



ENVIRONMENTAL SCIENCES DOCTORAL SCHOOL

**DEVELOPMENT AND APPLICATION OF ANALYTICAL
AND ISOTOPIC METHODS FOR THE
DETERMINATION AND MONITORING OF
POTENTIALLY TOXIC ELEMENTS IN
SOIL/WATER/SEDIMENT SYSTEMS**

PhD DISSERTATION THESIS BOOK

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Background and objectives of the work

My doctoral work was closely related to the OTKA 108558 project "Assessment of environmental mobility and temporal transformation of heavy metal contamination using sequential extraction methods", which was running from 2013 to 2017. The main objective of the work of the research group and thus of my PhD research was to further develop and refine the methodology of the BCR (3+1) step fractionation procedure, which is nowadays generally recommended in the European Union for the analysis of potentially toxic element (PTE) content in freshwater sediments, and to validate it for different matrices (soil and sediment) and PTEs and to develop and refine the procedure for the extraction of original species forms. Using these methods, we evaluated the changes in the environmental mobility of potentially toxic elements since the date of the first assessment in the lake system between Gödöllő and Isaszeg (1995-1998), an area previously investigated by us and subject to heavy metal pollution, using original and newly sampled layer sediment samples. Previously, we improved our supercritical fluid extraction (SFE) method developed for the extraction of the original species forms and performed radioanalytical measurements by γ -spectrometry. I have been actively involved in this method development work of the research group and the aims of my PhD research are summarised below:

I.) Improvement of analytical methods for speciation and fractionation of certain potentially toxic elements in sediment and soil samples; in particular, I aim to improve the various fractionation methods by reducing the shortcomings of the methods used so far, in particular the BCR sequential extraction method.

II.) To search for new fractionation methods that preserve the original species forms of the sample, to further develop a continuous flow leaching method instead of leaching methods, supported by kinetic equations (models). To this end, the optimisation of the supercritical fluid extraction (SFE) method previously developed in the department for the determination of potentially toxic element content. To evaluate the performance of the optimised technique for the extraction of the easily mobilisable element fraction.

III.) Comparison of pseudototal and fractionated element contents of lake sediment and soil samples, (geo)statistical evaluation and modelling of expected heavy metal contents for the determination of environmental mobility in relation to the EU Water Framework Directive (WFD) and human health legal requirements.

IV.) γ -radioanalytical analyses of lake sediment samples to assess the time course and variability of sedimentation.

Materials and methods

For my researches I used soil samples from Nagyhörcsök, Mosonmagyaróvár and Kecskemét, which were taken from the soil bank of the Soil Nutrient Management Group of the MATE Department of Soil Science. The soil samples from Budakalász, Imrehegy and Kiskunhalas were sampled in 2015 with an average depth of 0-30 cm. In all three areas, kitchen gardening is an agricultural activity with the use of organic manure. The river sediment sample from Vásárosnamény and the layer sediment samples from the fish ponds I.-VII. of Gödöllő-Isaszeg were collected in 2013 under the OTKA project 108558.

Determination of the pseudototal element content of potentially toxic elements

The total (pseudototal) element content was determined according to the MSZ 21470-50:2006 standard after digestion in a microwave-assisted ($\text{HNO}_3 + \text{H}_2\text{O}_2$) mixture, for which I used a CEM MARS 5 closed-chamber microwave device equipped with temperature and pressure sensors.

BCR sequence extraction

For the fractionation of the elemental content, I used the procedure recommended by the BCR (Community Bureau of Reference) (Table 1), initially according to the original protocol. Since the earlier experiments at my workplace started in 1993, they use the original protocol. The BCR recommendation is extremely time consuming (5-6 days) and the original form of each elemental species cannot be preserved in the extracts.

In order to compare the results, I performed my tests both with the old protocol and with a reference sample developed for the modified protocol (Rauret et al., 2001), so that I could subsequently evaluate the expected differences in leaching between the original and the modified protocol.

Table 1: EU BCR-recommended (3+1) step extraction method for elemental fractionation of sediments and soils (Ure et al. 1993)

Reagents applied in BCR sequential extraction steps	Chemical information
Step 1: 1.0000 g sample + 40 mL 0.11 mol dm ⁻³ HOAc	water soluble, exchangeable and carbonate-bound element content
Step 2: solid residue from Step 1 + 0.1 mol dm ⁻³ NH ₂ OH * HCl (pH = 2)	element content bound to reducible Fe/Mn oxides and hydroxides
Step 3: solid residue from Step 2 + 20 mL 8.8 mol dm ⁻³ H ₂ O ₂ (open system digestion), then + 50 mL 1 mol dm ⁻³ NH ₄ OAc (pH =2)	element content bound to organic matter and sulphides
Step (+1): microwave-assisted digestion of the residue from Step 3 with HNO ₃ /H ₂ O ₂	residual mobilizable element content (e.g. bound to silicate matrices)
Step (+2): microwave-assisted digestion of the original sample	pseudototal element content

Fractionation

At MATE (formerly SZIE) 15-20 years ago (Heltai et al., 2002; Heltai et al., 2005), a three-step continuous flow process based on CO₂ and H₂O in a supercritical extractor to preserve the as-received species was developed to complement the BCR recommendation (Table 2).

