effect on the flora and fauna of affected habitats, resulting in the uptake and accumulation of toxic chemicals in food chains and in some instances, in serious health problems and/or genetic defects in humans (Samanta et al., 2002). Remediation of areas contaminated by PAHs is a complicated process which does not exclude their persistence in the environment (Zeledón-Toruño et al., 2007; San Miguel et al., 2009) which is the main reason for thorough after-remediation monitoring of these substances.

More than 1800 burdens are registered in Slovakia, one of them being the Airport Sliac-South site which, according to the Register of Environmental Burdens in Slovak Republic, belongs to the sanitized or recultivated sites (SEA for the Minister of the Environment of the Slovak Republic, 2020). Organic contaminants in the investigated area of Sliac-South Airport fall exclusively into the category of non-polar extractable substances (NES) including NES-CI (C_{10} - C_{40} compounds), polycyclic aromatic hydrocarbons (PAHs), and come from the past military activities of the Soviet army in the region. The dominant contaminants in the studied locality are fuels, especially aviation gasoline, so it is not surprising that PAHs appear as contaminants in soil, groundwater, and surface water. Contamination of soil and groundwater by these substances was observed in the locality in question.

Several exploratory remediation works were carried out at the Sliac site, which took place in sequential stages. In the years 1981 – 1989, a survey and remediation pumping work took place at the site. In this phase, groundwater was pumped out and the oil matter was separated from it subsequently. Sources of pollution were not removed, and contamination proceeded (Méry & Vojtech, 1987). In the years 1990 – 1992, the pumping of the oil layer from the groundwater level continued and a survey of the pollution was carried out. Based on the results of the survey, remediation works (bioventing) were

carried out in 1998 – 2001, and subsequently, from 2001 – 2010 company INGEO carried out remediation pumping. During remediation works for groundwater decontamination, about 2,850,000 m³ of groundwater was treated. By 2008, about 23,000 L of free phase of petroleum substances were removed from the groundwater level, approximately 7,000 kg of petroleum substances determined as NES-IR, approximately 1,400 kg of petroleum substances determined as NES-CI, approximately 4.5 kg of aliphatic chlorinated hydrocarbons and 13 kg of aromatic hydrocarbons. During the remediation work, the concentration of NES-IR decreased to a value lower than the limit of 1,000 mg kg⁻¹ dry matter on the area of more than 50% of the contaminated area (Drahoš et al., 2011). In June 2015, selected infrastructure facilities left by the Soviet Army were rehabilitated. Ex-situ remediation of land was carried out in June 2015, together with the remediation of buildings after the Soviet army. Soil remediation consisted of excavating the contaminated soil at the site of the greatest pollution, removing it, and disposing of it on the decontamination area. The remaining areas were remediated by in-situ methods.

At the final phase of remediation works, the intensified sanitation of groundwater was performed in order to reach optimal values. Part of the remediated area (PHM Juh-1 silos) remained contaminated even after the process. The reason was that the physical removing of building objects was denied, so the access to the soil underneath was impossible (Schwarz et al., 2017).

2. MATERIALS AND METHODS

2.1. Characteristics of investigated region

Airport Sliac-South is located in the central part of Slovak Republic, on the northern edge of the Sliac municipality, in the urban territory of Sielnica (Zvolen district). The total area of the site is 1,514,702 m² (Ministry of Defense of the Slovak Republic, 2013). In the past, the location served the Soviet army as an airport, massive repositories for fuels, and part of the site was used for the barracks. At present, military activities (airport, repository, etc.) are carried out in the site without handling of oil substances.

The buildings used by Soviet army are located on the right bank of the river Hron. The Airport Sliac complex is located on both sides of the local road from Banská Bystrica to Zvolen.

