



# Effects of mine water discharge on river sediments: metal fate and behaviour, Upper Silesian Coal Basin

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## Abstract

The study aims to characterise the changes in elemental composition in the river sediments of streams influenced by mine waters enriched with radionuclides. The study took place in the vicinity of Ostrava, a city located in a coal mining region in the Czech Republic, namely the Upper Silesian Coal Basin. River sediments and waters of the Karvinský potok and Stružka streams were investigated. Field measurements were made for ambient dose equivalent rate (ADER). Laboratory gamma spectrometry and X-ray fluorescence were used to determine the content of radionuclides and elemental composition in river sediments. Water samples were analysed for the content of major ions and radionuclides. The field ADER measurement proved elevated content of radionuclides with values exceeding 1,000 nSv/h in both streams. The discharged mine waters were Na–Cl type, containing an <sup>226</sup>Ra (0.68–0.70 Bq/l) as a dominant radionuclide. Laboratory measurements of radionuclides in bottom sediments proved that the prevailing source of radiation are <sup>226</sup>Ra and <sup>232</sup>Th in both streams. The calculated enrichment factors showed extreme values for Sr, Cr, Pb, Zn, Cu, and Mo. The precipitation reactions forming Ca-minerals (calcite and aragonite), Fe-bearing minerals (hematite, goethite and amorphous Fe(OH)<sub>3</sub>) and hausmannite were found to be the primary geochemical process underway in the studied riverine systems. The correlation between elements and radionuclides demonstrated the significant role of geochemical barriers that lead to the precipitation of radionuclides from solution. The results show that the precipitation takes place preferentially in places where other waters enter the stream, or where recent organic matter is present.

**Keywords** Mine water · Metals · River sediments · Radionuclides · Coal mining

## Introduction

Neutral or alkaline mine water is much less common in mining areas than acid mine ones. Thus, a limited number of studies on the distribution of elements in river environments influenced by neutral/alkaline mine water has been published (Dahrazma and Kharghani 2012; Kelly et al. 2006). Coal, depending on the coalification stage, contains several

potentially risk elements. Among the elements of greatest concern are chalcophilic elements—As, Cd, Hg, Pb or Se (Swaine 1990; Suárez-Ruiz et al. 2006). Increased levels of radium (Ra) or uranium (U) have also been documented in the hard-coal coal basins (Bondaruk et al. 2015; Gombert et al. 2019; Janson et al. 2009; Schmid and Wiegand 2003).

Radium is an alkaline earth element with four natural isotopes (<sup>223</sup>Ra, <sup>224</sup>Ra, <sup>226</sup>Ra and <sup>228</sup>Ra), of which only two—<sup>228</sup>Ra ( $\beta$ ) and <sup>226</sup>Ra ( $\alpha$ )—are significant in terms of environmental protection. Other Ra isotopes have a very short half-life and/or extremely low concentration. These two isotopes are part of the decay series of primordial radionuclides: <sup>228</sup>Ra with a half-life of 5.75 years is a part of the thorium series (with parental isotope <sup>232</sup>Th) and <sup>226</sup>Ra with a half-life of 1600 y is a part of the uranium series (with parental isotope <sup>238</sup>U) (Bourdon et al. 2003; USEPA 2004; IAEA 2014). The activity of Ra isotopes in rocks and minerals is usually the same as that of its parent (Th or U), i.e., it is in secular equilibrium. The shift of this

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equilibrium is observed in the waters of the hypergenic zone (Chevychelov et al. 2021).

The  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  concentrations in the environment are extremely low. Carmichael (1989) stated that the concentration of  $^{226}\text{Ra}$  in the earth's crust was 0.9 ng/kg (33 Bq/kg); its concentration in soils was determined by Bowen (1979) to be 0.8 ng/kg. Activities in river water generally range between 0.5 and 20 mBq/L for  $^{226}\text{Ra}$ , though enhanced concentrations (of up to 300 mBq/L) have been reported (Vandenhove et al. 2010).

Uranium (U) and Th in the Earth's crust and soils are present in mg/kg, which are much greater concentrations than those of Ra. Rudnick and Gao (2003) stated that the abundance of U for the upper crust was 2.7 mg/kg (33 Bq/kg). In soils, the U concentration varies greatly—from 0.1 to 53.2 mg/kg. Thorium (Th) concentrations in rocks and soils are generally higher than those of U. According to Rudnick and Gao (2003), the concentration of Th is 10.5 mg/kg (43 Bq/kg) in the upper crust and is 3 to 4 times higher than that of U (Ahrland et al. 1973).

It was expected that the content of radioactive elements in the overburden is negligible compared to sulphates, chlorides, and other potentially hazardous elements such as Cd or Pb (Cravotta 2008; Chalupnik et al. 2017; Gombert et al. 2019). However, the increased level of naturally occurring radionuclides e.g. in shales, bituminous shales, igneous silicate rocks or phosphate rocks is documented across the world (Chalupnik et al. 2017; Paridaens and Vanmarcke 2001; Pujol and Sanchez-Cabeza 2000). Thus, the extraction of such raw materials can lead to the accumulation of radionuclides in tailings, fly ash or river sediments, even though the limits for the entry of radionuclides into the environment are met (Chalupnik et al. 2001; Lauer et al. 2017; Papastefanou 2010).

The first data on radioactivity in the Upper Silesian Coal Basin mines come from Poland and date back to the 1970s (Tomza and Lebecka 1981). Since 1989, the coal industry in Poland has been required to monitor the radioactivity of discharged mine waters and sediments (Chalupnik et al. 2001; Lebecka et al. 1994). Data on the occurrence of radionuclides in the surface water in the Czech part of the Upper Silesian Coal Basin (USCB) are available only to a limited extent in the monitoring schemes of state institutions (Hanslík et al. 2005). Besides the aforementioned radionuclides, the discharged mine waters contain elevated concentrations of Fe, Mn, Sr and Ba (Vöröš et al. 2021). Both Sr and Ba belong to alkaline earth metals and behave similarly to Ra. The similarity of element behaviour is used to monitor the fate of radium in the riverbed, but detailed studies evaluating elemental composition and radioactivity monitoring are scarce (Iyengar 1990). The main aim of the present study is to describe the changes in the elemental composition of the

riverine system influenced by mine water in the Upper Silesian Basin.