Table 2: Modified scheme for 3-step CO₂/H₂O sequential extraction

Step	Extractant	Chemical information (fraction)
1.	Supercritical CO ₂ (80 °C, 27 MPa, 60 min)	CO ₂ -soluble organic fraction and sulphides
2.	Subcritical H ₂ O (80 °C, 27 MPa, 90 min)	H ₂ O-soluble fraction
3.	Subcritical H ₂ O/CO ₂ (5%) (80 °C, 27 MPa, 90-180 min)	Fraction bound to carbonates

I reduced the size of the previously used large extraction column (from 40.40 cm³ to 22.78 cm³, a 43.61% reduction in volume), so that I could reduce the loaded sample to quartz sand ratio from about 1:20 to 1:2 sample to quartz sand ratio; which meant loading 0.5 g soil sample and 1.0 g quartz sand. I mixed 0.5000g of sample with 1.0000g of quartz sand and extracted for

2-4 h depending on the sample's organic content. I collected the extracts in 5-minute intervals, with the exception of the Nagyhörcsök sample, where I collected the extracts in 10-minute intervals.

As a final (+1) step of the SFE sequence extraction, my residues were microwave-digestion with a mixture of $\text{HNO}_3+\text{H}_2\text{O}_2$ using the same setup parameters as for the final (+1) step of the pseudototal elemental content and BCR protocol. The elemental content of aqueous and acid solutions obtained by the different procedures and of the detected residues was determined by ICP-OES.

Elemental content determination by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES)

The optimized operational parameters of the Horiba Jobin Yvon ACTIVA-M ICP-OES instrument we used were: incident power 1200 W; plasma argon gas flow 16 L/min; sheath argon gas flow 0.3 L/min; nebulizer pressure 2.89 bar; nebulizer sample uptake 0.83 mL/min; auxiliary gas flow 0.6 L/min; argon moistening; cyclon type spray chamber and Meinhard nebulizer.

The elements measured in my analyses were Ca, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn, S and V. For validation of elemental analysis measurements, I used the BCR CRM 701 reference element, in which the 3 steps of BCR fractionation were certified for Cd, Cr, Cu, Ni, Pb and Zn (USEPA 1994; Wai et al, 1997, Zemberyová et al, 2006; Zhan et al., 2014). The calibration was performed with extractant-solvent matrix matched multi-element standards (MERCK CertiPUR 1.11355.0100 ICP multi-element standard solution IV: 23 elements in dilute nitric acid) (Heltai et al., 2019).

Evaluation of column tests using first-order kinetics

For a more detailed evaluation, I ran different mathematical models. When studying the kinetic curves, I calculated with the following equation (Fit Exponential Decay/First Order):

$$y=A_1*\exp(-x/t_1)+y_o, \text{ from} \quad [1.]$$

A_1 is the "section" read on the y-axis from the starting point to the end point; t_1 is the reciprocal of the time factor ($1/t_1= k=$

kinetic constant), which multiplied by $\ln 2$ gives the half-life ($t_{1/2}$), which indicates the rate of leaching, and the y_0 factor is the asymptote. The half-life does not depend on the initial concentration of the reactant. This means that in a first order reaction the initial concentration is always halved at the same time interval.

Determination of recovered element content

The efficiency of the method and the accuracy of the results obtained are usually expressed as the sum of the elemental contents obtained with the different reagents used in the sequential extractions as a percentage of the total elemental content, described by the following equation:

$$\text{Recovery (\%)} = \left(\frac{c_{\text{watersol}} + c_{\text{red}} + c_{\text{ox}} + c_{\text{rez}}}{c_{\text{total}}} \right) \times 100 \quad [2.]$$

where: c_{watersol} , c_{red} , c_{ox} , c_{rez} és c_{total} are the water-soluble, exchangeable and carbonate-bound element content, reducible (iron)manganese oxyhydroxide-bound element content, oxidizable (e.g. bound to sulfides and organics) element content, residual fraction [mg/kg] and total element content expressed in [mg/kg] (Saleem et al., 2018).

Use of different geochemical indices to assess pollution and environmental risks

I have classified the (expected) risks of potentially toxic elements using different indices, to which I have assigned 1-1 value or category each; this will allow me to characterise my measurement results more clearly in the future. The indices were grouped into 2 categories:

- (a) indexes related to total element content and
- (b) indices related to fractional element content.