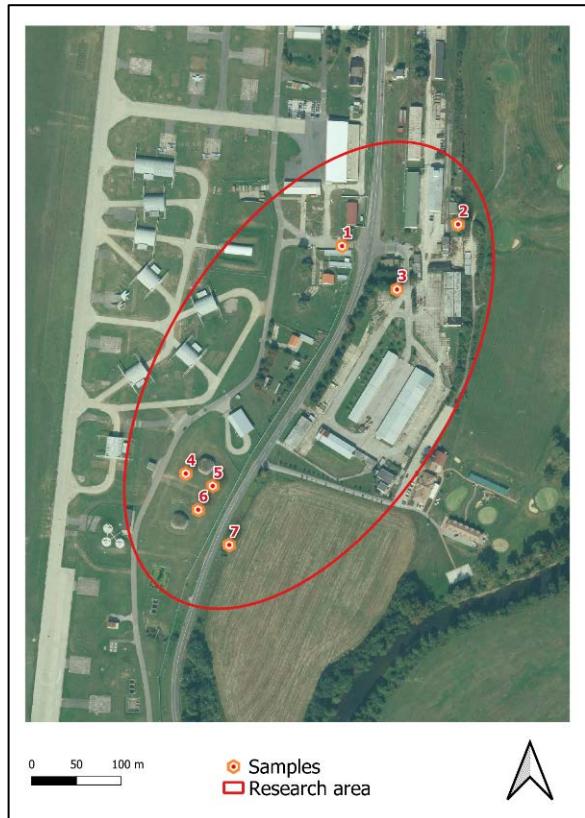


Figure 1. Situational map of investigated region, Airport Sliač-South, with marked sampling spots. The region investigated in this article is framed in red.

The Airport Sliač is classified as an environmental burden and the area is partially rehabilitated. According to the register of Environmental Burdens of Slovak Republic, the Airport Sliač-South site is classified as the environmental burden type C - remediated site (Strategic Environmental Assessment for the Ministry of the Environment of the Slovak Republic, 2020). The direction and intensity of possible contamination spread from the burden is influenced mainly by the hydrogeological regime of groundwater related to the river Hron. External precipitation water enters the burden and is enriched with soluble and insoluble substances. Such contaminated water reaches the groundwater level after passing the aeration zone and may contaminate the aquifer in its vicinity. Any contamination then spreads further in the direction of groundwater movement into the environment in the form of a contamination cloud. The direction of groundwater flow at the site is south-east, towards river Hron. Soil contamination is located about 6 m below the ground and groundwater contamination was located throughout the collector, including the collector base.

2.2. Sampling

The sampling of the surface watercourses was supplemented by the sampling of groundwaters since March 2015. The list and characteristics of monitored sites (sampling spots) in Airport Sliač-South, also depicted in Fig. 1, is given in Table 1.

Table 1. List and characteristics of sampling spots in Airport Sliač-South

Sampling spot	Function	Label
1	Source drill	VN30-1
2	Indication drill	VN30-2
3	Source drill	VN30-3
4	Source drill	VN30-4
5	Source drill	VN30-5
6	Source drill	VN30-6
7	Indication bore	VN30-7

Groundwater samples were taken after approximately 20 minutes of pumping, after stabilization of the basic physico-chemical parameters of water (temperature, conductivity, pH, acidity, O₂ saturation). The sampling of water was performed by a GIGANT pump, 20% power, from the middle of the water column. In case of a low water column, a groundwater collector (bailer) designed by the Eijkenkamp company, was used. Groundwater samples were taken twice a year into prepared sample boxes - Glass wide-mouth bottles delivered to the State Geological Institute of Dionýz Štúr laboratory in Spišská Nová Ves within 24 hours after collection, where the analysis was performed to determine polyaromatic hydrocarbons (PAHs), hydrocarbon index (NES CI) and total organic carbon (TOC). The analytical procedure also included sample pretreatment to prepare the samples for the analysis itself.

2.3. Extraction and analysis

PAHs were isolated from the water samples by solid phase extraction (SPE, Empore disk). Water sample (100 ml) was passed through a Bond elut C18 – 500 mg / 6 ml. The columns were dried, adsorbed analytes were eluted with 5 ml of n-hexane, and after concentrating the extract on the 0,2 ml, the samples were analyzed by gas chromatography with a mass detector. An Agilent GC 7890B – MS 5977B was used for analysis.