## The studied area

The studied streams are situated in the Czech part of the Upper Silesian Coal Basin (USCB), which, with a total area of 7,500 km<sup>2</sup> (from which only 1,550 km<sup>2</sup> is in the Czech Republic), is one of the biggest hard coal basins in Europe and has a long mining history. The basin is divided into the Ostrava and Karviná sub-basins (Dopita and Kumpere 1993). The present USCB is only a denudation remnant of what was initially a much larger basin structure filled with Devonian and Lower to Upper Carboniferous sediments (Jirásek et al. 2018; Geršlová et al. 2016).

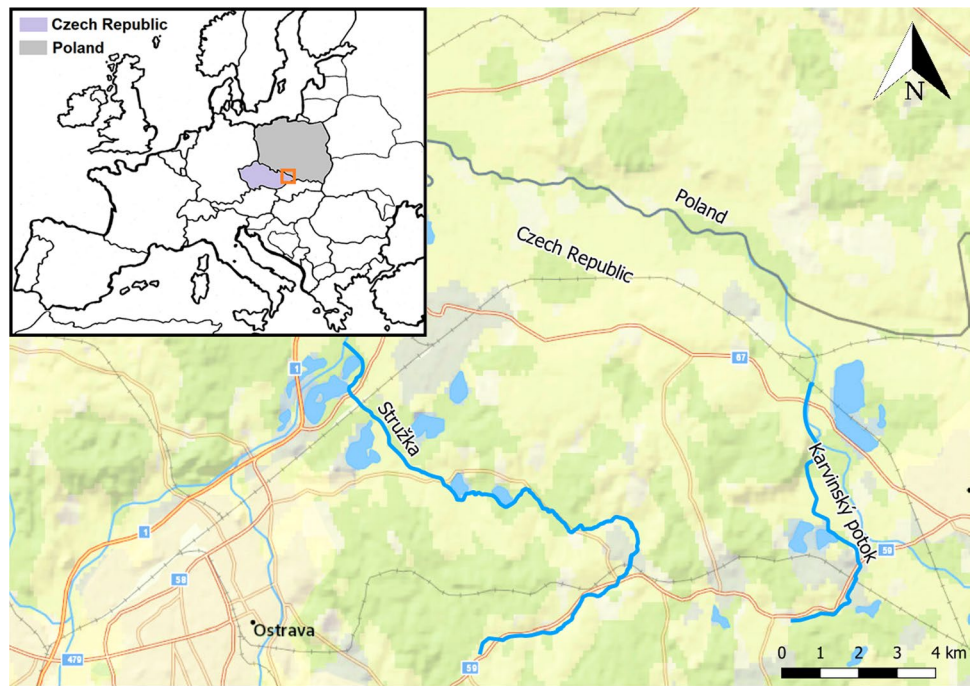
### Karvinský potok

Karvinský potok is located in the Karviná sub-basin and its channel was artificially modified. The stream's length is 8.4 km and it begins as a stream draining an old reclaimed coal heap. The mine water discharge is located 2.6 km away from the beginning of the stream and enters a small artificial pond (approximate dimensions: 15 × 60 m) with a bottom and sides made of concrete (points S1-2 and S1-3). Moving down the stream, the broad floodplain area (points S1-4) is followed by a channelled section; at sampling location S1-8 the river bed is lined with a concrete bottom and banks and flows to Lake Kozinec, which is a subsidence lake. Downstream of the lake, Karvinský potok is a regulated stream and takes the form of a straightened channel with steep banks. The natural part of Karvinsky potok has an occasional flow that works mainly during the rainy season, otherwise the mine waters are the main water source (Fig. 1).

### Stružka

Stružka is located in the Ostrava sub-basin. The total length of the stream is 14.2 km; it is affected by mine subsidence with a total grade of 60 m and is naturally low in water. The beginning of Stružka has characteristics of a dale with many rather small rapids and erratic bottoms. After 270 m from the mine water discharge, another mine water-influenced stream flows into the Stružka. Approximately 1.4 km from the mine waters discharge, there is a wastewater treatment plant, which represents a significant water input. The remaining part of the stream is a regulated stream bed with stabilized banks. The discharged mine water represents approximately 1/4 of the total volume of water. The estimate is based on average volumes of discharged water and measured flow data during the year.

**Fig. 1** Map of the studied area with a marked position of investigated streams



## Methodology and equipment

Two streams into which mine water is discharged were selected – Karvinský potok and Stružka. To evaluate the mine water's influence and its extent, conductivity, pH and radioactivity levels were monitored directly in the field (Tables 1 and 2, Fig. 2). Eighteen samples of river sediments, 12 samples of surface water and 2 samples of discharged mine water were collected from both streams. The content of 7 radionuclides and 19 elements was determined in the river sediments and the content of main ions and radionuclides was determined in the water samples.

### Field measurements

The in-situ measurement of ADER was done using a gamma spectrometer GT-40 (Georadis Ltd.) equipped with a NaI (TI) detector with a volume of 345 cm<sup>3</sup>, diameter 76 mm (cylinder). GT-40 is a device capable of accurate measurement of ADER values from 20.0 nSv/h to 0.5 mSv/h within an energy range of 25 keV to 3 MeV. The in-situ measurement was done by walking directly in the streams and the device was carried in hand partially submerged under the water so the detector would be still at the same distance from the sediment. Conductivity, pH and temperature were measured by Multimeter WTW Multi 350i (with accuracy  $\pm 0.01$  for pH,  $\pm 0.5\%$  for conductivity and  $\pm 0.1$  °C for temperature) equipped with probe WTWSenTix 41 for pH and temperature and probe WTW TetraCon 325 for conductivity measurements. The conductivity, temperature and pH

of stream water were measured in a single day during active mine water discharging straight into the streams.

### Sampling and sample preparation

The samples of sediments from the streams were taken at places with elevated ADER values. The samples were dark brown to black clayey silty sand with a rare occurrence of bigger grains. Sample S1-2 contains corroded fragments of piping (up to 2 cm in size). The fine-grained river sediments were taken with a scoop and placed in plastic bags. Dry samples were sieved with a mesh size of 2 cm to remove plant debris and larger detrital pieces. The obtained fraction was used for the laboratory gamma spectrometry and particle size distribution. The Marinelli beakers with a volume of 600 ml were filled and hermetically sealed for 30 days to reach the secular equilibrium for <sup>226</sup>Ra and <sup>222</sup>Rn. For the XRF analyses, the homogenized samples were crushed to a fraction less than 0.2 mm.

