Innovative Sample Preparation Method Using a Conductively Heated Digestion System for Trace Element Analysis of Radioactive Oil Sludge by ICP-OES

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Electronic supplementary information

Validation of analytical procedure

The method developed was validated according to the parameters outlined in the DOQ-CGCRE-008 document (ref. 25), issued by the Brazilian National Institute of Metrology, Quality and Technology, INMETRO (*Instituto Nacional de Metrologia, Qualidade e Tecnologia*, in Portuguese).

The Horwitz equation is used to assess the acceptability and precision characteristics of the proposed method, using HORRAT (HO_R) values. The HO_R values are obtained using the following equation (S1):

$$HO_{R} = \frac{DPR_{E}}{DPR_{H}}$$
 (S1)

 DPR_E represents the relative standard deviation obtained experimentally and DPR_H the relative standard deviation obtained from the Horwitz equation, at the concentration of interest (ref. 25).

To assess the accuracy of the method, the z-score is applied, obtained using the following equation (S2):

Z

$$=\frac{x_{lab} - x_{v}}{s}$$
 (S2)

 x_{lab} is the value obtained experimentally, x_v is the value accepted as real and s is the standard deviation of the proficiency test.

The relative error (RE%) enables to evaluate the accuracy of the method. It is obtained using the following equation (S3):

$$RE\% = \frac{X_{det} - X_{cert}}{X_{cert}} \times 100$$
(S3)

 X_{det} is the arithmetic mean of the values obtained experimentally and X_{cert} is the value of the certified material.

The relative standard deviation (RSD%) allows for the assessment of the precision of the proposed method. It is calculated using the following equation (4):

$$RSD\% = \frac{\sigma}{x_D} \times 100$$
 (S4)

 σ represents the standard deviation and x_D the average of the values obtained experimentally.

The linear working range is the interval in which the results of the method are proportional to the concentration of the analyte. This criterion was applied when constructing the calibration curve for quantifying the analytes of interest in the samples. Linearity is checked according to the following equation (S5):

$$y = a + bx$$
 (S5)

y is the measured response (intensity), x is the concentration, a is the linear coefficient and b is the angular coefficient (sensitivity).

The limit of detection (LD) corresponds to the smallest amount of analyte that can be detected, but not necessarily quantified. For the proposed methodology, the LD was calculated using the following equation (S6):

$$LD = 3.3 \frac{s}{b}$$
 (S6)

s is the standard deviation of the blank response and b is the angular coefficient of the analytical curve.

The limit of quantification (LQ) consists of the smallest amount of the analyte that can be effectively quantified with acceptable precision and accuracy. For the proposed methodology, the LQ was calculated using the following equation (S7):

$$LQ = 10 \frac{s}{b}$$
 (S7)

s is the standard deviation of the blank response and *b* is the angular coefficient of the analytical curve.

The recovery test on the NIST 2709 reference material was carried out according to the guidelines of the certificate (ref. 24). The sample preparation method was applied to the certified reference material (CRM) and the recovery was calculated according to the following equation (S8):

Recovery (%) =
$$\left(\frac{\text{median of the range}}{\text{certified concentration value}}\right) \times 100$$
 (58)

The median is calculated based on the range of leaching concentrations and the certified concentration value corresponds to the value specified on the CRM certificate.

To calculate the uncertainty of a measurement, the combined standard uncertainty calculation is applied (ref. 30). In this method, the sources of uncertainty that affect the measurement are considered. After identifying each source of uncertainty and determining the standard uncertainty value of each source (mass, volume, etc.), the combined standard uncertainty is obtained using the following equation (S9):

$$u_{\rm C} = 2 \sqrt{u_1^2 + u_2^2 + \ldots + u_n^2}$$
 (S9)

Where u_c is the combined standard uncertainty and u_1, u_2 and u_n the standard uncertainty of each source of uncertainty that impacts on the measurement.



Figure S1: Oil sludge sample before CHDS (a). Oil sludge sample after CHDS in a closed vessel (b).



Figure S2: Vessels closed and sealed with the samples (a). Vert Simplify digester containing the vessels (b).