Indexes related to total element content

Contamination factor [CF]

The degree of contamination of toxic substances in a medium is described by the contamination factor (CF) (Hakanson, 1980), which is:

$$CF = \frac{c_n}{B_n}, \text{ where} \quad [3.]$$

c_n is the concentration of the element measured and B_n is the background concentration of the element measured in [mg/kg].

Enrichment Factor [EF]

A widely used metric (Buat-Menerd and Chesselt, 1979) to describe how much the concentration of a given element in a sampling medium has increased relative to its natural abundance due to anthropogenic influences.

$$EF = \frac{c_n(\text{element})/c_{\text{ref}}(\text{element})}{B_n(\text{background})/B_{\text{ref}}(\text{background})}, \text{ where} \quad [4.]$$

$c_n(\text{element})$ is the concentration of the measured element, $c_{\text{ref}}(\text{element})$ is the measured concentration of the element chosen as the reference element, $B_n(\text{background})$ is the background concentration of the measured element and $B_{\text{ref}}(\text{background})$ is the background concentration of the reference element in [mg/kg].

Geo-accumulation Factor [I_{geo}]

It was originally defined by Müller in 1969 to describe the level of contamination for potentially toxic elements in 2 micron fractions. The formula used globally is:

$$I_{\text{geo}} = \log_2 \frac{c_n}{1,5 \times B_n}, \text{ where} \quad [5.]$$

c_n is the concentration of the measured element, B_n is the background concentration of the element of interest in [mg/kg] and a constant of "1.5" is introduced to account for natural fluctuations and very small anthropogenic effects.

Pollution Load Index [PLI]

This empirical index provides a simple, comparative tool to map the extent of contamination by potentially toxic elements (Saleem et al., 2018). It describes how soils and sediments can

be/are associated with potentially toxic heavy metals that may affect their flora and fauna.

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{\frac{1}{n}}, \text{ where} \quad [6.]$$

CF_n= contamination factor obtained for sample number "n".

Indexes related to fractionated element content

Bioavailable Metal Index [BMI]

Regulatory frameworks for the characterisation and remediation of contaminated soils have moved towards a risk-based approach, taking into account the bioavailability of (potential) contaminant sources for the whole biosphere. To describe this, Rosaldo and colleagues developed the following index in 2015:

$$BMI = \left[\frac{C_{F_1}^1}{c_R^1} \times \dots \times \frac{C_{F_1}^i}{c_R^i} \times \dots \times \frac{C_{F_1}^n}{c_R^n} \right]^{\frac{1}{n}}, \text{ where} \quad [7.]$$

C_{F₁}ⁱ= easily absorbable, water-soluble element content (first step of BCR sequence extraction = F1) for element 'i'

C_Rⁱ = easily absorbable, water soluble element content for the background sample and

n = number of PTE (potentially toxic elements) tested.

Bioavailability Factor [BF]

It defines the individual physical, chemical and biological interactions that determine the exposure of flora and fauna to potentially toxic heavy metals in soil and sediments (Okbah et al., 2020).

$$BF = \frac{c_{bio}}{c_{total}}, \text{ where} \quad [8.]$$

c_{bio}= the water-soluble, easily available element content [mg/kg] and

c_{total}= total element content [mg/kg].

Individual Contamination Factor [ICF]

An important aspect is the determination of the contamination factor of the tested elements, which indicates the degree of risk of a given element for the environment in relation to its retention

time (Forghami et al., 2012), which can be described by the following equation.

$$\text{ICF} = \frac{\text{EXC} + \text{CARB} + \text{RO} + \text{OM}}{\text{RES}}, \text{ where} \quad [9.]$$

EXC = exchangeable element content

CARB = carbonate-bound element content

RO = element content bound to reducible oxides

OM = element content bound to organic substances, and

RES = residual fraction.

Risk Assessment Code [RAC%]

It determines the bioavailability of elements at different stages and their associated risks in aquatic ecosystems (Saleem et al., 2015). The risk assessment of trace elements is determined using RAC% index; which depends on the first (F1) and second (F2) steps of sequential extraction (Perin et al., 1985, Zhao et al., 2012.), developed for the old BCR sequential extraction.

$$\text{RAC} = \text{EXC}\% + \text{CARB}\%, \text{ where} \quad [10.]$$

EXC% = exchangeable element content (F1) and

CARB% = carbonate-bound element content (F2).

A modified (3+1 step) BCR protocol, due to $\text{RAC} = \text{F1}(\%)$

Radioanalytical analysis by γ -spectroscopy

Radioanalytical analysis of sediment samples was performed by γ -spectroscopy. Dried and homogenized samples were loaded into a 38 cm³ airtight plastic box similar geometry to Marinelli containers and a solid-state detector (Canberra HPGe) with a 1333 keV half-width detector connected to a multichannel analyzer was used. The activity of radionuclides was determined in [Bq/kg] units. In my thesis I compared the activity of ¹³⁷Cs of Chernobyl origin in each layer (Heltai et al. 2018.)