Samples of water were collected into plastic sampling bottles directly from the streams and were transferred to the laboratory within 24 h and immediately analysed.

### Laboratory measurements

#### Water samples

<sup>226</sup>Ra was analysed by the JKA 300 analyser equipped with probe NS 9502 E using an accredited radiochemical methodology. The 1 L of water is chemically processed

**Table 1** Hydrochemical data of surface and mine waters analysed. For location see Fig. 2 with ID

ID	Locality, distance from discharge in meters	226Ra [Bq/l]	U-238 [Bq/l]	226Ra/238U	EC [ $\mu$ S/cm]	pH	HCO <sub>3</sub> - [mg/l]	Cl- [mg/l]	(SO <sub>4</sub> ) <sub>2</sub> - [mg/l]	Ca <sup>2+</sup> + [mg/l]	Mg <sup>2+</sup> + [mg/l]	Na + [mg/l]	K + [mg/l]	Ba <sup>2+</sup> + [mg/l]	Fe [mg/l]
S1 W2 DIS	Karvin-ský potok, dis-charge	0.70	0,008	93.7	20,700	7.3	115.4	6 974.3	77.4	320.4	96.4	3 805.7	50.8	6.4	0.18
S2 W2 DIS	Stružka, dis-charge	0.68	0,010	67.3	16,600	7.4	828.1	6 849.5	26.8	141.4	117.7	4 317.6	39.6	15.5	0.18
S2 W1	Stružka, -100	0.005	0,008	0.6	660	7.2	151.7	64.1	107.8	53.0	17.3	51.7	10.6	0.07	0.16
S2 W3	Stružka, 120	0.04	0,011	3.4	820	6.9	204.3	55.2	205.0	88.7	34.4	53.1	8.4	6.90	0.12
S2 W4	Stružka, 250	0.03	0,020	1.6	653	7.3	150.8	51.3	133.8	70.5	22.1	38.0	9.1	0.72	0.18
S2 W5	Stružka, 280	0.05	0,016	3.0	2,350	7.5	211.7	537.1	200.0	72.6	66.3	327.2	21.3	0.52	0.15
S2 W6	Stružka, 360	0.03	0,013	2.3	4,910	6.8	312.1	1 381.2	308.7	188.7	91.0	799.5	21.6	ND	0.16
S2 W7	Stružka, 890	0.04	0,027	1.4	7,440	7.0	373.8	2 274.0	391.8	246.5	128.1	1 310.3	30.9	ND	0.12
S2 W8	Stružka, 4400	0.02	0,019	1.0	440	6.9	135.9	43.4	54.3	39.4	14.9	27.1	5.1	ND	0.15
S1 W1	Karvin-ský potok, -750	0.005	0,025	0.2	1,360	7.8	338.3	100.7	322.3	77.1	33.4	139.0	13.8	0.05	0.16
S1 W3	Karvin-ský potok, 10	0.06	0,026	2.2	24,800	7.3	183.7	8 837.7	214.0	427.2	295.8	6 901.8	67.7	0.84	0.16
S1 W4	Karvin-ský potok, 100	0.23	0,011	20.9	10,760	6.9	217.7	4 024.6	186.0	218.5	67.4	2 273.0	35.1	0.68	0.18
S1 W5	Karvin-ský potok, 260	0.05	0,018	2.9	4,290	7.7	292.6	1 137.9	279.1	111.9	38.6	737.5	19.4	0.23	0.16

Table 1 (continued)

ID	Locality, distance from discharge in meters	<sup>226</sup> Ra [Bq/l]	U-238 [Bq/l]	<sup>226</sup> Ra/ <sup>238</sup> U	EC [μS/cm]	pH	HCO <sub>3</sub> - [mg/l]	Cl- [mg/l]	(SO <sub>4</sub> ) <sub>2</sub> - [mg/l]	Ca <sup>2+</sup> + [mg/l]	Mg <sup>2+</sup> + [mg/l]	Na + [mg/l]	K + [mg/l]	Ba <sup>2+</sup> + [mg/l]	Fe [mg/l]
S1 W6	Karvin-ský potok, 2300	0.04	0,017	2.2	2,030	8.1	183.8	8 837.7	214.0	249.6	502.2	4 596.5	67.7	0.28	0.16

ND not detected

and the residue after evaporation is subsequently mixed with ZnS(Ag) and measured. For <sup>226</sup>Ra each sample was measured twice with the efficiency of 0.66 for 2000 s and the background of 23imps. According to the activity of the sample, the average uncertainty is up to 20%.

The content of chlorides was determined by the precipitation with silver nitrate via titration with a measurement uncertainty of 10%. The equivalence point was determined potentiometrically (electrode pH/mV meter Seven2Go S2 Std-Kit Mettler Toledo). The concentration of bicarbonate ions was analysed by potentiometric method at pH 4.5 and pH 8.3 with 10% measurement uncertainty. The content of sulphates ions was determined by precipitation of barium ions under moderate acid condition with 10% measurement uncertainty. Barium sulphate was filtrated and weighed. The Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup> and Ba<sup>2+</sup> contents were determined by atomic absorption spectrometer Agilent 280FS AA. For each element the appropriate wavelength was used. All methods meet requirements for accredited analyses in accordance with ISO/IEC 17025 (control of calibration, blanks and standards measurements, interlaboratory comparisons).

The resulting charge balance errors were at the level of ± 10%.

**Sediment samples**

Particle size distribution was carried out on sieved homogenized samples using the CILAS 1064 instrument.

Gamma spectrometry was done by HPGe (high purity germanium) coaxial spectrometers with high resolution and efficiency of 4% (HK-8G with carbon window), 25% (HK-1G and HK-2G with beryllium window), 30%, 35% and 40% and with 8,192 channels to 3,000 keV. The measurement time was at least 24 h. Activity of 7 radionuclides was determined: <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>238</sup>U, <sup>228</sup>Th, <sup>232</sup>Th and <sup>210</sup>Pb with deviations of up to 12% for <sup>226</sup>Ra, 10% for <sup>228</sup>Ra, 36% for <sup>235</sup>U, 31% for <sup>238</sup>U, 10% for <sup>228</sup>Th, 10% for <sup>232</sup>Th and 75% for <sup>210</sup>Pb. We used the following sources as a standard for energy and efficiency calibration for laboratory gamma spectrometric determination of the concentration of radionuclides: Activity standard: Type CBSS 2; produced by the Czech Metrological Institute, a mixture of radionuclides – Am-241; Cd-109; Ce-139; Co-57; Co-60; Cs-137; Ba-133; Sr-85; Y-88; Cr-51 a Pb-210. Activity standard: type MBSS 2X; produced by the Czech Metrological Institut; a mixture of radionuclides – Ra-226 a Th-23.