Concentration of elements								
	Ni	Ni	Zn	Zn	Cu	Cu		
Sample	Average	Composite	Average	Composite	Average	Composite		
	Aliquots	Sample	Aliquots	Sample	Aliquots	Sample		
1	1.11 ± 0.06	1.07 ± 0.05	1.16 ± 0.06	1.64 ± 0.08	0.50 ± 0.03	0.71 ± 0.04		
1	(mg kg ⁻¹)							
	0.46 ± 0.02	0.48 ± 0.02	2.56 ± 0.13	3.09 ± 0.15	0.32 ± 0.02	0.52 ± 0.03		
Z	(mg kg ⁻¹)							
	0.32 ± 0.02	0.29 ± 0.01	1.57 ± 0.08	1.94 ± 0.10	< 2.00	< 2.00		
5	(mg kg ⁻¹)	(mg kg⁻¹)	(mg kg⁻¹)	(mg kg ⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)		
1	0.76 ± 0.04	0.76 ± 0.04	3.09 ± 0.16	3.59 ± 0.18	0.61 ± 0.03	0.79 ± 0.04		
4	(mg kg ⁻¹)							
	< 4.00	< 4.00	< 2.00	< 2.00	< 2.00	< 2.00		
5	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)					
6	0.64 ± 0.03	0.65 ± 0.03	1.48 ± 0.07	2.03 ± 0.10	< 2.00	< 2.00		
U	(mg kg ⁻¹)	(µg kg⁻¹)	(µg kg-1)					
7	0.64 ± 0.03	0.63 ± 0.03	6.21 ± 0.31	6.75 ± 0.34	1.73 ± 0.09	1.87 ± 0.09		
/	(mg kg ⁻¹)	(mg kg⁻¹)	(mg kg⁻¹)	(mg kg ⁻¹)	(mg kg⁻¹)	(mg kg ⁻¹)		
0	20.14 ± 1.01	20.04 ± 1.00	36.86 ± 1.84	38.14 ± 1.91	14.90 ± 0.74	12.98 ± 0.65		
0	(mg kg ⁻¹)							
0	1.25 ± 0.06	1.24 ± 0.06	9.20 ± 0.46	9.63 ± 0.48	1.08 ± 0.05	1.21 ± 0.06		
9	(mg kg ⁻¹)							
10	< 4.00	< 4.00	16.17 ± 0.81	13.07 ± 0.66	1.96 ± 0.10	2.04 ± 0.10		
10	(µg kg-1)	(µg kg⁻¹)	(mg kg ⁻¹)					
11	1.02 ± 0.05	1.01 ± 0.05	13.78 ± 0.69	14.29 ± 0.71	0.77 ± 0.04	0.93 ± 0.05		
11	(mg kg ⁻¹)							
	Pb	Pb	Cr	Cr	Mn	Mn		
Sample	Average	Composite	Average	Composite	Average	Composite		
	Aliquots	Sample	Aliquots	Sample	Aliquots	Sample		
1	< 8.00	< 8.00	5.01 ± 0.25	5.26 ± 0.26	1.76 ± 0.09	1.91 ± 0.10		
	(µg kg⁻¹)	(µg kg-1)	(mg kg ⁻¹)					
2	< 8.00	< 8.00	0.98 ± 0.05	1.30 ± 0.06	18.52 ± 0.93	18.03 ± 0.90		
	(µg kg⁻¹)	(µg kg ⁻¹)	(mg kg ⁻¹)					
з	< 8.00	< 8.00	0.78 ± 0.04	1.06 ± 0.05	1.04 ± 0.05	1.04 ± 0.05		
	(µg kg ⁻¹)	(µg kg-1)	(mg kg ⁻¹)	(mg kg ⁻¹)	(mg kg⁻¹)	(mg kg ⁻¹)		
4	< 8.00	< 8.00	3.35 ± 0.17	3.53 ± 0.18	3.40 ± 0.17	3.37 ± 0.17		
	(µg kg⁻¹)	(µg kg-1)	(mg kg ⁻¹)	(mg kg ⁻¹)	(mg kg⁻¹)	(mg kg ⁻¹)		
5	< 8.00	< 8.00	2.22 ± 0.11	2.51 ± 0.13	12.52 ± 0.63	12.01 ± 0.60		
	(µg kg ⁻¹)	(µg kg⁻¹)	(mg kg ⁻¹)	(mg kg ⁻¹)	(mg kg⁻¹)	(mg kg ⁻¹)		
6	< 8.00	< 8.00	4.63 ± 0.23	4.60 ± 0.23	0.89 ± 0.04	0.91 ± 0.05		
	(µg kg ⁻¹)	(µg kg⁻¹)	(mg kg ⁻¹)					
7	< 8.00	< 8.00	2.51 ± 0.13	2.77 ± 0.14	3.23 ± 0.16	3.21 ± 0.16		
,	(µg kg ⁻¹)	(µg kg ⁻¹)	(mg kg ⁻¹)					
8	2.42 ± 0.12	2.87 ± 0.14	8.36 ± 0.42	8.51 ± 0.43	3.44 ± 0.17	3.43 ± 0.17		
	(mg kg ⁻¹)							
9	< 8.00	< 8.00	3.50 ± 0.18	3.83 ± 0.19	3.58 ± 0.18	3.57 ± 0.18		

