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Full Length Research Paper

Vertical distribution of natural and artificial (¹³⁷Cs) radionuclides in soils surrounding the phosphogypsum deposit at Taiba Ndiaye, Senegal

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Résumé :

L'objectif de ce travail est d'étudier la distribution verticale des radionucléides naturels (²³⁸U, ²²⁶Ra, ²¹⁰Pb, 232 Th, et 40 K) et 137 Cs dans des profils de sol aux alentours de l'industrie de production d'acide phosphorique de Taïba Ndiaye (Sénégal). Les profils de sol prélevés ont été mesurés par spectrométrie gamma. Les concentrations des activités de 226Ra, 232Th, et 40K trouvées sont inferieur aux valeurs recommandées par UNSCEAR (1982). Le rapport A_{Pb}/ A_{Ra} calculé montre un excès de ²¹⁰Pb. Les distributions verticales de ²³⁸U, 226 Ra, 210 Pb, 232 Th, 40 K, et 137 Cs ont été représentées. L'activité équivalente de radium (Raeq) est de 32.03 ± 2.91 Bq.kg⁻¹, elle est inférieur à la valeur recommandée qui est de 370 Bq.kg⁻¹. Les valeurs du débit de dose absorbée (D) et de l'équivalent de dose efficace annuelle (AEDE) sont de 14.45 ± 1.37 nGy.h⁻¹ et 17.72 ± 1.68 µSv.y⁻¹ respectivement, ils sont inferieur aux valeurs recommandées par UNSCEAR (2000).

Abstract:

In this study, natural radionuclides (²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²³²Th, and ⁴⁰K) and ¹³⁷Cs in soil profiles in the vicinity of phosphoric acid production industry in Taiba Ndiaye (Senegal) were determined using gamma-ray spectrometry. The activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K in this study were lower than their values given by UNSCEAR (1982). A_{Pb}/ A_{Ra} ratio was calculated and showed an excess of ²¹⁰Pb. A study of the vertical distribution of ²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²³²Th, ⁴⁰K, and ¹³⁷Cs was also conducted. The radium equivalent activity (Raeq) calculated is 32.03 ± 2.91 Bq.kg⁻¹, which was lower than the recommended limit of 370 Bq.kg⁻¹. The obtained values of absorbed dose rate (D) and the annual effective dose rate (AEDE) were 14.45 ± 1.37 nGy.h⁻¹ and 17.72 \pm 1.68 µSv.y⁻¹, respectively and found less than the recommended limit values reported by UNSCEAR (2000).

Keyword: Natural radionuclides, ¹³⁷Cs, Vertical distribution, Absorbed dose rate, and Annual effective dose.

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1. Introduction

Since the formation of the earth, there is natural radioactivity that is widely distributed in the different compartments of the environment (soil, water, air, and plants) (Nenadović et al., 2012; Ramli et al., 2005; Tsabaris et al., 2007). This natural radioactivity represents about 90% of the exposure received by humans with an annual effective dose of 2.4 mSv.y-1 according to UNSCEAR (2000).

Natural radionuclides in the ground such as the uranium, actinium, and thorium series, and potassium are for UNSCEAR (1993) the main component of the exposure coming from the background. Their concentrations depend on the geological formation of an area and the content of natural radionuclides in the rocks from which the soils are derived. Soils are complex mixtures of solids that can contain moisture which plays a major role in the distribution of radionuclides (Kabata-pendias and Pendias, 2001). The radionuclides present in the soil can be dissolved in solution, exchanged by ions in reaction, complexed with the organic matter of the soil, or precipitated in the pure or mixed solid state (Dragovic and Onjia, 2006). They can move through the soil, water, air, and food products (Gavrilescu et al., 2009).

