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EVALUATION OF THE EFFICIENCY OF SOME MINERALOGICAL AND ORGANIC MATERIALS TO REMOVE SOME HEAVY METALS FROM CONTAMINATED SOIL

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Abstract

To evaluate the efficiency of some organic materials (corncob and active charcoal) and mineralogical (phosphate rock and Bentonite) to remove some heavy metals (Pb, Zn, Ni, Cd) from soil and contaminated water. A biological experiment was conducted in the plastic house of the Department of soil Science and Water Resources – College of Agricultural Engineering Sciences – University of Baghdad – Jadiriya in spring season 2018 using two types of calcareous soils with different textures, the first was silty clay loam (SiCL) and the second was loam (L), capacity in plastic pots 20 kg soil. were treated with the above materials at one level 5% weight/weight (individually), 5 seeds of Maize plant was planted for each pot, reduced to three plants per pot after a week of germination. A complete randomization design (CRD) was used. It included 30 experimental units (2 textures × 3 replicates × 4 processing materials + 6 control), the fertilizers was added according to fertilizers recommendation, the plants were irrigated with tap water at a height of plant 5 cm above the soil surface, and then irrigated by contaminated water with heavy metals (Pb, Zn, Ni, Cd) in concentrations of 20 $mg.L^{-1}$ for both of them after the depletion of 50% of the available water for the plant by weight method , soil samples were taken before the flowering stage For laboratory analysis. The results showed that processing materials significant effect on reducing the available concentrations of all studied elements and increasing their total concentrations. The order of materials effect was as follow:

Active charcoal (M_2) > Phosphate rock (M_3) > Corncob (M_1) > Bentonite (M_4)

Soil texture also significantly affected on reducing the available concentrations and increased the total concentrations of heavy metals (Pb, Zn, Ni and Cd). While the available Nickel was not affected, The texture of SiCL gave the lowest value of the available concentrations comparing with L texture, The results also indicated the exceeded of the active charcoal treatment of both textures by giving the highest efficiency of reducing the available concentrations and increasing the total concentrations of heavy metals.

Key words : Processing materials - heavy metals - removing efficiency of heavy metals.

Introduction

The pollution issue is one of the most pressing environmental problems that began to take serious environmental, economic and social dimensions, especially after the industrial revolution in Europe and industrial expansion and backed by modern technology therefore, the human being is working to balance the environmental components in terms of industrial activity, population expansion and the expansion of cities that have a devastating impact on the environment as a result of the tremendous development witnessed by the world, this has resulted in a lot of damage to the components of the environment as a result of contamination with many pollutants, including heavy metals (Karaca *et al.,* 2010).

That heavy metals resulting from a various activities remain in the soil to reach plants, seeds and other soil components. The presence of these pollutants in the environment affects the health of humans, animals, agricultural crops and wildlife. The soil contaminated with heavy metals constitute the biggest environmental challenges facing the whole world. Heavy metals lie in their cumulative qualities in the bodies of living organisms and cause harm to humans when eating food contaminated with these elements, Heavy metals such as Lead, Cadmium and Nickel are very dangerous

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pollutants of soil, water and air (Pierzynski, *et al.,* 2000), Therefore, it is necessary to work on reduce the damage for heavy metals in the soil and water, been working on many techniques and technological means that reduce the presence of these elements and mitigate from effects, including use of some organic and mineral for reduce from of heavy metals harmful to soil, water and plant. Many studies,showed Such as study Heike, (2004) The many material such as active charcoal, corncob, phosphate rock and bentonite and others can be used as processing materials for heavy metals the following materials used in the study:

Active charcoal

Carbonized materials are the product of incomplete combustion of the plant, referred to combustion products coal, ash, and charcoal with black carbon (BC), Black carbon is a mixture resistant for decomposition biological and chemical. Nouri *et al.,* (2003) explained on their study when used active charcoal for adsorb three types of aromatic hydrocarbons containing different effective aggregates, found that adsorption efficiency depends on the size of active functional group and its nature and tendency to adsorbent and surface area, the results of their study showed that coal is a good alternative can be used for remove dyes from waste water resulting from textile industries. Proved Aslam *et al.,* (2005) explained the reason of efficient use the activated charcoal as a adsorbent to remove solutes in water is due to the large number of surface and internal pores that make the surface area exposed to adsorption big relative to the actual effective volume.