Results and discussion

Determination of pseudototal element content in sediment samples

Sediments from lakes I (3 sites) and VII (4 sites) of the Gödöllő-Isaszeg lake system were sampled again in 2013. The stratigraphic samples were analysed in 10 cm intervals, from which, after microwave digestion according to MSZ 21470-50:1998, the concentrations of 11 elements were determined by ICP-OES, as follows: Cadmium (Cd), Cobalt (Co), Chromium "total" (Cr), Copper (Cu), Iron (Fe), Manganese (Mn), Nickel (Ni), Lead (Pb), Zinc (Zn), Sulphur (S) and Vanadium (V).

In evaluating my results, I examined how the elements behaved relative to each other in the 10 cm divisions of a given section, and how they behaved relative to each other in a given vertical line, which I also compared with the results from the 1996 sampling year. Research from previous years has shown that PTE accumulation of communal origin is observed in Lake I and industrial origin in Lake VII, which are also supported by the results I have presented. As it is well known, in the case of Lake I, the treated wastewater of the city of Gödöllő was discharged until 1995, while in Lake VII, the wastewater of the adjacent engineering works was discharged.

The concentration of Cd exceeds the limit values in the whole longitudinal sections 1, 2 and 3 of Lake I. In the case of Lake VII, the limit values are exceeded at the upper 50 cm at point 9, at the upper 20 cm at point 10, at the upper 20 cm at point 11 and at the upper 10 cm at point 12, mainly for chromium and zinc.

In sediment core samples from Lake VII in 2013, a downward trend in PTE concentrations was observed for most elements, with the exception of vanadium and zinc. The concentrations of Cd, Co, Cr, Cu, Ni, Pb and Zn in the deeper layers decreased significantly in 2013, but are still very high in the upper 0-10 cm layer, in some cases exceeding the contamination limits.

Concentrations of Fe and Mn increased significantly in all layers in 2013, probably due to oxidative conditions during the 2003 sediment removal operation. In general, the removal of PTE contamination from Lake VII was incomplete in 2003.

Sequential extraction according to BCR protocol

The certified sediment sample I use, BCR CRM-701, is certified for 6 elements, namely cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn). It is also important to mention here that the results obtained in steps (+1) and (+2) are only indicative.

In fraction 1 of the BCR, the proportion of PTEs decreased significantly, which can be attributed to the fact that during the sediment removal in 2003, the most mobile fraction was transported further with this runoff water.

The proportion of the BCR 2 fraction increased significantly for several elements, which can be explained by the formation of insoluble iron and manganese oxides, as the upper layer of the sediment was in direct contact with the air.

The conclusion I made when evaluating the results of the pseudototal element content is also correct here, that the removal of PTE contamination from Lake VII was not complete in 2003.

Supercritical fluid extraction (SFE) method development and kinetic study of Step 3.

My kinetic studies were based on an earlier method development by Halász (2010), in which the previously used column construction was scaled down and a 0.5000g sample loading was applied in addition to the blank sample.

The samples used were:

- ✧ analytical grade quartz sand (VWR)
- ✧ soil from Nagyhörcsök
- ✧ soil from Kecskemét
- ✧ soil from Mosonmagyaróvár

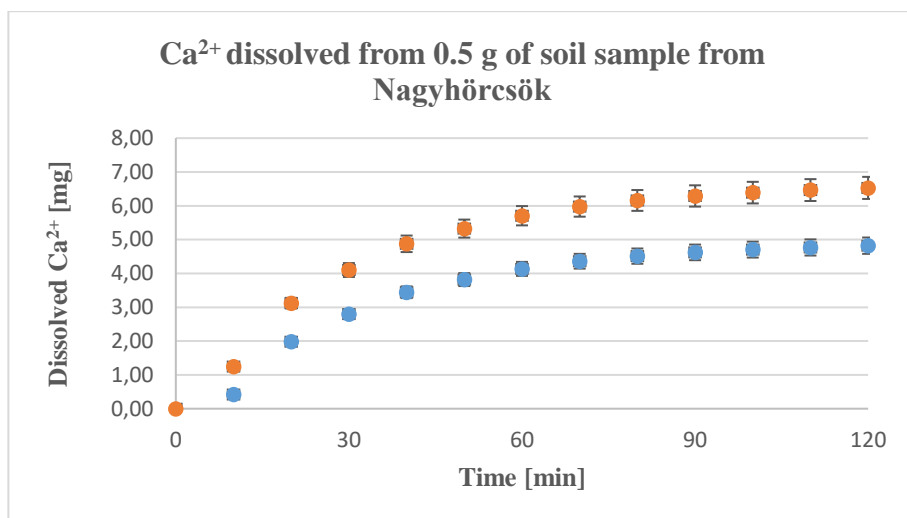


Figure 1: Ca²⁺ leaching from the soil sample in Nagyhörsök (blue points: 2011, red points: 2015)

Comparing the concentration of the corrected Ca²⁺ ions as a function of extraction time and total Ca²⁺ concentration of the samples (Figure 1); it can be said that the new column extracted fractions, based on their combined concentrations, resulted in 17.81% more Ca²⁺ extracted in the case of the Nagyhörsök sample, which is illustrated in Table 3. From the graphical representation of saturation times, it can be said that 6.55 mg Ca²⁺ could be extracted in 120 min for the Nagyhörsök sample.