The distribution of radionuclides can be influenced by human activities. Since 1956, a phosphoric acid and fertilizer production industry has been established in Taïba Ndiaye in the Thiès region. The conversion of Apatite from phosphate rock by sulfuric acid to form phosphoric acid and sparingly soluble gypsum generates a large quantity of solid waste called phosphogypsum. During production, the radionuclides are redistributed between phosphoric acid, fertilizer, and phosphogypsum depending on their chemical properties. This redistribution can thus lead to the reinforcement of the natural radioactivity in the phosphogypsum. The open pit phosphogypsum deposits around the production area may be altered by climatic conditions. This can lead to radioactive contamination of the immediate environment, thus constituting a source of exposure for the workers and residents. To contribute to the knowledge of the distribution of radionuclides and the radiological situation in soils around a phosphate mining area in Senegal, this work aimed to: determine the activity concentration of ²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²³²Th, ⁴⁰K, and ¹³⁷Cs; evaluate the physicochemical parameters such as soil texture, pH, and OM and the vertical distribution of natural radionuclides and ¹³⁷Cs; and assess the equivalent radium activity, the annual dose rate, and the annual effective dose.

2. Materials et methods

The sampling site is located in the commune of Taïba Ndiaye. Taïba Ndiaye is situated in the region of Thies, 100 km from the capital of Senegal (Dakar). It is between latitude $15^{\circ}02'19N$ and longitude $16^{\circ}52'42W$. Three (3) sampling points were selected around this area. At each point, a sample was collected over a depth of 20 cm using a sample corer. Each soil profile of a 20 cm depth sample is then subdivided into four (4) parts of 5 cm. After removal of the debris, the samples are packed in plastic bags, then labeled S_xH_y (S_x : sampling point and H_y : sampling depth).

The samples were dried at a temperature of 105°C with a dry oven to eliminate humidity. They were then crushed and sieved with a sieve with a mesh diameter of 2 mm. After drying, grinding, and homogenization, the samples were transferred to 60 mL beakers with an internal diameter of 72 mm. After conditioning, the samples are sealed in vacuum aluminum to prevent any loss of radon during the period of establishment of the secular equilibrium between ²²⁶Ra and its short-lived descendants. This period was at least 4 weeks during the study.A low-background Broad Energy Germanium (BEGe) detector was used for determining the activity

concentrations of ²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²³²Th, ⁴⁰K, and ¹³⁷Cs in soil profiles. The relative efficiency of the detector was 50% at 1332 keV. The detector has an energy resolution of 0.7 keV at 60 keV and 1.8 keV at 1332 keV measured with ²⁴¹Am and ⁶⁰Co, respectively. A standard of mixed radionuclides containing ²⁴¹Am, ²¹⁰Pb, ¹³⁹Ce, ¹³⁷Cs, ¹¹³Sn, ¹⁰⁹Cd, ⁸⁸Y, ⁸⁵Sr, ⁶⁰Co, ⁵⁷Co, and ⁵¹Cr in the same geometry as the samples was used for energy and efficiency calibration of the detectors. Sample measurements were carried out with Genie 2000 software for a counting time of 80,000 s. The activity concentration of ²³⁸U has been estimated using the gamma-ray emission of ²³⁴Th at 63.3 keV. The gamma-ray emissions of ²¹⁴Pb at 295.2 keV and of ²¹⁴Bi at 609.3 keV were used for analysis. For ²³²Th, its activity concentration was calculated from the gamma-ray emissions of ²²⁸Ac at 911.2 keV and ²⁰⁸Tl at 583.2 keV. The activity concentrations of ²¹⁰Pb, ⁴⁰K, and ¹³⁷Cs were calculated using their gamma-ray emissions at 46.5 keV, 1460.8 keV, and 661.6 keV, The activity concentration of respectively. the radionuclides of interest was calculated using the following equation (Ndour et al., 2021):

$$A(Bqkg^{-1}) = \frac{N_{net,i}}{P(E_i) \times \varepsilon(E_i) \times t \times m \times F_{c,i}}$$
[Eq.1]

Where $N_{net,i}$ represents the net counts under the full absorption peak, $P(E_i)$ the emission probability, $\epsilon(E_i)$ the detector efficiency at energy E_i , t the counting time, m the sample mass, and $F_{c,i}$ the corrective factors taking into account the self-attenuation, the coincidence summing, and the radioactive decay. The self-attenuation and the coincidence summing were corrected using a transmission bench and the GESPECOR software package based on the Monte-Carlo simulation method. The propagation of uncertainties law was used to calculate the uncertainty of the activity concentration described by the following equation:

$$\frac{u(A)}{A} = \sqrt{\frac{u^2(N_{net,i})}{N_{net,i}^2} + \frac{u^2(F_{c,i})}{F_{c,i}^2} + \frac{u^2(P(E_i))}{P^2(E_i)} + \frac{u^2(\varepsilon(E_i))}{\varepsilon(E_i)}}$$
[Eq.2]