The elemental composition was measured by XRF Delta Premium with Rh source, in the settings of Geochem Vanad. Metranal 19, Metranal 34, Nist 2702, Nist 2781 and IRM 5718As were used as reference materials. For each sample, the measurement time was 300 s. The content of 19 elements (Si, Al, Fe, Ti, K, Ca, Mn, Sr, S, Cr, Zr, Rb, Ni, Zn, Cu, Pb, As, Mo, Th) was used for further evaluation. The LODs



**Table 2** The results of gamma spectrometry analyses in the river sediments of Karvinský potok (S1) and Struzka (S2)

ID	K-40	Ra-226	Ra-228	U-238	Th-228	Th-232	Pb-210	Ra-226/ U-238
	Bq/kg	Bq/kg	Bq/kg	Bq/kg	Bq/kg	Bq/kg	Bq/kg	
S1-2	510	10,575	3293	323	2336	5630	109	32.74
S1-3	490	2494	1327	127	741	2068	175	19.71
S1-4	501	1346	670	100	392	1062	114	13.46
S1-5	430	1102	361	113	371	733	270	9.75
S1-6	496	568	159	33	160	319	172	17.16
S1-7	282	946	589	150	200	789	66	6.30
S1-8	360	777	230	80	192	422	262	9.71
S2-1	411	1547	851	304	283	1133	139	5.08
S2-2	469	1964	1084	258	266	1351	190	7.62
S2-3	433	3000	1473	233	528	2001	113	12.88
S2-4	449	5600	2532	657	1015	3547	189	8.53
S2-6	445	5885	2853	286	1056	3909	301	20.30
S2-7	503	5299	2116	235	1287	3403	166	22.55
S2-8	415	3883	1886	163	731	2617	114	23.86
S2-9	415	2299	1026	82	634	1660	154	28.03
S2-10	287	1963	702	46	554	1256	152	43.07
S2-11	438	1055	593	317	180	772	123	3.33
S2-12	332	744	254	37	197	451	123	20.11

declared by the InnovXSystem is as follows for Si < 0.5%, Al < 0.5%, Fe = 10 mg/kg, Ti = 7–15 mg/kg, K = 40–60 mg/kg, Ca = 25–40 mg/kg, Mn = 10 mg/kg, Sr = 1–3 mg/kg, S = 150–300 mg/kg, Cr = 5–10 mg/kg, Zr = 1–3 mg/kg, Rb = 1–3 mg/kg, Ni = 10–20 mg/kg, Zn = 3–5 mg/kg, Cu = 5–7 mg/kg, Pb = 2–4 mg/kg, As = 1–3 mg/kg, Mo = 1–3 mg/kg, Th not declared.

## Data processing

The results were processed using Microsoft Excel and QGIS 3.2.2. The spectra were evaluated using G2k and GENIE 3.2.3 (Mirion–Canberra). The results were corrected using MEFFTRAN (freeware).

Data normalisation was applied to the assessment of anomalous metal contribution using Al as a reference element. Data from the publication by Rudnick and Gao (2003) were used as the background values. The following formula was used for enrichment factor calculation (Zoller et al. 1974 and Sinex and Helz 1981):

$$EF = (A/An)/(B/Bn)$$

A = the content of the element in the river sample.  
An = the content of the reference element in the river sample.  
B = the content of the element in the background samples.  
Bn = the content of the reference element in the background samples.

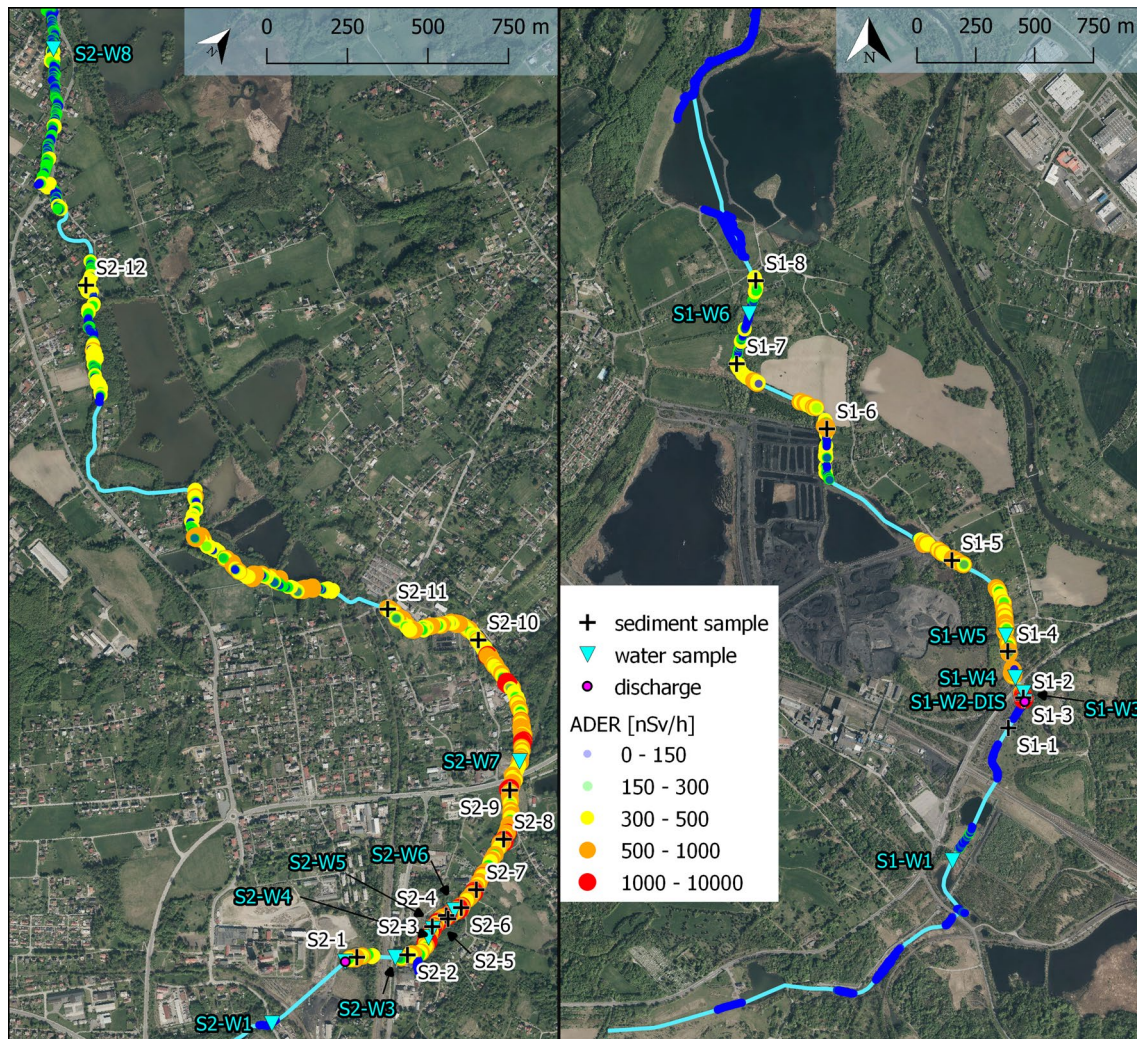
Hydrochemical modelling was conducted to study the solid–liquid interactions using PHREEQC software. This