Table 3: Rate of Ca²⁺ extraction from the Nagyhörsök sample

Nagyhörsök			
	Extraction time [min]	Dissolved ΣCa^{2+} [mg]	Corrected dissolved Ca ²⁺ [mg]
Old column	120	8,54	4,82
New column	120	10,25	6,55

It can be said that by reducing the size of the extraction column, we have found a solution to a significant extraction problem, such as successfully reducing the interference from the blank value.

Methodological comparisons, determination of the recoverable element content in sediment layers

The following two graphs (Figure 2. and Figure 3.) illustrate the distribution of PTE concentrations between the sediment

reference sample BCR-701 and the sediment sample from the Gödöllő-Isaszeg section of Lake VII/10 between SFE (sum of elemental contents extracted with subcritical H₂O and subcritical H₂O/CO₂), BCR (step 1 and sum of step 4) and microwave digestion. It is clearly visible in the two graphs that the elements that can be easily mobilized; copper, nickel; but most of all, the elemental content of zinc stands out during the SFE process.

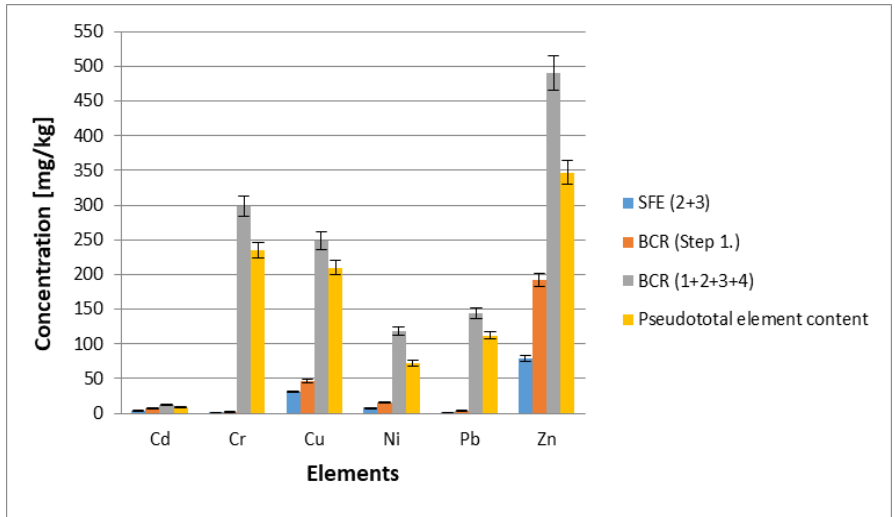


Figure 2: Comparison of BCR-701 reference sample values (with 5% error range)

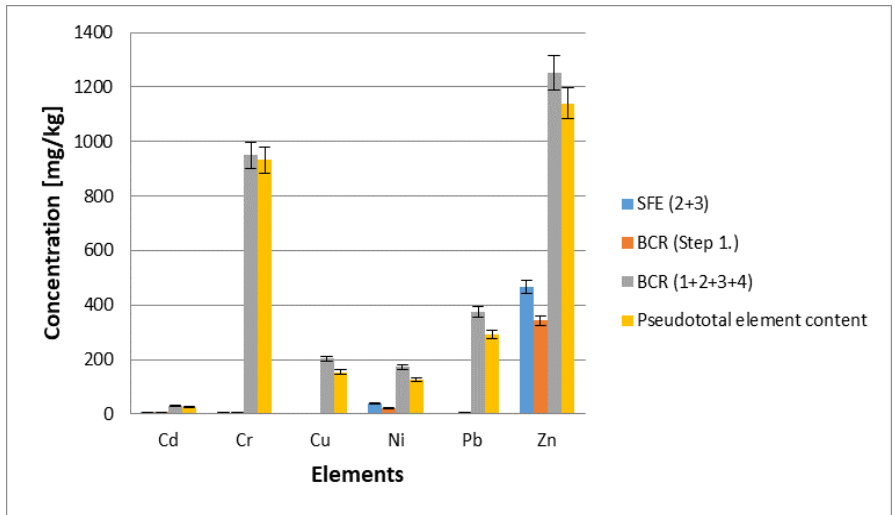


Figure 3: Comparison of sediment sample values (0-10cm) from Lake Gödöllő-Isaszeg V (with 5% error range)

I compared the fractionation results obtained with the BCR sequential extraction method and the improved SFE method using the BCR CRM-701 sample and the sediment sample from fish pond VII of the Gödöllő-Isaszeg lake system.