The carbon assay is done by the modified Black & Wakley method (Bahadori and Tofighi, 2016). Organic carbon is oxidized with a mixture of 1N potassium dichromate ($K_2Cr_2O_7$) and concentrated sulfuric acid (H_2SO_4). The carbon dosage is done by spectrophotometer at 600 nm. The percentage of organic matter in the soil was calculated based on the carbon content in the soil.

The texture and pH of the soil samples were evaluated using a laser diffractometer Mastersize 3000 and a pH meter.

3. Results and discussions

3.1. Physicochemical parameters of soil samples

Soil texture includes the percentage of sand, silt, and clay particle in a sample. The sizes of clay, silt, and sand particle are < 0.002 mm, 0.002-0.006 mm, and 0.06-2 mm, respectively.

The first profile with a pH of 6.39-6 .89 and organic matter content between 0.95-1.2% includes 0.24-0.48% of clay, 16.56 -25.08% of silt, and 74.45-83.25% of sand. The proportion of clay, silt, and sand in soil texture was between 0.25-0.45%, 16.52-29.2%, and 70.35-83.08%, respectively for the second profile. Its pH varied between 6.39-6.57 and organic matter content between 0.32-0.98 percent.

The third profile is composed of 0.19-0.57% of clay, 12.04-20.20% of silt, and 69.48-87.77% of sand with a pH between 5.66-6.66 and organic matter content between 0.32-0.62percent.

Based on USDA (1999), the sampled soils were characterized as sandy soil.

Table 1. Soil texture, pH, and organic matter in soil profiles.

Sample	Soil text	ure	pН	OM	
number	(%)				(%)
	Clay	Silt	Sand		
S_1H_1	0.24	23.89	75.87	6.89	0.99
S_1H_2	0.26	18.55	81	6.63	1.01
S_1H_3	0.2	16.56	83.25	6.39	1.20
S_1H_4	0.48	25.08	74.45	6.8	0.95
S_2H_1	0.45	29.2	70.35	6.57	0.98
S_2H_2	0.25	19.12	80.64	6.41	0.52
S_2H_3	0.4	17.45	82.15	6.39	0.40
S_2H_4	0.39	16.52	83.08	6.51	0.32
S_3H_1	0.40	20.2	79.4	6.66	0.61
S_3H_2	0.57	29.95	69.48	6.39	0.62
S ₃ H ₃	0.41	15.01	84.58	6.01	0.34
S ₃ H ₄	0.19	12.04	87.77	5.66	0.32

Fig.1 shows the trend of soil texture, pH, and organic matter in soil profiles. There is an increase of sand with the depth. In the first profile, the bottom layer shows less sand and OM and more silt – like in the most upper layer. The OM is highest in the third layer in this profile. In the second and third profiles, the OM is decreasing steadily. The silt and clay are correlating and the pH and sand are slightly anti-correlation in the three profiles.



Figure 1. Trend of soil texture, pH, and organic matter in soil profiles.

3.2. Activity concentration of natural radionuclides and ¹³⁷Cs in soil samples

The activity concentration of natural radionuclides and ¹³⁷Cs in soil samples is presented in Table 2. The activity concentration of ²³⁸U, ²²⁶Ra, and ²¹⁰Pb in soil samples ranged from 11.2 ± 1.9 Bq.kg⁻¹ to 20.8 ± 2.5 Bq.kg⁻¹, from 11.6 ± 0.3 Bq.kg⁻¹ to 21.7 ± 0.8 Bq.kg⁻¹, and from 11.8 ± 1.8 Bq.kg⁻¹ to 30.9 ± 3.1 Bq.kg⁻¹, respectively. Their respective average values were 16.1 ± 2.7 Bq.kg⁻¹, 17.4 ± 3.1 Bq.kg⁻¹, and 24.1 ± 6.2 Bq.kg⁻¹ to 40.5 Bq.kg⁻¹, 20.5 ± 3.4 Bq.kg⁻¹ to 30.2 ± 4.3 Bq.kg⁻¹, and 0.3 ± 0.1 Bq.kg⁻¹ to 0.6 ± 0.1 Bq.kg⁻¹ for ²³²Th, ⁴⁰K, and ¹³⁷Cs, respectively with average values of 8.9 ± 0.8 Bq.kg⁻¹, 25.2 ± 3.0 Bq.kg⁻¹, and 0.4 ± 0.1 Bq.kg⁻¹.