included the calculation of the mineral saturation index (SI) for both streams to compare the mineral stabilities in the water. The water composition (concentration of dissolved ions and anions), pH, and T values were used as input parameters for the SI calculation. SI is defined as an ion activity product in relation to solubility product on a logarithmic scale where SI value > 0 means saturation of mineral phases in the water, while SI values < 0 represent subsaturation.

## Results and Discussion

### Discharged water samples

The discharged mine waters have a stable temperature (23–25 °C) according to long-term measurements done by Diamo a.s. The pH of discharged waters was comparable (7.3–7.4), as was EC with the values of 20,700 μS/cm and 16,600 μS/cm for Karvinský potok and Stružka, respectively (Table 1). Both discharged waters (S1-W2-DIS and S2-W2-DIS) are classified as chloride types. In the cationic part, the proportion of alkali metals Na<sup>+</sup> and K<sup>+</sup> is more than 80 eq.%. It is generally assumed that the mine waters of USCB contain a substantial concentration of sulphates (Grmela and Jelínek 1999). These opinions are also supported by findings of sulphate minerals inside the mines (Matýšek et al. 2014) and the general assumption that coal contains an increased amount of sulphur (Deurbrouck 1972). However, the concentration of sulphates (SO<sub>4</sub>)<sup>2-</sup> in the studied mine water



**Fig. 2** Field ADER in-situ measurements on Stružka (left) and Karvinský potok (right) with marked sampling points. Mine water discharge points are adjacent to the S2-1 and S1-2 points

discharges was low (not exceeding 77 mg/l). This deficit of  $\text{SO}_4^{2-}$  in mine water can be explained as either insufficient oxidation of sulphides in the mined bedrock or reached saturation limits of gypsum and/or jarosite and their precipitation (Banks 2006). A low content of mainly pyritic sulphur in coal-bearing strata of the Ostrava and Karvina Formations was found in boreholes in the Karvina region (Vöröš et al. 2022). Thus, low sulphates content will be conditioned by a lower pyrite content contained in the rock massif. A lower amount of Ba reaching 6.4 and 15.5 mg/l was found in the discharge water of Karvinský potok and Stružka, respectively (Table 1). However, the values are not as high as from the discharge of an active mine (up to 99 mg/l, Vöröš et al. 2021). Even higher content of Ba was measured in Polish parts of USCB, where in water samples the concentration was up to 2,000 mg/l (Lebecka et al. 1994). The measured activity of  $^{226}\text{Ra}$  in discharged waters was 0.70 Bq/l and

0.68 Bq/l for Karvinský potok and Stružka, respectively. Both mine waters has an extreme excess of  $^{226}\text{Ra}$  compared to  $^{238}\text{U}$  (Tab. 1).

### Surface water samples

According to the ion composition the surface waters are classified as carbonate-sulphate, however, after the mine water discharge, it changes to chloride type. The pH values do not show a visible trend along both streams and are in the range of 6.8–8.1. The discharge of mine waters significantly increased the electrical conductivity values in both streams (Table 1). The subsequent development of conductivity is different for individual streams. In Karvinský potok, the EC decreases from 24,800 to 2030  $\mu\text{S}/\text{cm}$  within a 5.7 km long section. In Stružka, the EC it does not fall immediately after discharge, but on the contrary rises and then

falls. The activity ratio between  $^{226}\text{Ra}/^{238}\text{U}$  is significantly lower (1.0–20.9) compared to its value in the discharged mine waters (93.7 in Karvinský potok and 67.3 in Stružka; Table 1).

### River sediment samples

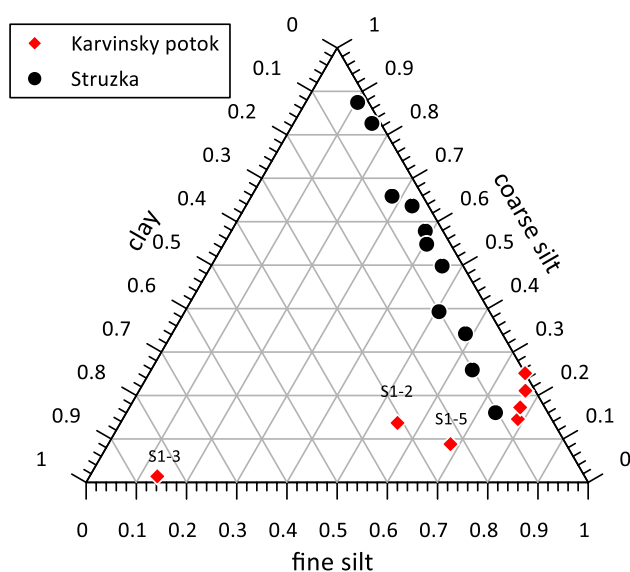
The field ADER measurement proved an increased content of radioactivity in both streams (Fig. 2). Highest measured radioactivity (up to 4000 nSv/h), with an average of 1,831 nSv/h was found directly after mine water discharge in the Karvinský potok. No place with ADER above 200 nSv/h has been found upstream of the mine water discharge. The highest ADER values exhibited in the Stružka exceeding 2000 nSv/h were measured 200 m after mine water discharge with an average of 898 nSv/h (Fig. 2). Laboratory gamma spectrometry of bottom sediments proved that the most active radionuclides in river sediments are  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in both studied streams (Table 2).