NES CIs were isolated from water samples by the liquid-liquid extraction. Pollutants were extracted from the water samples with n-hexane. After separating the polar extractables by adsorption on silica gel and

concentration, the extract was analyzed by gas chromatography with a FID detector. A Varian GC FID 3900 instrument was used for analysis.

Table 2. Results of after-remediation monitoring of contaminants in the groundwater of Airport Sliač-South carried out from 2015 to 2020

Sampling spot/ Label	Year	Total PAH	NAPH	TOC	NES CI	DGW
		[$\mu\text{g L}^{-1}$]	[$\mu\text{g L}^{-1}$]	[mg L^{-1}]	[mg L^{-1}]	[m]
1/ VN30-1	2015	0.68	0.19	3.20	0.00	4.64
	2016	1.72	1.59	2.38	0.00	4.50
	2017	0.85	0.52	2.30	0.00	4.55
	2018	0.05	0.00	2.30	0.00	4.30
	2019	1.73	1.13	2.00	0.00	4.82
	2020	0.00	0.00	1.80	0.03	4.76
2/ VN30-2	2015	0.49	0.33	1.60	0.00	5.78
	2016	0.05	0.00	0.66	0.00	5.58
	2017	0.24	0.24	1.10	0.00	5.70
	2018	0.02	0.00	1.40	0.00	5.57
	2019	1.55	0.71	1.30	0.00	5.85
	2020	0.13	0.10	1.80	0.00	5.84
3/ VN30-3	2015	3.11	2.91	4.90	0.00	5.18
	2016	1.40	1.30	3.07	0.00	5.30
	2017	0.10	0.10	3.00	0.00	5.19
	2018	0.01	0.00	2.40	0.00	4.98
	2019	1.33	0.71	2.50	0.00	5.36
	2020	0.03	0.00	3.20	0.05	5.32
4/ VN30-4	2015	188.98	186.00	12.20	4.90	3.28
	2016	17.21	15.90	11.30	2.94	3.26
	2017	27.23	26.70	13.50	1.41	3.04
	2018	4.33	4.12	10.40	2.22	2.98
	2019	2.64	1.87	11.30	2.30	3.36
	2020	2.23	0.98	10.20	0.07	3.29
5/ VN30-5	2015	86.90	85.50	11.70	1.15	3.28
	2016	136.91	136.00	11.20	2.26	3.30
	2017	45.76	45.50	9.50	1.06	3.20
	2018	0.06	0.00	10.00	3.82	5.05
	2019	3.48	1.87	10.40	2.00	3.40
	2020	0.76	0.49	8.90	1.08	3.32
6/ VN30-5	2015	148.75	147.00	15.00	1.42	3.20
	2016	90.33	89.30	15.50	2.97	2.97
	2017	30.08	29.40	12.30	3.13	2.93
	2018	7.21	6.99	7.10	0.75	2.63
	2019	2.58	1.42	8.60	1.13	3.00
	2020	0.30	0.09	5.70	1.32	2.86
7/ VN30-7	2015	4.22	3.99	3.10	0.02	4.43
	2016	6.76	6.59	2.32	0.00	4.54
	2017	0.25	0.25	2.50	0.00	4.40
	2018	0.03	0.00	2.50	0.13	4.70
	2020	0.09	0.05	2.20	0.07	4.46