According to UNSCEAR (1982), the worldwide average activity concentration of ²²⁶Ra, ²³²Th, and ⁴⁰K are 35 Bq.kg⁻¹, 30 Bq.kg⁻¹, and 400 Bq.kg⁻¹, respectively which are greater than the activity concentrations reported in this study.

 Table
 2. Activity concentration of natural radionuclides and ¹³⁷Cs in soil sample

Sample	Activity concentration (Bq.kg ⁻¹)					
number	²³⁸ U	²²⁶ Ra	²¹⁰ Pb	²³² Th	⁴⁰ K	¹³⁷ Cs
S_1H_1	17.6 ± 2.2	20.3 ± 0.7	27.7 ± 3.1	8.1 ± 0.5	28.7 ± 3.9	0.5 ± 0.1
S_1H_2	16.2 ± 2.1	20.2 ± 0.7	27.7 ± 3.1	8.2 ± 0.7	27.4 ± 3.7	0.6 ± 0.1
S_1H_3	20.8 ± 2.5	21.7 ± 0.8	30.9 ± 3.1	8.2 ± 0.7	30.2 ± 4.3	0.5 ± 0.1
S_1H_4	18.8 ± 2.6	19.9 ± 0.4	26.8 ± 3.1	8.4 ± 0.5	27.6 ± 4.1	0.4 ± 0.1
S_2H_1	16.6 ± 2.1	19.2 ± 0.7	30.0 ± 3.3	9.5 ± 0.6	24.9 ± 3.8	0.3 ± 0.1
S_2H_2	18.1 ± 2.4	19.4 ± 0.6	30.2 ± 3.4	8.8 ± 0.4	22.3 ± 3.6	0.3 ± 0.1
S_2H_3	12.8 ± 1.8	13.8 ± 0.7	18.6 ± 2.2	9.8 ± 0.6	24.6 ± 3.7	0.3 ± 0.1
S_2H_4	11.2 ± 1.9	11.6 ± 0.3	11.8 ± 1.8	10.1 ± 0.5	24.3 ± 3.8	< 0.6
S ₃ H ₁	16.7 ± 1.9	18.4 ± 0.2	26.1 ± 2.6	9.8 ± 0.7	26.8 ± 3.2	0.5 ± 0.1
S_3H_2	16.0 ± 2.0	15.5 ± 0.5	23.0 ± 2.6	7.8 ± 0.5	21.3 ± 3.1	0.4 ± 0.1
S ₃ H ₃	14.1 ± 2.0	15.7 ± 0.9	20.6 ± 2.6	8.4 ± 1.0	20.5 ± 3.4	0.3 ± 0.1
S ₃ H ₄	13.7 ± 2.0	13.1 ± 0.7	15.8 ± 2.1	9.1 ± 0.8	24.1 ± 3.6	0.4 ± 0.1
Mean ± SD	16.1 ± 2.7	17.4 ± 3.1	24.1 ± 6.2	8.9 ± 0.8	25.2 ± 3.0	0.4 ± 0.1

3.3 ²²⁶Ra and ²¹⁰Pb equilibrium state

The equilibrium state between ²¹⁰Pb and ²²⁶Ra was studied by calculating the ratios of their activity concentrations (A_{Pb}/A_{Ra}). The obtained values were greater than unity in all samples. There is therefore disequilibrium between ²¹⁰Pb and ²²⁶Ra with an enrichment of ²¹⁰Pb. This can be explained by the exhalation of radon from phosphogypsum deposits which escape into the air where it will disintegrate into ²¹⁰Pb. The rainfall leads to a deposit and an accumulation of ²¹⁰Pb at a certain depth.