The difference between the measured and validated values for the reference sample BCR CRM-701 shows in few cases a significant difference at the $p=5.0\%$ level, these differences are not significant at the $p=0.1\%$ level. It can be concluded that the measured and validated concentrations for BCR CRM-701 show an acceptable agreement, i.e. the BCR sequence extraction was reliably applied. Based on these results, it can be concluded that the fractionation results obtained for the Gödöllő-Isaszeg sediment sample are also reliable.

A comparison of the fractionation results of the BCR CRM-701 and the Lake Gödöllő-Isaszeg sediment samples shows that both sediment samples contain significant amounts of PTE in all fractions and that the distribution of the elements between the fractions reflects the origin of the contamination. It can be seen that the most mobilisable Fraction 1, with the exception of copper, contains PTEs of the same magnitude in both samples. The BCR Step 1 solubilizes similar, but not exactly the same amount of PTEs as the summed three SFE steps, i.e. our SFE procedure most accurately characterizes the easily mobilizable PTE fractions.

Based on previous experience (Heltai et al. 2002), there were order of magnitude differences between the two methods for Cd, Pb, and Zn, which we were able to eliminate by increasing the extraction times. The mobile element fraction is characterized by the acetic acid leaching alone according to the BCR recommendation, whereas the SFE procedure separates it into two fractions, water-soluble and carbonate-bound fractions

From the ratio $(\text{BCR } 3)/(\text{CO}_2)$, we can infer organic matter of apolar nature and possibly elemental sulphur. Comparing the samples, in both cases zinc and cadmium are more easily mobilised than the other elements. In the case of lake VII, sample 10, the high ratio suggests that a significant proportion of nickel, lead and cadmium are bound to organic matter.

Geochemical characterisation of sediment samples from fish ponds by sediment layers, risk assessment for mobilisation of PTEs

The values of the geochemical indices are assigned a category, which usually (depending on the index) have values between 1-5/1-6/1-7. I have chosen Fe as the reference element, which is recommended by several literatures. I calculated indices for cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), manganese (Mn), nickel (Ni), lead (Pb) and zinc (Zn).

The geochemical indices I used compare very well with the pseudototal and sequential extraction results of the PTEs, as well as with the background sedimentation system of the MSZ and EPA standards. I think it is important to note that while $\text{HNO}_3+\text{H}_2\text{O}_2$ MW digestion is preferred in environmental analysis, „aqua regia” exploration is preferred in environmental geochemistry.

In the literature, there is no mandatory matrix for the evaluation of geochemical indices, so there may be minor differences. In the following I present my results for 8 geochemical indices.

Evaluation of indices related to total element content

Contamination factor [CF]

As for cadmium CF, very high values were obtained for the Lake I/1-3, which is also true for the Lake VII/10, 11. Low levels of contamination are observed for cobalt, manganese and nickel, although for nickel there are already moderate levels of contamination in 6 layers. For chromium, copper, lead and zinc, there are significant levels and/or very high levels of contamination in some layers, which are characteristic of the upper 30 cm layer thickness.

Pollution Load Index [PLI]

The CF index is usually evaluated together with the PLI. This PLI value above 1 pollution exists. Lake I is 0-20 cm in the 1st site, 0-40 cm in the 2nd site and 50-60 cm, while 0-10 cm in the 3rd site the possibility of contamination was confirmed in its depth. The possibility of pollution was confirmed in the upper 40 cm in the 9th extension of the Lake VII, in the upper 10 cm in the

10th site and in the 0-20 cm depth of the 11th site. The PLI index of part of 12 does not indicate pollution.

Enrichment Factor [EF]

For cadmium, enrichment ranges from significant to extremely high levels. Only significant levels of enrichment are found in the deeper parts of the Lake VII/ 10-12. For cobalt, manganese and nickel elements, enrichment is minimal, sometimes moderate. For chromium, nickel, lead and zinc, however, moderate to significant or significant to very high enrichments are observed in the upper 30 cm layers.

In the upper 10 cm of the Lake VII/10 and 11, the EF value for lead is extremely high, while for zinc, extremely high EF values were determined in the 10-20 cm of the Lake VII/9 and in the 0-10 cm of the Lake VII/11.