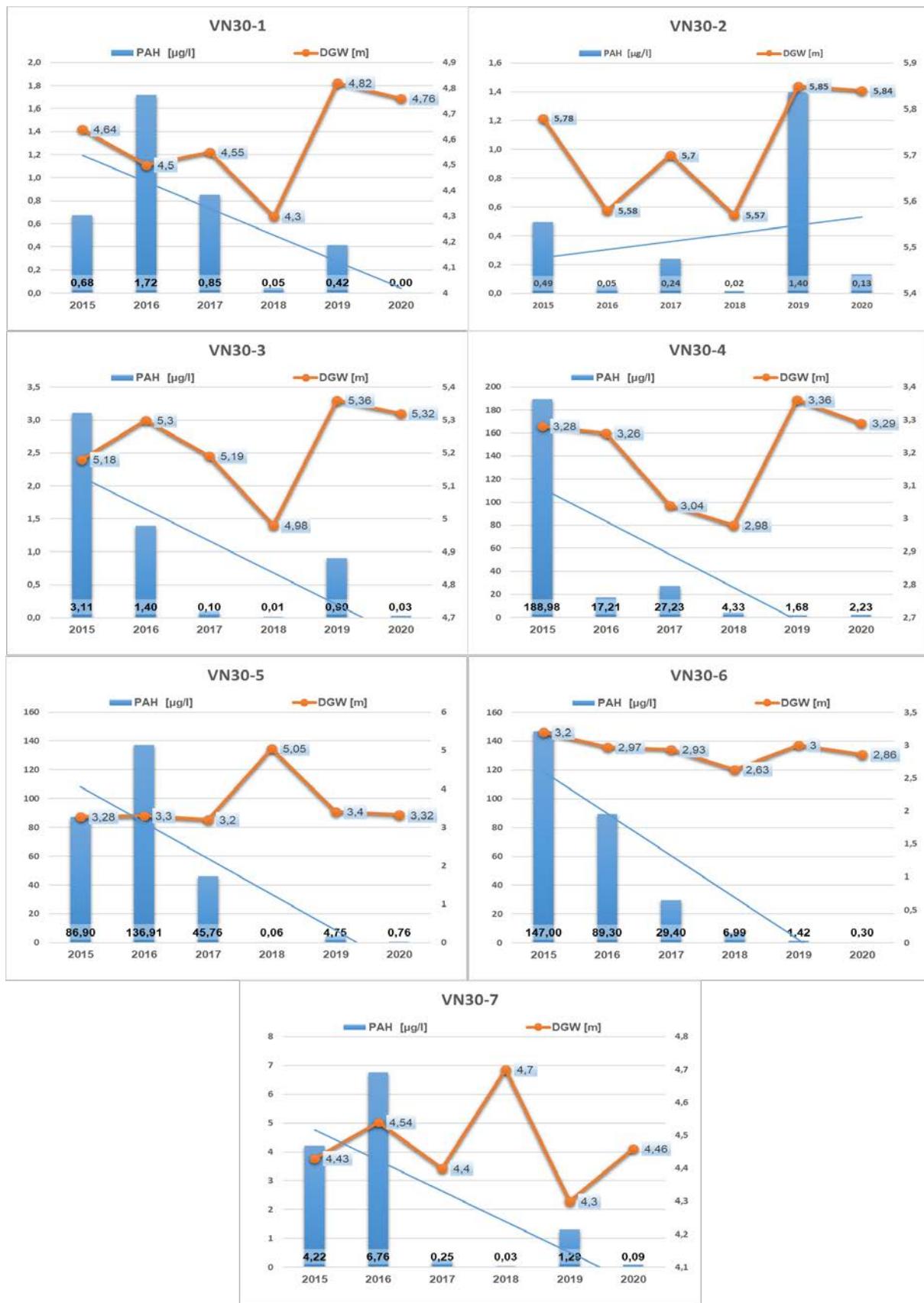


Figure 2. Change of PAH concentration in the sampling spots according to depth and year of sampling (2015 – 2020)

TOC A Tim-V-CPN organic carbon analyzer from Shimadzu with accessories (ASI-V autosampler) was used to determine the TOC. The essence of the method is the high-temperature oxidation of the carbon present in the samples in the oxygen stream, as a result of which the carbon is converted to carbon dioxide, which is detected on a non-dispersive infrared detector. The TOC is determined by the method of the difference between total and inorganic carbon (TC - TIC = TOC). The instrument is calibrated for determination in the concentration range from 0.05 to 40%.

Groundwater level (DGW) measurements were performed with a Solinst - Model: 107 TLC portable level meter and a G30 portable level meter. The groundwater level was measured from upper edge of the metal drill guard.

2.4. Statistics

The interrelationship between the contents of PAHs, naphthalene and TOC were interpreted by Spearman rank correlation coefficients where the significance was detected at $p \leq 0.05$. The analyses were processed in software application R, version 4.0.0. Graphical outputs were prepared in IBM SPSS Statistics 19; and in Microsoft Excel 2016.