Figure 2. 210 Pb and 226 Ra activity concentrations ratios (A_{Pb}/A_{Ra}) in soil samples.

3.4 Vertical distribution of natural radionuclides and ¹³⁷Cs in soil

The vertical distribution of 238 U in soil profiles is presented in Fig.3 (a). The third profile showed a decrease of 238 U with depth while the first presented a decrease of 238 U, then its increase followed by a decrease. For the second profile, 238 U increase in depth followed by a decrease.



Figure 3. Vertical distribution (a) of ²³⁸U; (b) of ²²⁶Ra and (c) of ²¹⁰Pb in soil profiles.

According to Schulz (1965), many studies have shown that the dominant process is between uranium and organic matter in surface soil layers. The organic matter content and activity concentration are consistent in all soil profiles (see Fig.4).



Figure 4. Trend of organic matter, ²³⁸U, ²²⁶Ra, and ²¹⁰Pb in soil profiles.

Fig.3 (b) shows the vertical distribution of ²²⁶Ra in soil profiles. In the first and third profiles, the activity concentration of ²²⁶Ra decreased slightly followed by an increase and a decrease. In the second profile, there is an increase followed by a progressive decrease of ²²⁶Ra the activity concentration. The vertical distribution of ²²⁶Ra in soil profiles follows that of ²³⁸U. According to Atwood (2010), organic matter can control the retention of radium. The obtained results confirm the concordance between organic matter and the activity concentration of ²²⁶Ra.

The vertical distribution of ²¹⁰Pb in soil profiles is shown in Fig.3 (c). In the first and third profiles, the activity concentration of ²¹⁰Pb decreased and increased slightly, respectively. That is followed by a progressive decrease of the activity concentration of ²¹⁰Pb for the third profile and an increase then a decrease for the first profile. The vertical distribution of ²¹⁰Pb in soil profiles is similar to that of ²²⁶Ra.

The vertical distribution of 232 Th in soil profiles is illustrated in Fig.5 (a). In the first profile, there was an increase of 232 Th activity concentration in depth. The second and third profiles presented a decrease followed by an increase of 232 Th activity concentration. The activity concentration of 232 Th increased with depth except for the second and third profiles.



Figure 5. Vertical distribution (a) of ²³²Th; (b) of ⁴⁰K; and (c) of ¹³⁷Cs in soil profiles.

Fig.5 (b) shows the vertical distribution of 40 K in soil profiles. In the first and second profiles, there was a decrease and increase of 40 K activity concentration and then a decrease. In the third profile, the 40 K activity concentration decreased and increased at the end layer. The vertical distribution of 137 Cs in soil profiles is shown in Fig.5 (c). In the first profile, the activity concentration of 137 Cs increased at the second layer and decreased until the fourth layer. The second profile presented a decrease of 137 Cs activity concentration at

the second layer which became constant at the third layer and decreased at the end layer. In the third profile, the activity concentration of 137 Cs decreased until the third layer and increased at the fourth layer. The activity concentration of 137 Cs varied slightly in the soil profiles.

3.5 Radium equivalent activity, absorbed dose rate, and annual effective dose

The radium equivalent activity was calculated according to the following equation:

Raeq
$$(Bq.kg^{-1}) = A_{Ra} + 1,43A_{Th} + 0,077A$$
 [Eq.3]

Where A_{Ra} , A_{Th} , and A_K are the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K, respectively. The coefficients of equation (1.1) are calculated by estimating that 370 Bq.kg⁻¹ of ²²⁶Ra, 259 Bq.kg⁻¹ of ²³²Th, and 4810 Bq.kg⁻¹ of ⁴⁰K each produce the same gamma dose rate (Fall et al., 2023). The ²²⁶Ra activity concentration is used since it is responsible for 98.5% of the radiological effects of the uranium series (Ndour et al., 2020).

The radium equivalent activity in soil samples varied from 27.99 Bq.kg⁻¹ to 35.76 Bq.kg⁻¹ with an average value of 32.03 ± 2.91 Bq.kg⁻¹. This average value is lesser than compared to the recommended value of 370 Bq.kg⁻¹ (Ndour et al., 2021; UNSCEAR, 2000).