Corncob

That addition Corn residues, such as corncob, prepare from effective strategies on reducing the damage of pollution and salinity of irrigation water and increased the plant Bearing They are improves the distribution of soil pores that increase from ability hold water and elements, and improve the secretions of the roots such as organic acids that regulate soil reaction and reduce from the harmful effect for salts and heavy metals on soil solution (El-Dardiry, 2007). The important fields that used it corncob was on treat that water contaminated and saline and then use that the water in irrigation, in a study of Abed *et al.,* (2012) included best treatment in water processing, used Four sources of plant organic residues were: Champlain, Corncob, Wheat residues and sunflower stalks. The results showed that the best of these materials for processing of salt water is Corncob and Wheat residues.

Phosphate rock

Phosphate rocks referred to as phosphors are sedimentary rocks and most of these rocks are deposited contain high concentrations of phosphorus element and is one of the most important raw sources of Phosphorus. Phosphate rock is one of the mineral material which used to processing contaminated soils through adsorption, and chelation to formed complexes with contaminants found in the soil, Several studies showed that phosphate rock has a high ability to remove heavy metals from aqueous solutions and soil Brown *et al.,* (2005) studed the ability or phosphate rock to remove heavy metals in the soil and it different the according to contaminated element in order Pb > Cu > Zn with a adsorption capacity 138, 114, 83.2 mmol / kg phosphate rock.

Bentonite

Bentonite is a commercial name for a type of clay consists form the Basically of smectite and the dominant metal is montmorlonite. Mention Yueh *et al.,* (2016) finally showedthat a lot of research has been done using the physiochemical processes for decontaminate on the soil and water using several materials, known as adsorption, which is the process of transferring the material from the liquid phase to the solid surface, of these materials is Bentonite clay, which consider from the materials adsorption important economically because of its high efficiency in removing heavy metals from soil and water. Mention Mohajeri *et al.,* (2018) shows that Bentonite is a suitable absorber and installed for heavy metals contaminants in the soil, even when the concentrations are high, this is because bentonite has a high cation exchange capacity (CEC) this characteristic and other properties enable it to react with contaminants that are dangerous when present in the solution. The aim of this research is to evaluate the efficiency of some organic and mineral substances in removing some heavy metals from contaminated soil and water.

Materials and Methods

Pots experiment was conducted in the plastic house for the department soil science and water resources - Faculty of Agricultural Engineering Sciences - University of Baghdad - Jadiriya for the spring agricultural season (2018) using two soils calcareous different texture the first silty clay loam (SiCL) and the second is loam (L), dried the soil pneumatically and grinded, from which a sample was taken and sieved whith a 4 mm For agriculture on plastic pots of 20 kg with diameters of 40 cm and diameter base 20 cm and height of 39 cm filled pots with soil after being treated with two types of organic materials are (corncob and active charcoal) and two types of mineral materials are (phosphate rock and bentonite) individually use on the processing 5% of soil weight equivalent of 19 kg soil to 1 kg of organic or mineral material after sieving these materials with a sieve diameter of its holes 4 mm, table 1 shows some characteristics of these materials.

Taking part of the two soil and air dried, grinded and gave up a sieve diameter of 2 mm for some chemical and physical analyzes before planting and shown in Table (2 and 3), A complete randomization design (CRD) was used it included 30 experimental units (2 textures × 3 replicated × 4 processing materials + 6 control) included : two types of Soil texture : silty clay loam (SiCL) symbolized (T_1) and Loam symbolized (T_2), processing materials included : Corncob (M_1), Active charcoal (M_2), Phosphate rock (M_3), Bentonite (M_4) three replicated : (R_1, R_2, R_3). Corn seeds Planted class (Fajer 1) on 9/4/2018 5 seeds per pot, faded to 3 Plants after a week of germination. NPK fertilizers were added according to the Maize fertilizer recommendation (120 – 60 – 240) (K – P – N Kg.h^{-1}) Respectively (Al – Bahrani, 2015) To soil tap water used to irrigation at the beginning of the experiment until the plant reaches a height of 5 cm above the soil surface, After this stage done irrigated the plant with water contaminated by heavy metals at concentrations of 20 ppm for all elements, Which attended from the salts $CdSO_4$, $NiSO_4$, $ZnSO_4$, $PbSO_4$, mixed together to irrigate for the during period from the age of the plant, Irrigation after depleting 50% of the available water for plant by weighing the pot with the soil and plant and adding the contaminated water to the plants when needed on the basis of the loss of available water by the weight method. (Table 4) shows the amount of concentrations the elements of added during stages of plant growth. Soil samples taken and all treatments before the flowering stage on 25/6/2018. Samples were preserved for analysis.