3. RESULTS

After-remediation monitoring of the contaminants in seven different sampling spots within the Airport Sliac-South brought results that indicated significant change in the concentration of total PAHs, naphthalene (NAPH), total organic carbon (TOC) and NES CI. The remediation took place in 2015 and the systematical sampling started off in March 2015. Table 2 demonstrates the concentrations of studied contaminants in samples from individual sampling spots since 2015 to 2020. In Figure 2, the results are depicted graphically, also considering the respective groundwater levels (DGW) in individual sampling spots.

The highest groundwater levels have been documented in reference drills (sampling spots 4, 5, 6). The lowest groundwater levels were measured in drills located at the exit from the airport area and closest to the surface flow (sampling spots 2 and 3). Changes in the concentration of total PAH have a decreasing character in all sampling spots except for sampling spot 2. The highest value of total PAH concentration, $188.98 \mu\text{g L}^{-1}$, was measured in 2015 in the groundwater sample from the sampling spot 4, and after this year, the

concentration of total PAH had significantly decreasing trend. In 2019, the total PAH concentration in the same sampling spot decreased to $2.64 \mu\text{g L}^{-1}$. Significantly declining trend in the total PAH concentrations appeared in all sampling spots, especially after 2017, when the maximum concentration did not exceed $7.5 \mu\text{g L}^{-1}$, as depicted in Figure 2. Significant part of total PAH concentration in all sampling drills throughout the whole monitoring period was presented by naphthalene (NAPH), as demonstrated in Table 2. In the groundwater samples taken from the monitored drills, changes in the NES and TOC concentrations with declining character were observed throughout the monitoring period. The concentration of NESs in a sample from drill 4 was 4.90 mg L^{-1} in 2015 and slightly decreased to 0.07 mg L^{-1} in 2020. Also, reduction in TOC content occurred in drill 6, where the initial concentration 15.00 mg L^{-1} dropped throughout the monitoring period to 5.70 mg L^{-1} .

Also, the highly significant correlation can be described, with Spearman rank correlation coefficient almost equal to 1, between the concentrations of total PAH and naphthalene (Fig. 3).

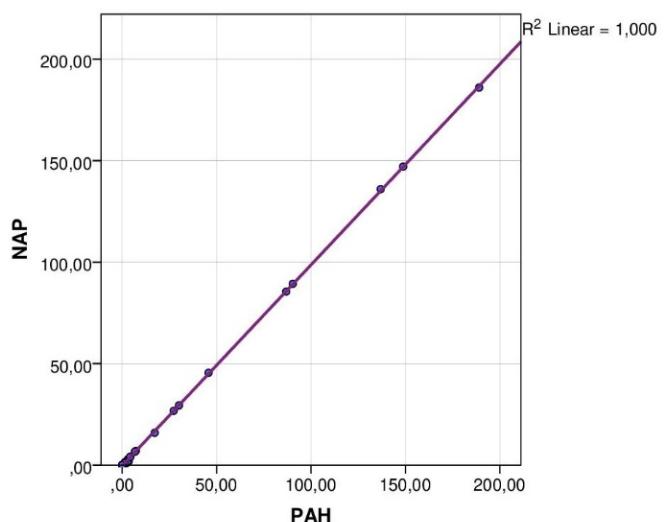


Figure 3. Graphical depiction of the correlation between total PAH/ NAPH concentrations

The data about the content of individual contaminants were processed for mutual correlation expressed by means of Spearman rank correlation coefficient for PAH/ NES content ($r = 5.57$) and PAH/ TOC content ($r = 0.72$) with both tuning out to be significantly positive. The graphical displays of correlation data for both PAH/ NES and PAH/ TOC concentrations are shown in Figure 4.