Table S1: Results of the metals analysis of samples 1, 2, 3, 4, 5, 6, 7, 8, 9, 10 and 11, obtained by comparing the averages of the aliquots with the composite sample of the aliquots.

	(µg kg-1)	(µg kg-1)	(mg kg-1)	(mg kg-1)	(mg kg-1)	(mg kg-1)
10	< 8.00	< 8.00	0.70 ± 0.04	0.97 ± 0.05	3.10 ± 0.16	2.99 ± 0.15
10	(µg kg-1)	(µg kg⁻¹)	(mg kg-1)	(mg kg-1)	(mg kg ⁻¹)	(mg kg-1)
11	< 8.00	< 8.00	1.84 ± 0.09	2.11 ± 0.11	4.82 ± 0.24	4.78 ± 0.24
11	(µg kg⁻¹)	(µg kg⁻¹)	(mg kg-1)	(mg kg-1)	(mg kg-1)	(mg kg-1)

Table S2: Results of the metals analysis of sample 1, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

Sample 1								
Element	SD	RSD%	RE%	HO _R	Z-score			
Ni	0.02	1.80	4.30	0.11	0.23			
Zn	3.00E-03	0.29	28.99	0.02	13.84			
Cu	1.00E-03	0.25	28.98	0.02	16.50			
Pb	-	-	-	-	-			
Cr	0,08	1.56	4.77	0.10	0.32			
Mn	8.00E-03	0.45	7.86	0.03	1.91			

Table S3: Results of the metals analysis of sample 2, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 2								
Element	SD	RSD%	RE%	HO _R	Z-score				
Ni	6.80E-04	0.15	2.73	0.01	1.91				
Zn	5.05E-03	0.20	16.42	0.01	10.03				
Cu	4.40E-04	0.14	38.91	0.01	45.63				
Pb	-	-	-	-	-				
Cr	1.81E-03	0.18	24.18	0.01	17.34				
Mn	0.04	0.24	2.71	0.02	1.08				

Table S4: Results of the metals analysis of sample 3, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

Sample 3								
Element	SD	RSD%	RE%	HO _R	Z-score			
Ni	3.30E-03	1.04	7.76	0.07	0.69			
Zn	1.84E-02	1.17	19.17	0.07	2.03			
Cu	-	-	-	-	-			
Pb	-	-	-	-	-			
Cr	0.02	2.78	26.03	0.17	1.27			
Mn	7.73E-03	0.74	0.36	0.05	0.05			

Table S5: Results of the metals analysis of sample 4, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

Sample 4									
Element	SD	RSD%	RE%	HO _R	Z-score				
Ni	1.73E-03	0.23	0.36	0.01	0.16				
Zn	0.04	1.20	14.09	0.07	1.37				
Cu	2.01E-03	0.33	23.66	0.02	9.35				

Pb	-	-	-	-	-
Cr	0.05	1.46	5.12	0.09	0.37
Mn	0.02	0.60	0.89	0.04	0.15

Table S6: Results of the metals analysis of sample 5, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 5								
Element	SD	RSD%	RE%	HO _R	Z-score				
Ni	-	-	-	-	-				
Zn	-	-	-	-	-				
Cu	-	-	-	-	-				
Pb	-	-	-	-	-				
Cr	0.01	1.80	11.52	0.11	2.07				
Mn	0.06	1.73	4.25	0.11	0.82				

Table S7: Results of the metals analysis of sample 6, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 6								
Element	SD	RSD%	RE%	HO _R	Z-score				
Ni	3.68E-03	0.58	1.70	0.00	0.30				
Zn	0.02	1.53	27.19	0.00	2.44				
Cu	-	-	-	-	-				
Pb	-	-	-	-	-				
Cr	0.07	1.56	0.60	0.00	0.04				
Mn	1.05E-03	0.12	1.95	0.00	1.70				