 Table 3. Radium equivalent activity, absorbed dose

 rate, and annual effective dose in soil samples.

Sample	Raeq	$D(nGy.h^{-1})$	E (μ Sv.an ⁻¹)
number	$(Bq.kg^{-1})$		
S_1H_1	34.01	15.43	18.92
S_1H_2	34.09	15.45	18.95
S_1H_3	35.76	16.24	19.92
S_1H_4	34.11	15.45	18.95
S_2H_1	34.76	15.67	19.22
S_2H_2	33.75	15.23	18.68
S_2H_3	29.83	13.37	16.40
S_2H_4	27.99	12.51	15.34
S_3H_1	34.49	15.54	19.06
S_3H_2	28.31	12.77	15.66
S ₃ H ₃	29.23	13.16	16.14
S_3H_4	28.00	12.56	15.41
Mean ± SD	32.03 ±	14.45 ±	17.72 ±
	2.91	1.37	1.68
UNSCEAR	370	57	70
(2000)			

The absorbed dose rate in the air at 1 m above the ground level was calculated using the following equation (Durusoy and Yildirim, 2017)

$$D(nGy.h^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K}$$
 [Eq.4]

Where A_{Ra} , A_{Th} , and A_K are the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K, respectively. The coefficients 0,462, 0,604, and 0, 0417 nGy.h⁻¹/ Bq.kg⁻¹ are conversion factors of the activity concentrations of $^{226}\text{Ra},\,^{232}\text{Th},\,\text{and}\,^{40}\text{K}$ into absorbed dose rate in air.

The absorbed dose rate in soil ranged from 12.51 $nGy.h^{-1}$ to 16.24 $nGy.h^{-1}$. Its average value was 14.45 ± 1.37 $nGy.h^{-1}$, this value is less than the worldwide average value of 57 $nGy.h^{-1}$ (UNSCEAR, 2000).

To know the radiological risk to which an individual is exposed, the dose rate absorbed in the air at 1 m above the ground level is converted into an annual effective dose in the human body by using a conversion coefficient of 0.7 Sv.Gy^{-1} . The annual effective dose is given by the following equation (Miah et al., 2013; UNSCEAR, 2000):

$E[\mu Svan^{-1}] = D[nGyh^{-1}] \times 8760[han^{-1}] \times 02007[SvGy^{-1}] \times 10^3$ [Eq.5]

This equation takes into account that people spend about 20% of their time outdoors and the annual exposure time is 8760 h.

The annual effective dose was in the range of 15.34 μ Sv.an⁻¹ to 19.92 μ Sv.an⁻¹ with an average value of 17.72 ± 1.68 μ Sv.an⁻¹. This value is less than the worldwide average value of 70 μ Sv.an⁻¹.

4. Conclusion

Vertical distribution of natural radionuclides and ¹³⁷Cs in soil profiles around the phosphate industry in Taiba Ndiaye (Senegal) were measured. Twelve soil samples were measured by gamma spectrometry to assess the activity concentrations of natural radionuclides (²³⁸U, ²²⁶Ra, ²³²Th, ²¹⁰Pb, and ⁴⁰K) and ¹³⁷Cs. The average activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K are compared to global mean values and found to be lower. The radiological parameters were estimated and found lower than 370 Bq.kg⁻¹, 57 nGy.h⁻¹, and 70 µSv.an⁻¹, respectively for the radium equivalent activity, the absorbed dose rate at 1 m above the ground, and the annual effective dose rate. Apb/ ARa ratio was calculated to estimate the equilibrium between ²¹⁰Pb and ²²⁶Ra. All the ratios were greater than the unity. The vertical distributions of the prospected radionuclides have been studied taking into account the pH, the soil texture (clay, silt, sand), and organic matter. The vertical distributions of radionuclides reveal that the activity concentration of ²³⁸U, ²²⁶Ra, and ²¹⁰Pb and organic matter content are consistent but also the presence of the phosphoric acid industry leads to an enrichment of ²¹⁰Pb.

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Competing interests

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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