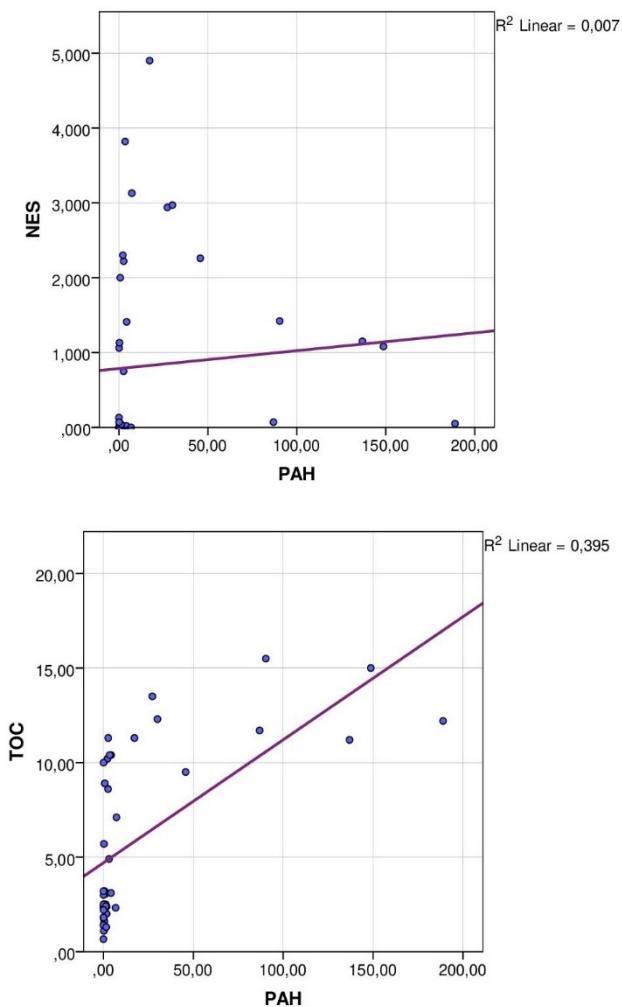


Figure 4. Graphical depiction of the correlation between total PAH/ NES concentrations (up) and total PAH/ TOC concentrations (down)

4. DISCUSSION

The measured values of total PAH, NES and TOC concentrations in individual drills in the investigated region after the remediation confirmed the decreasing trend in the occurrence of organic contaminants. A higher level of pollution was observed in samples from sampling spots located in the source area of environmental load, particularly in drills 4, 5 and 6, where are the unremoved fuel silos located. There can be presumed that the contamination is still persistent. When comparing the concentration of total PAH content in the groundwater samples to the values of indication ($60 \mu\text{g L}^{-1}$) and intervention criteria ($120 \mu\text{g L}^{-1}$), according to the Directive of the Ministry of the Environment SR 1 / 2015-7 for risk analysis, both of them were greatly exceeded in 2015, especially in

groundwater samples taken from boreholes 4 and 6 as well as in 2016, in the groundwater sample taken from borehole 5. The intervention criterion ($60 \mu\text{g L}^{-1}$) according to the directive (Ministry of Environment of the Slovak Republic, 2015) was exceeded in the groundwater samples taken from drill 5 in 2015 and drill 6 in 2016. As demonstrated in the results, there is a strong correlation between PAHs and naphthalene concentrations, and for this reason, even when the values of the indication and intervention criteria are exceeded, the results are very similar. There are smaller differences due to the lower limit for the naphthalene indication criterion, which according to (Minister of Environment of the Slovak Republic, 2015) is set at $25 \mu\text{g l}$ and the intervention criterion at $50 \mu\text{g/l}$. The values of the indication criterion were demonstrated for naphthalene in groundwater samples taken from drills 4, 5 and 6 in 2017. The values of the intervention criterion for naphthalene exceeded groundwater samples in 2015 for drills 4, 5 and 6 and in 2016 for drills 5 and 6. As mentioned by Schwarz (2017), increased contamination level in years 2015 and 2016 interrelates with intense sanitation of groundwater in final phase. So therefore, a higher content of PAHs here was expected. The other 15 PAHs, which are on the US Environmental Protection Agency's (EPA) list of priority pollutants, and which were also monitored in groundwater samples, did not exceed the indication or intervention criteria. For this reason, we can agree with the statement (Kerr, JM, 2001), which assessed in crude oils and condensates collected from oil production sites around the world and the potential risk of direct exposure of soil and leaching to groundwater for 15 of the 16 priority PAH pollutants were not significant (Kerr, 2001). found that naphthalene in particular is PAH, which may be present in concentrations that exceed the level of risk.