Stružka river sediments were composed of silt (10.5–75.4% w/w) and sand fraction (16.1–87.5% w/w). On the contrary, in the Karvinský potok sediments the granulometric distributions of the particles were less variable and the most abundant fraction is silt fraction (Median 80.5% w/w). Sample S1-3 is an exception with the prevailing sand fraction (85.1% w/w) and 1.2% w/w content of clay fraction (Fig. 3).

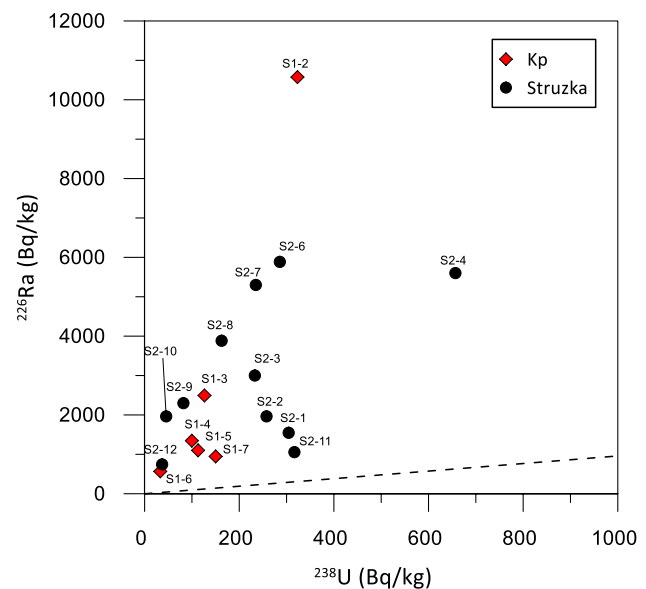
The  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratios of siliciclastic rocks can be used as a parameter sensitive to sediment recycling and the weathering process. With increasing sediment maturity, quartz is

enriched compared to feldspars, mafic minerals and lithics (Roser et al. 1996). The average  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratios in unaltered igneous rocks ranged from 3.0 (basic rocks) to 5.0 (acidic rocks). The  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio in the Karvinský potok ranges from 3.2 to 5.0, which confirms basic to intermediate maturity. Values of  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio  $> 5.0$  in Stružka sediments pointed to increased geochemical sediment maturity. There has been found no correlation between Al and Si ( $r=0.14$ ). It follows from the above that sediment material of both rivers is made up mainly of detrital components of weathered rocks with a minor portion of soil flushes from the stream surroundings.

A strong positive correlation was found for Al and K ( $r=0.92$ ) in both streams, and this is interpreted as the presence of clay minerals or feldspars. The sample S1-2 completely deviates from the evaluated set, as it contained corroded fragments of piping with a diameter of up to 2 cm (Fe content = 204 508 mg/kg). Samples S2-1, S2-2 and S2-11 are enriched in Fe content (over 48 720 mg/kg). The different element distribution was found in samples S2-3 and S2-4 where a low content of Fe (31 955 mg/kg and 23 321 mg/kg, respectively) and an increased content of Ca (average of 35 000 mg/kg) was measured (Appendix 1). The ratio of  $^{226}\text{Ra}$  and  $^{238}\text{U}$  in the river sediments of Karvinský potok is in the range 6.3–32.7 and in Stružka in the interval 3.3–43.1 (Table 2). All evaluated sediments contain  $^{226}\text{Ra}$ , which is not in secular equilibrium with its parent radionuclide  $^{238}\text{U}$  (Fig. 4). The ratio of  $^{226}\text{Ra}$  and  $^{238}\text{U}$  in sediments decreases downstream, but a systematic gradual decrease could not



**Fig. 3** Ternary diagram of the granulometric distributions (%) of the finest fractions (clays  $< 2 \mu\text{m}$ ,  $2 \mu\text{m} < \text{fine silts} < 20 \mu\text{m}$  and  $20 \mu\text{m} < \text{coarse silts} < 63 \mu\text{m}$ ) in the stream sediments of the Karvinský potok and Stružka streams



**Fig. 4** Activity concentration for  $^{226}\text{Ra}$  and  $^{238}\text{U}$  in the stream sediments of the Karvinský potok (Kp) and Stružka. The dashed line expresses the line of secular equilibrium between the monitored radionuclides



be traced. This is probably due to the natural movement of sediments within the stream. The most abundant radionuclide  $^{226}\text{Ra}$  correlates with  $^{228}\text{Ra}$  and  $^{232}\text{Th}$  in all evaluated samples, which reflects the different mobility of uranium, radium, and thorium in the environment (Fig. 5). Uranium is highly soluble under oxidising conditions, whereas radium is mobile regardless of oxidation–reduction environmental conditions. Thus, radium movement in the environment is controlled by precipitation or sorption.