Geo-accumulation Factor [I_{geo}]

Only for cadmium I obtained extreme levels of contamination, detectable up to 40 cm, which is characteristic of samples from Lake I/2 and Lake VII/9. For cobalt and manganese there is no contamination, while for nickel there is also no contamination except for 4 layers. The other 4 elements generally show low to moderate levels of contamination in the upper 30 cm, and in some cases 40 cm; however, in the 0-10 cm of the Lake VII/10, severe levels of contamination were detected for lead and zinc, while in Lake VII/11, heavy and high levels of contamination were detected for chromium, lead and zinc.

Evaluation of indices related to fractionated element content

Bioavailable Metal Index - [BMI]

The BMI is calculated by comparing the concentrations obtained in step 1 of the BCR with the concentrations in the bedrock, taking into account the number of PTEs measured, and thus giving the following results: 0.14 for 0-10 cm; 0.1 for 0-20 cm; 0.04 for 20-30 cm; 0.03 for 30-40 cm; 0.01 for 40-50 cm; 0.04 for 50-60 cm and 0.03 for 60-70 cm. Based on these results, I conclude that the BCR of the PTEs relative to the background according to Step 1 resulted in very low indices.

Bioavailability Factor [BF], Individual Contamination Factor [ICF] and the Risk Assessment Code RAC%

There is no category system for BF, because it is essentially a ratio. The ICF index can be characterized by a system of 4 categories: low, moderate, moderate and high, while the RAC% is classified as: 1- none, 2- low, 3- moderate, 4- high and 5- very high risk. BF is the highest for cadmium, i.e. 1.

Chromium is high in the top 10 cm, cobalt in the 50-70 cm, no significant variation is observed for copper, iron is prominent in the 0-10 cm and 50-70 cm, manganese is above 0.50 in the whole layer, nickel is very high in the top 10 cm but more significant in the 10-70 cm, lead is above 0.89 in the total layers and zinc is high in the 0-10 cm.

Chromium is high in 0-10 cm and then low. Cadmium is high throughout the whole length, cobalt is low and moderate, copper is low throughout the whole length, iron is low between 0-60 cm and moderate CF values are found in 60-70 cm.

Manganese is moderately high between 40-70 cm and nickel is high in the upper 10 cm. Lead is high up to 60 cm and low up to 70 cm. Finally, zinc is high between 0-10 cm, moderate in 10-20 cm and low in the deeper layers.

For RAC, the values for each metal are as follows: chromium is low between 20-70 cm, cadmium is moderate between 0-10 cm and low between 10-20 cm, cobalt is moderate between 0-10 cm and high between 60-70 cm, copper and iron are low risk in the whole section. Manganese is medium in 50-60 cm but high in the other layers. The RAC values for nickel, lead and zinc in the layers are low to medium risk.

Radioanalytical results using γ -spectroscopy

The γ -spectrometric measurement of the samples collected in 2013 was carried out in 2016. The nuclide activity of ^{137}Cs at sampling point 1 of Lake I: 0-33.74 Bq/kg, at sampling point 2: 0-179.26 Bq/kg and at sampling point 3: 0-55.22 Bq/kg; while for the 4 sampling points of Lake VII: 0-88.11 Bq/kg, 2.37-56.24 Bq/kg, 0-148.91 Bq/kg and 0-40.91 Bq/kg are characterized.

In 2018 (Heltai et al. 2018), our group published these results (for radionuclides of Chernobyl origin) in comparison with the 1996 measurements for the Lake I/1 and Lake VII/10 sections, and the following conclusions were reached. The concentrations measured in 1996 have been corrected to 2016 to take into account the decay rate, and a decrease in activity concentrations by one

order of magnitude was observed in 2013 at both sites, which is not due to radioactive decay only. This decrease is explained by the leaching and transport of ^{137}Cs by water flow through the lakes. The vertical distribution of PTEs was similar to that of ^{137}Cs , indicating that their accumulation intensified in the period 1986-1996.

Conclusions and recommendations

The fraction of contaminants that are currently bioavailable or soluble under different conditions in the soil-sediment-water system can be estimated from the possible physico-chemical interactions. The fractionation methods developed for this purpose are based on the sequential or individual application of solvents modelling different environmental conditions.

The primary purpose of the risk assessment is the fractionation that characterizes the environmental mobility of the outgoing PTEs in the short and long term. The methods used so far for this purpose need to be further developed, since until now they have mainly been used for the examination of aquatic sediments. For the validation of BCR fractionation, the sediment sample BCR 701, certified for six elements (Cd, Cu, Cr, Ni, Pb, Zn), was available. In order to extend the validation to soils and other sediment types and to increase the number of contaminants that could be tested, it was necessary to optimise the ICP-OES method and to minimise the chemical and physical matrix effects that hamper the calibration.

The ICP-OES elemental analysis, based on matrix calibration and the use of an internal yttrium standard, allowing flexible element selection, could be successfully applied to the determination of pseudotal and BCR fractionated PTE in lake and river sediments and in different types of soils. I was also able to validate the measurements for the different sample types (sediment and soil) by examining the BCR 701 CRM samples matched to the respective measurement series and by comparing the BCR fractionation residues with the pseudototal element concentrations for the fractionation of the unproven elements.