Table S8: Results of the metals analysis of sample 7, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 7								
Element	SD	RSD%	RE%	HO _R	Z-score				
Ni	1.26E-03	0.20	1.09	0.01	0.54				
Zn	4.92E-03	0.08	8.05	0.00	11.04				
Cu	2.99E-03	0.17	7.71	0.01	4.82				
Pb	-	-	-	-	-				
Cr	4.32E-03	0.17	9.20	0.01	5.90				
Mn	4.43E-03	0.14	0.61	0.01	0.45				

Table S9: Results of the metals analysis of sample 8, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

Sample 8										
Element	Element SD RSD% RE% HO _R Z-score									
Ni	0.12	0.61	0.51	0.04	0.08					
Zn	0.03	0.10	3.36	0.01	3.62					

Cu	0.03	0.24	14.76	0.01	2.66
Pb	0.02	0.72	15.62	0.04	2.59
Cr	0.03	0.33	1.76	0.02	0.54
Mn	7.82E-03	0.23	0.49	0.01	0.21

Table S10: Results of the metals analysis of sample 9, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 9							
Element	SD	RSD%	RE%	HO _R	Z-score			
Ni	1.19E-03	0.10	0.90	0.01	0.94			
Zn	2.06E-02	0.22	4.55	0.01	2.12			
Cu	3.18E-03	0.30	11.50	0.02	4.39			
Pb	-	-	-	-	-			
Cr	0.03	0.98	8.56	0.06	0.95			
Mn	1.51E-02	0.42	0.27	0.03	0.06			

Table S11: Results of the metals analysis of sample 10, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

Sample 10						
Element	SD	RSD%	RE%	HO _R	Z-score	
Ni	-	-	-	-	-	
Zn	0.04	0.22	23.69	0.01	0.51	
Cu	0.02	0.96	3.77	0.06	0.41	
Pb	-	-	-	-	-	
Cr	4.65E-03	0.66	27.94	0.04	5.85	
Mn	0.01	0.35	3.69	0.02	1.03	

Table S12: Results of the metals analysis of sample 11, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

Sample 11						
Element	SD	RSD%	RE%	HO _R	Z-score	
Ni	3.30E-03	0.33	0.11	0.02	0.03	
Zn	0.05	0.33	3.55	0.02	1.09	
Cu	6.19E-03	0.81	17.90	0.05	2.70	
Pb	-	-	-	-	-	
Cr	8.68E-03	0.47	12.82	0.03	3.11	
Mn	1.35E-02	0.28	0.87	0.02	0.31	