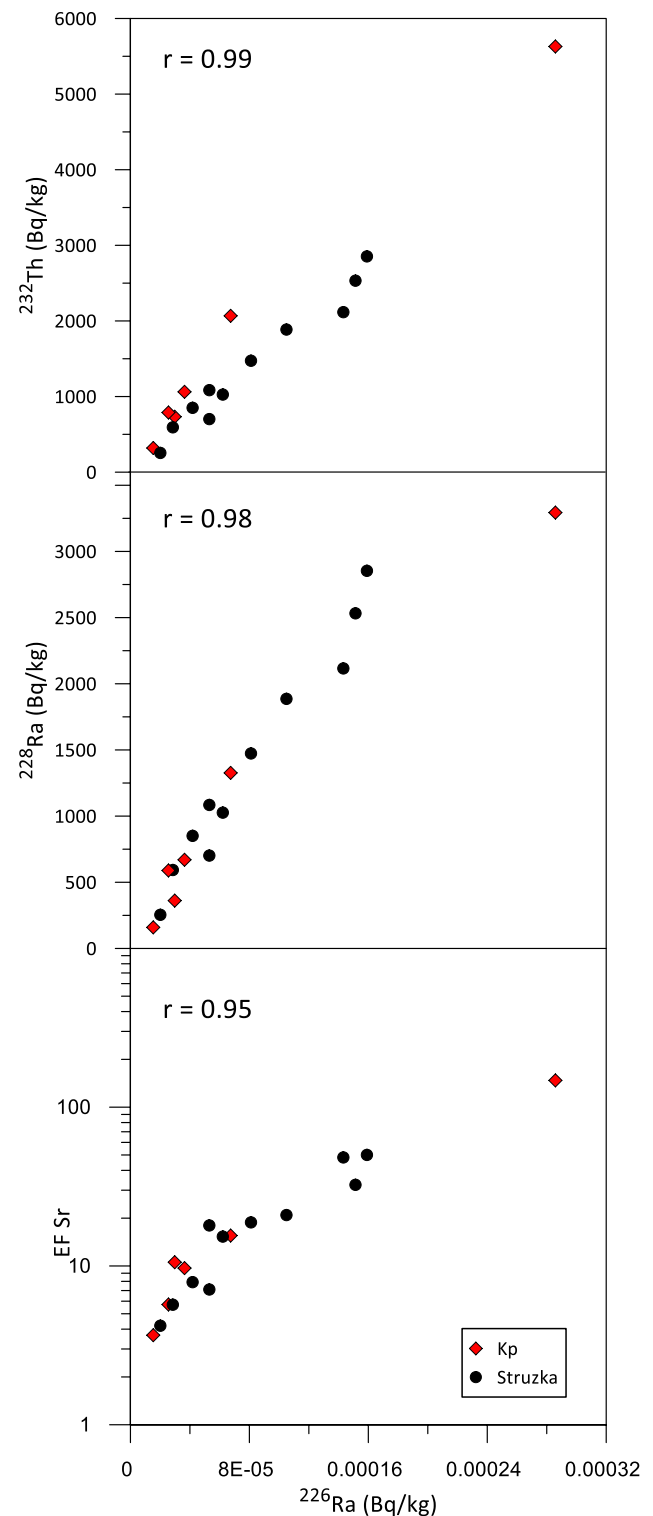
To assess the mutual relationship between river sediment composition and radionuclides, it was necessary to eliminate the effect of grain size. Thus, the enrichment factor (EF) using Al as the normalising element was calculated. The EF for both streams reached extreme values for Sr, Cr, Mo, Th, Pb, Zn and Cu (Fig. 6).

A mutual correlation was found between the enrichment factor of Sr and the content of  $^{226}\text{Ra}$  (Fig. 5, Appendix 2). This is related to their oxidation states (+2) which do not change under typical groundwater conditions, thus their mobility in aquifers is indirectly affected by changes in geochemical conditions leading to precipitation or sorption onto negative mineral surfaces.

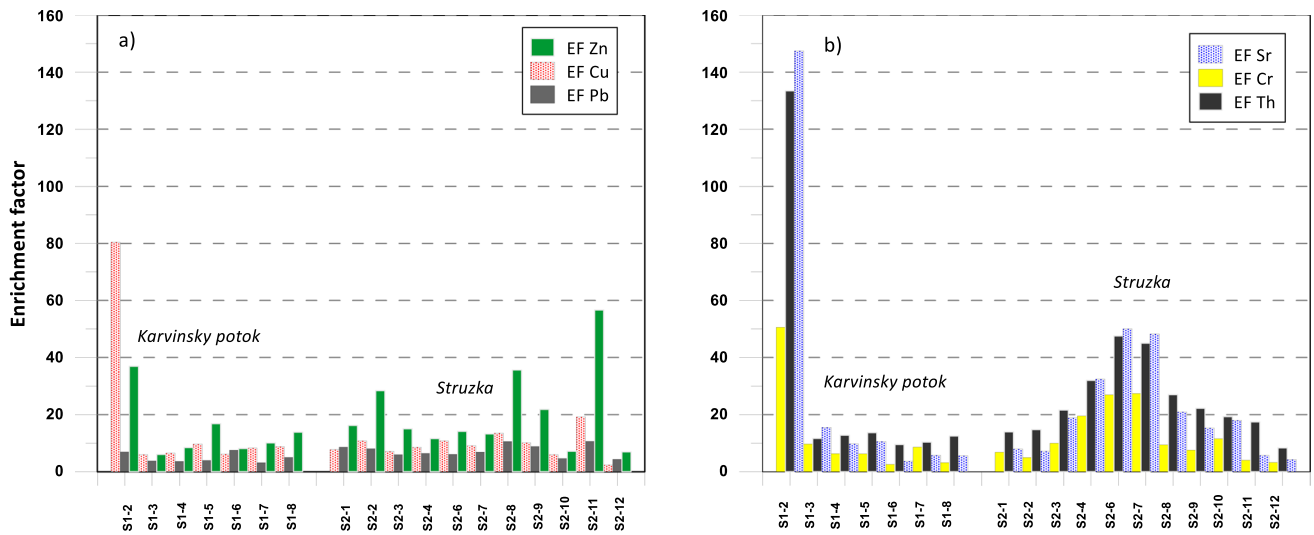
### Saturation index and mineral dissolution

Two geochemical models applied for the results from discharge mine water (MW) and mixing discharge water with surface water (MIX) were evaluated in both Stružka and Karvinský potok (Table 3). The mineral saturation indices (SI) were calculated from the quarter-month monitoring data and revealed positive values for calcite and dolomite, averaged of 0.41 and 0.97 for Stružka; averaged of 0.49 and 0.93 for Karvinský potok (MW), respectively. This observation indicates supersaturation in respect to their solubilities (Table 3). Other hydrous carbonate phases such as aragonite showed lower SI values compared to aforementioned minerals, averaged of 0.27 for Stružka, and 0.35 for Karvinský potok.