Improved SFE method using supercritical carbon dioxide, subcritical water and their mixture as solvents has been shown to be suitable for the analysis of easily mobilizable elemental content in environmental samples. The development was implemented with a reduced column size compared to previous ones, which gave very favourable results in the kinetic study. The information content of the SFE differs from the BCR protocol. Step 1 of the BCR recommendation, using acetic acid leaching, provides an aggregate characterization of the fraction that can be mobilized by water solubilization and hydrogen carbonate formation. In contrast, the SFE procedure breaks this down into two fractions; a water-soluble fraction and a carbonate-bound fraction. Its time

demand is a fraction of that of the BCR protocol. Another advantage is that it is possible to search and identify species in their original state from the extracts.

The main question of the geochemical assessments carried out at the different sample sites was whether the detected element concentrations were from natural sources or influenced by anthropogenic or in situ emissions and/or activities, taking into account the geochemical mechanisms regulating the concentrations in the sediment. In turn, the individual geochemical indices helped to understand the relationship between the elemental contents of pseudototal, sequentially extracted fractions and the properties of the parent rock. Other tasks to be carried out include extending the mathematical models and determining their missing parameters based on soil analyses (e.g. pore volume), which can be used to run the already appropriate models.

The radioanalytical investigations have allowed the interpretation of the temporal variation and environmental mobility of sedimentation and PTE contamination in the lake system between Gödöllő and Isaszeg. The ^{137}Cs concentrations measured in 1996 were corrected for the decay rate in 2016. A decrease in activity concentrations by an order of magnitude was observed in 2013 at both sites, which is not attributable to radioactive decay. This decrease can be explained by leaching and transport by water flow through the lakes. The vertical distribution of PTEs was similar to that of ^{137}Cs , indicating that the temporal variation of sedimentation was more intensive directly active the Chernobyl accident during the period 1986-1996.

New scientific results

1. I have successfully applied the flexible multi-element ICP-OES method, also developed with the collaboration of our group, to determine the pseudototal and BCR-fractionated PTE content of different sample types (sediments, soils). The application of the ICP-OES method with matrix-matched calibration with BCR extraction solvents and an internal standard of yttrium allowed a broader validation of the BCR fractionation method than previously possible for other sample types and elements using the BCR 701 CRM, previously certified for the fractionation of only lake sediments and six PTE (Cd, Cr, Cu, Ni, Pb, Zn), for other sample types and elements. For the additional elements included in the study, the total elemental contents leached in the BCR fractions and the pseudototal elemental contents showed acceptable agreement.

2. Improved SFE-extraction using supercritical carbon dioxide, subcritical water and their mixture as solvent has been shown to be suitable for the determination of water-soluble and carbonate-bound elemental content in different environmental samples (soil, sediment) separately, thus allowing the search and identification of as-prepared species from these extracts. The advantage of my method is that more detailed information on the elemental content bound to carbonates and bicarbonates can be obtained. The extraction times are optimised depending on the different carbonate contents of different types of samples. The optimisation of the method was achieved by a full kinetic study of different samples. Due to the complex matrix components and the sometimes high Ca content of the sample, the optimization of the ICP-OES measurement parameters (increase of plasma power, adjustment of gas flow) was necessary.

3. During the 2013-2016 repetition of the sediment pollution survey of the lake system between Gödöllő and Isaszeg, I established the following in 1996:

a. It can be said that the exceptionally high element concentration appears primarily in the upper 30 cm of the sites (with a few exceptions); however, what is even more worrying is that I found element concentrations above the limit in many samples.

b. In the deeper layers, the concentration of Cd, Co, Cr, Cu, Ni, Pb and Zn decreased significantly in 2013, but it is still very high in the upper 0-10 cm layer, which in some cases even exceeds the contamination limits.

c. I showed that as a result of the 2003 sediment removal operation, the concentration of iron and manganese increased in all layers as a result of the oxidative conditions. As a result of the pollutant transported further by the runoff, the BCR decreased in the 1st fraction; however, it increased in 2nd fraction, which is a consequence of the sediment removal operations that took place in the meantime.

d. In 1995, during γ -spectrometry tests, the accumulation of the ^{137}Cs isotope from the 1986 Chernobyl reactor accident was clearly detected in the sediment layer of the lakes in the upper 30-40 cm thick layer of the sediment. During the evaluation of the repeated sampling in 2013, the activity concentration showed a similar depth distribution, but the decrease in activity was greater than the value calculated from the rate of radioactive decay.

4. I used different geochemical indices to characterise the different sampling areas and show the relationships between pseudototal and sequentially extracted elemental content.

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