	Concentration of elements									
	Er	Er	Eu	Eu	Lu	Lu	Nd	Nd	Sm	Sm
Sample	Average	Composite	Average	Composite	Average	Composite	Average	Composite	Average	Composite
	Aliquots	Sample	Aliquots	Sample	Aliquots	Sample	Aliquots	Sample	Aliquots	Sample
1	< 4.64	< 4.64	< 1.09	< 1.09	< 0.60	< 0.60	< 1.47	< 1.47	< 5.42	< 5.42
	(µg kg-1)	(µg kg-1)	(µg kg⁻¹)	(µg kg-1)	(µg kg-1)	(µg kg⁻¹)	(µg kg-1)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)
2	0.43 ± 0.02	0.41 ± 0.02	< 1.09	< 1.09	< 0.60	< 0.60	2.00 ± 0.11	1.90 ± 0.10	0.60 ± 0.03	0.45 ± 0.02
	(mg kg ⁻¹)	(mg kg-1)	(µg kg-1)	(µg kg⁻¹)	(µg kg-1)	(µg kg-1)	(mg kg ⁻¹)	(mg kg-1)	(mg kg-1)	(mg kg ⁻¹)
3	< 4.64	< 4.64	< 1.09	< 1.09	< 0.60	< 0.60	0.57 ± 0.03	0.50 ± 0.01	< 5.42	< 5.42
	(µg kg-1)	(µg kg-1)	(µg kg⁻¹)	(µg kg-1)	(µg kg-1)	(µg kg-1)	(mg kg ⁻¹)	(mg kg ⁻¹)	(µg kg-1)	(µg kg-1)
4	< 4.64	< 4.64	< 1.09	< 1.09	< 0.60	< 0.60	0.88 ± 0.04	0.81 ± 0.04	< 5.42	< 5.42
	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg ⁻¹)	(mg kg ⁻¹)	(mg kg ⁻¹)	(µg kg-1)	(µg kg⁻¹)
5	< 4.64	< 4.64	< 1.09	< 1.09	< 0.60	< 0.60	< 1.47	< 1.47	< 5.42	< 5.42
	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg-1)
6	< 4.64	< 4.64	< 1.09	< 1.09	< 0.60	< 0.60	0.51 ± 0.03	0.44 ± 0.02	< 5.42	< 5.42
	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)	(µg kg-1)	(mg kg ⁻¹)	(mg kg ⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)
7	< 4.64	< 4.64	< 1.09	< 1.09	< 0.60	< 0.60	< 1.47	< 1.47	< 5.42	< 5.42
	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)	(µg kg-1)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)
8	< 4.64	< 4.64	< 1.09	< 1.09	< 0.60	< 0.60	< 1.47	< 1.47	< 5.42	< 5.42
	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg-1)	(µg kg ⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)	(µg kg⁻¹)
9	< 4.64	< 4.64	< 1.09	< 1.09	< 0.60	< 0.60	0.61 ± 0.03	0.55 ± 0.03	< 5.42	< 5.42
	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)	(mg kg ⁻¹)	(mg kg ⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)
10	< 4.64	< 4.64	< 1.09	< 1.09	< 0.60	< 0.60	< 1.47	< 1.47	< 5.42	< 5.42
	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg-1)	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)
11	< 4.64	< 4.64	< 1.09	< 1.09	< 0.60	< 0.60	0.60 ± 0.03	0.54 ± 0.03	< 5.42	< 5.42
	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg-1)	(mg kg ⁻¹)	(mg kg ⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)
	Sc	Sc	Y	Y	Yb	Yb	U	U	La	La
Sample	Average	Composite	Average	Composite	Average	Composite	Average	Composite	Average	Composite
	Aliquots	Sample	Aliquots	Sample	Aliquots	Sample	Aliquots	Sample	Aliquots	Sample
1	< 0.96	< 0.96	< 0.73	< 0.73	< 0.84	< 0.84	< 1.10	< 1.10	< 1.50	< 1.50
	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg⁻¹)				
2	0.64 ± 0.03	0.47 ± 0.02	0.77 ± 0.04	0.81 ± 0.04	< 0.84	< 0.84	< 1.10	< 1.10	2.86 ± 0.15	2.71 ± 0.14
	(mg kg ⁻¹)	(mg kg ⁻¹)	(mg kg ⁻¹)	(mg kg ⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(mg kg ⁻¹)	(mg kg ⁻¹)
3	< 0.96	< 0.96	< 0.73	< 0.73	< 0.84	< 0.84	< 1.10	< 1.10	< 1.50	< 1.50
	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)	(µg kg-1)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg⁻¹)
4	< 0.96	< 0.96	< 0.73	< 0.73	< 0.84	< 0.84	< 1.10	< 1.10	< 1.50	< 1.50
	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg-1)	(µg kg ⁻¹)	(µg kg-1)
5	< 0.96	< 0.96	< 0.73	< 0.73	< 0.84	< 0.84	< 1.10	< 1.10	< 1.50	< 1.50
	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)
6	< 0.96	< 0.96	< 0.73	< 0.73	< 0.84	< 0.84	< 1.10	< 1.10	< 1.50	< 1.50
	(µg kg-1)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg ⁻¹)				
7	< 0.96	< 0.96	< 0.73	< 0.73	< 0.84	< 0.84	< 1.10	< 1.10	< 1.50	< 1.50
	(µg kg⁻¹)	(µg kg ⁻¹)	(µg kg-1)	(µg kg ⁻¹)						
8	< 0.96	< 0.96	< 0.73	< 0.73	< 0.84	< 0.84	< 1.10	< 1.10	< 1.50	< 1.50
	(µg kg⁻¹)	(µg kg ⁻¹)								
9	< 0.96	< 0.96	< 0.73	< 0.73	< 0.84	< 0.84	< 1.10	< 1.10	< 1.50	< 1.50
	(µg kg⁻¹)	(µg kg ⁻¹)								
10	< 0.96	< 0.96	< 0.73	< 0.73	< 0.84	< 0.84	< 1.10	< 1.10	< 1.50	< 1.50

Table S13: Results of the rare earth and uranium analysis of samples 2, 3, 4, 6, 9 and 11, obtained by comparing the averages of the aliquots with the composite sample of the aliquots.