The calculated internal partial pressure of  $\text{CO}_2$  showed negative  $\log(p\text{CO}_2)$  values in both streams, not exceeding  $\log(p\text{CO}_2)$  for the Earth's atmosphere -3.4. It reflects on the potential degassing of  $\text{CO}_2$  from the solutions to the atmosphere (Boch et al. 2015). According to the results, both streams are supersaturated with respect to the amorphous and hydrous iron minerals, such as hematite, goethite and Fe-oxo hydroxides ( $\text{FeOH}_3$ ). The SI values highly fluctuated and their values differed from each other. Amorphous ( $\text{FeOH}_3$ ) exhibited the lowest SI values in Stružka and Karvinský potok, averaged of 1.71 and 2.25, respectively. Goethite and hematite showed much higher SI values in both streams with a slight difference between the two systems. This is in agreement with the observation of the first section of Stružka where Fe-hydroxide coatings were observed. The



**Fig. 5** Correlation of the activity concentration for  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ ,  $^{228}\text{Ra}$  and Sr enrichment factor (EF Sr) in the Karvinský potok (Kp) and Stružka stream sediments



**Fig.6** Enrichment factor (EF) values calculated for stream sediments from the Karvinsky potok and Struzka for Zn, Cu, Pb (a) and Sr, Cr, Th relative to the standardization element Al

**Table 3** Calculated Saturation index (S.I.) of minerals for discharged mine waters and mixture of mine water and surface water

Mineral S.I	Discharged water						
	Struška			Karvinský potok			
	MIN	MAX	Average	MIN	MAX	Average	
SI <sub>Goethite</sub>	6.91	8.20	7.56	7.34	8.96	8.15	
SI <sub>Hematite</sub>	15.83	18.41	17.12	16.68	19.93	18.31	
SI <sub>Fe(OH)3</sub>	1.09	2.33	1.71	1.44	3.06	2.25	
SI <sub>Calcite</sub>	0.31	0.51	0.41	- 0.43	1.41	0.49	
SI <sub>Dolomite</sub>	0.77	1.17	0.97	- 0.90	2.75	0.93	
SI <sub>Aragonite</sub>	0.17	0.36	0.27	- 0.58	1.27	0.35	
SI <sub>Barite</sub>	0.83	1.38	1.11				
SI <sub>Jarosite-K</sub>	- 6.38	- 1.59	- 3.99	- 4.03	- 0.27	- 2.15	
Mineral S.I	Mixture of mine and surface water						
	Struška			Karvinský potok			
	MIN	MAX	Average	MIN	MAX	Average	
SI <sub>Goethite</sub>	7.84	9.09	8.46	2.00	8.71	5.36	
SI <sub>Hematite</sub>	17.67	20.18	18.93	17.59	19.42	18.51	
SI <sub>Fe(OH)3</sub>	2.03	3.31	2.67	2.00	2.91	2.46	
SI <sub>Calcite</sub>	0.12	0.25	0.19	0.63	1.30	0.97	
SI <sub>Dolomite</sub>	0.66	0.87	0.77	1.30	2.51	1.91	
SI <sub>Aragonite</sub>	- 0.03	0.10	0.04	0.49	1.16	0.83	
SI <sub>Manganite</sub>				3.61	3.96	3.79	
SI <sub>Hausmannite</sub>				5.81	6.87	6.34	
SI <sub>Pyrolusite</sub>				6.50	6.85	6.68	
SI <sub>Barite</sub>	1.85	1.98	1.91				
SI <sub>Jarosite-K</sub>	- 1.52	2.33	0.41				

SI values for goethite and hematite were averaged of 7.56 and 17.12 in Stružka; averaged of 8.15 and 18.31 in Karvinský potok, respectively.

The variability of SI values for the mineral Jarosite-K was found in both streams. Only negative averaged SI values were observed in Stružka and Karvinský potok (− 3.99 and − 2.15, respectively). When the system does not reach the equilibrium with carbonate phases (calcite and dolomite), thus Jarosite-K remains undersaturated. Precipitation/dissolution of Jarosite-K is considered when the mine water reaches the equilibrium with carbonate phases or is mixed with surface water (positive and negative SI values, Table 3), which is controlled by various factors, such as temperature, alkalinity, content of sulphates and iron in the solution regardless of the equilibrium with these phases (Elwood Madden et al. 2012). Barium (Ba) solubility in the Stružka stream seems to be controlled with respect to barite that is found to be supersaturated (average SI > 1). According to the mineralisation, pH and the content of dominant anions, Ra can co-precipitate when incorporated in barite, gypsum or even calcite, but the co-precipitation with gypsum and calcite is negligible compared to barite (Bosbach et al. 2010; Yoshida et al. 2009; Vinson et al. 2012). A low content of sulphates is not favourable for Ra<sup>2+</sup> precipitation as RaSO<sub>4</sub>. In water with dominant Cl<sup>−</sup> anion, Ra prefers to remain in solution (Beneš et al. 1982). Radium is readily and strongly adsorbed especially by the fine fraction of soils, but not in the Fe-oxide fraction. In addition, Mn solubility in the Karvinský potok is governed by the mineral phases of pyrolusite, manganite, and hausmannite that have been found supersaturated as well (average SI > 3 for manganite, and average SI > 6 for hausmannite and pyrolusite). The results of geochemical modelling lead to a conclusion that all presented supersaturated mineral phases are important for Fe, Ba, Ra and Mn removal from the discharged water in both streams.

## Conclusion

The riverine environment in the Ostrava region has been influenced by discharged mine water for decades. The mine waters studied are of Na–Cl type with increased <sup>226</sup>Ra activity. The study proved increased concentrations of Sr, Mo, Cr, Zn, Pb and Cu in river sediments and also an increased content of <sup>226</sup>Ra. Detailed field ADER mapping and conductivity measurements showed that there is a relationship between both parameters. Isotopes of Ra and Th have exhibited strong correlations with the enrichment factor of Cr, Sr and Mo in sediments of both streams supporting their relationship to the mine water. The results of geochemical modelling proved that the geochemical fate of elements in two riverine systems is governed by the precipitation reactions

forming Ca-minerals (calcite and aragonite), Fe-bearing minerals (hematite, goethite and amorphous Fe(OH)<sub>3</sub>) and Mn-bearing mineral hausmannite. Barium (Ba) was found in lower amounts in the mine water, which results in its instantaneous precipitation. The correlation of the <sup>226</sup>Ra with the enrichment factor of Sr is explained as a result of the precipitation from solution in the Karvinský potok where mine water is the main component. In the case of the Stružka stream, mine waters are diluted with surface waters, which reduces the probability of precipitation from the solution, but rather leads to the spread of contaminants further down the stream. A weak correlation of Al and K (components of silicates) with the major contaminants has proved that the sorption to clay minerals is not the governing mechanism that removes radionuclides from the water.

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**Data Availability** Data available in appendix and from the corresponding author upon request.

## Declarations

**Competing interests** The authors declare no competing interests.

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