The results of the analytical determination of selected organic indicators (Table 1) proves a generally reduced groundwater quality in the whole examined area in the TOC indicator, where the value of the indication criteria of 2 mg L^{-1} was exceeded in almost all samples except groundwater samples in drill 2 and intervention criteria 5 mg L^{-1} in drills near the source area of contamination 4, 5, 6. The relationship of PAHs with TOC has also been investigated and a positive correlation was observed between total PAH and TOC concentrations. This indicates that (Bozek et al., 2011) partitioned in organic matter may be the possible source of PAHs in water (Saba et al., 2012) and in study in Europe (Nam et al., 2008). However, it is clear from the

measured concentrations and correlations of TOCs and PAHs that PAHs affect the concentration of TOCs, but are not the only source.

The NES indicator exceeded the limit values according to (Minister of Environment of the Slovak Republic, 2015) determined for the indication criteria to the value of 0.25 mg L^{-1} and the intervention criteria to the value of 0.50 mg L^{-1} in groundwater samples from boreholes located in the source area. environmental load (drills 4, 5, 6). According to the conclusions from the works where the remediation work also took place (Drahoš et al., 2011) the results of the risk analysis documented that at a distance of 100 m from the original pollution sources (reference distance) the content of pollutants determined as NES CI to the level of the limit values for their determination. It was stated that no risk of spreading groundwater pollution was identified (Drahoš et al., 2011). Similarly, from the results of numerical modeling of groundwater flow (Drahoš & Kovářík 2011) it was concluded that the immediate source of pollution in the locality Sliač fuel warehouses - South does not pose a risk from the spread of NES-CI pollution in groundwater.

To a certain extent, the statement of the final report (Drahoš et al., 2011). contrasts with the above statements, where the authors state that in the pre-remediation period before 1992, the pollution in the rock environment could migrate, and for this reason it cannot be ruled out that part of this pollution also reached the deeper part of the Neogene gravels. Furthermore (Drahoš, et al., 2011). state that the transport of pollution is very, very slow ($<30 \text{ m / year}$) and it follows from the above that pollution in deeper circulation can appear on the surface only after many decades. from the period of heavy pollution of the Quaternary gravel aquifer (Drahoš et al., 2011). Many studies (Zhou et al., 1996; Bour et al., 2003; Brasington et al., 2007; Onwurah et al., 2007; Schwarz et al., 2011; Bouzid et al., 2017) point to the fact that petroleum hydrocarbons are able to cause long - term pollution of water layers and to be trapped in soil sediments and can be partially removed by different remediation methods depending on the nature of the contamination. Several of the studies also admit the possibility, as he also stated (Drahoš et al., 2011), that pollution may appear in even deeper circulation after several decades. The results of analyses performed within the monitoring period also point to this possibility, after which the concentrations of individual contaminants have a decreasing trend after remediation, but it is still not possible to say that the quality of

groundwater due to organic pollution is still not at such a level as not to pose a possible risk to the environment. For this reason, groundwater monitoring should continue to be an important part of this area.

5. CONCLUSION

After-remediation monitoring of the Airport Sliač-South ranging from 2015 to 2020 points to the declining nature of organic pollution, especially in the indicator of PAHs, which in 2018 and 2020 did not exceed the values of the indication or intervention criteria. They also point to a strong linear correlation between naphthalene and total PAH concentration and also to its strong representation in the monitored groundwater samples. A linear correlation was also observed in TOC and total PAH, NES and total PAH. It is clear from the results that with decreasing concentration of PAHs, a lower concentration of other indicators of organic pollution was also observed, although PAHs alone are not their only source. The above study concluded that post-ground groundwater monitoring is an important part of monitoring changes in pollution. It is therefore important to include it in the monitoring of the state of the environment in the studied locality and further, in the long term. We see its justification mainly for substances that tend to persist for a long time in the environment, because pollution in the deeper circulation cannot appear on the surface until many decades.

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