	(µg kg-1)	(µg kg⁻¹)	(µg kg-1)	(µg kg-1)	(µg kg-1)	(µg kg-1)				
11	< 0.96	< 0.96	< 0.73	< 0.73	< 0.84	< 0.84	< 1.10	< 1.10	< 1.50	< 1.50
11	(µg kg⁻¹)	(µg kg-1)								



Figure S3: Concentration of rare earth elements and uranium (< 0.1%) in oil sludge samples. The missing columns mean that the values are < LQ. The margin of error for all measurements is less than 5%.

Table S14: Results of the rare earth and uranium analysis of sample 2, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 2						
Element	SD	RSD%	RE%	HO _R	Z-score		
Er	1.00E-04	0.02	2.68	0.00	16.56		
Eu	-	-	-	-	-		
Lu	-	-	-	-	-		
Nd	8.00E-03	0.42	4.78	0.03	24.33		
Sm	2.00E-03	0.32	34.10	0.02	8.00		
Sc	2.00E-04	0.04	37.54	0.00	77.38		
Y	2.00E-03	0.22	5.53	0.01	2.71		
Yb	-	-	-	-	-		
U	-	-	-	-	-		
La	0.01	0.44	5.43	0.03	1.17		

Table S15: Results of the rare earth and uranium analysis of sample 3, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 3						
Element	SD	RSD%	RE%	HO _R	Z-score		
Er	-	-	-	-	-		
Eu	-	-	-	-	-		
Lu	-	-	-	-	-		
Nd	3.00E-03	0.54	14.49	0.03	2.33		
Sm	-	-	-	-	-		
Sc	-	-	-	-	-		
Y	-	-	-	-	-		
Yb	-	-	-	-	-		
U	-	-	-	-	-		
La	-	-	-	-	-		

Table S16: Results of the rare earth and uranium analysis of sample 4, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 4							
Element	SD	RSD%	RE%	HO _R	Z-score			
Er	-	-	-	-	-			
Eu	-	-	-	-	-			
Lu	-	-	-	-	-			
Nd	1.15E-03	0.13	8.12	0.01	5.75			
Sm	-	-	-	-	-			
Sc	-	-	-	-	-			
Y	-	-	-	-	-			
Yb	-	-	-	-	-			
U	-	-	-	-	-			
La	-	-	-	-	-			

Table S17: Results of the rare earth and uranium analysis of sample 6, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 6						
Element	SD	RSD%	RE%	HO _R	Z-score		
Er	-	-	-	-	-		
Eu	-	-	-	-	-		
Lu	-	-	-	-	-		
Nd	1.40E-03	0.26	14.59	0.02	4.74		
Sm	-	-	-	-	-		
Sc	-	-	-	-	-		
Y	-	-	-	-	-		
Yb	-	-	-	-	-		
U	-	-	-	-	-		
La	-	-	-	-	-		

Table S18: Results of the rare earth and uranium analysis of sample 9, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 9						
Element	SD	RSD%	RE%	HO _R	Z-score		
Er	-	-	-	-	-		
Eu	-	-	-	-	-		
Lu	-	-	-	-	-		
Nd	1.00E-03	0.50	11.26	0.03	6.51		
Sm	-	-	-	-	-		
Sc	-	-	-	-	-		
Y	-	-	-	-	-		
Yb	-	-	-	-	-		
U	-	-	-	-	-		
La	-	-	-	-	-		

Table S19: Results of the rare earth and uranium analysis of sample 11, obtained by comparing the averages of the aliquots with the composite sample of the aliquots. SD is the standard deviation, RSD% the relative standard deviation, HO_R the HORRAT and the Z-score values. The elements not represented were in concentrations below the limit of quantification in these samples.

	Sample 11						
Element	SD	RSD%	RE%	HO _R	Z-score		
Er	-	-	-	-	-		
Eu	-	-	-	-	-		
Lu	-	-	-	-	-		
Nd	5.60E-04	0.32	10.98	0.02	10.64		
Sm	-	-	-	-	-		
Sc	-	-	-	-	-		
Y	-	-	-	-	-		
Yb	-	-	-	-	-		
U	-	-	-	-	-		
La	-	-	-	-	-a		