1. Total heavy metals in soil (Pb, Zn, Ni, Cd) : Take 0.5 g of soil to estimated the total elements, by digestion method using sulfuric acid (Black, 1965).
2. Available heavy metals in soil (Pb, Zn, Ni, Cd) : Take 0.5 g of soil to estimated the available elements and estimated by extraction using chelating compound (Diethylene triamine penta aceticacid) (DTPA) (Norvell and Lindsay, 1978).
3. Calculate the efficiency of added materials through the equation :

Removed efficiency =

$$\frac{\text{Concentration before planting - Concentration after planting}}{\text{Concentration before planting (original + added)}} * 100$$

Data were analyzed statistically according to the analysis of variance (ANOVA) by using a complete randomization design (CRD), The averages Were compared with the least significant difference test (LSD) use a program Genstat under the operating system (Windows 10) For statistical analysis.

Results and Discussion

Available heavy metals

The results of table 5 showed that the processing materials (M) had effected significantly on the values of available Lead element in the study soil. It gave the phosphate rock treatment (M_4) lowest value of available lead was 9.50 mg pb. Kg^{-1} soil while control treatment (M_0) (Irrigation with contaminated water without adding processing materials) gave highest value of available Lead was 14.26 mg Pb. Kg^{-1} soil thus the treatment of phosphate rock gave the highest decrease rate compared with reached other treatments 33.38%.

The results of tables (6, 7, 8) showed that processing materials (M) effect significant in reducing the concentrations of Zinc, Nickel and Cadmium available in the soil, treatment of charcoal (M_2) gave the

Table 1: Some physical and chemical properties of processing materials.

Properties	Unit	Processing materials (M)			
		Corncob (M_1)	Active charcoal (M_2)	Phosphate rock (M_3)	Bentonite (M_4)
Soil raction	-	6.50	7.43	7.5	7.12
Electrical conductivity	ds.m^{-1}	2.23	3.5	5.9	3.9
Organic Carbon (O.C)	gm.kg^{-1}	247.19	382.80	-	-
Organic matter (O.M)	gm.kg^{-1}	426.20	659.94	-	-
C / N	-	69.44	95.22	-	-
Total Nitrogen	gm.kg^{-1}	3.56	4.02	2.5	0.64
Total Phosphorus	gm.kg^{-1}	1.02	3.15	102.20	0.23
Total Potassium	gm.kg^{-1}	3.50	3.86	0.728	3.97
CEC	cmol.g-1	61.50	84.61	49.44	59.13
Total Lead	mg.kg-1	1.84	1.03	0.71	3.18
Total Zinc	mg.kg-1	2.81	2.12	2.44	4.60
Total Nickel	mg.kg-1	1.86	1.02	1.15	1.75
Total Cadmium	mg.kg-1	0.13	0.18	0.28	0.14

Table 2: Some physical and chemical qualities of study soil.

Qualities		Value		Unit
		Abu Ghraib soil (T_1)	Jadriyah soil (T_2)	
Soil reaction (pH)		7.30	7.37	-
Electrical conductivity (EC)		1.84	1.48	$ds.m^{-1}$
CEC		23.45	19.18	$cmol.g^{-1}$
Organic matter O.M		8.23	7.71	$gm.kg^{-1}$
Gypsum		0.26	0.15	$gm.kg^{-1}$
Carbonate minerals		221.12	180.00	$gm.kg^{-1}$
Dissolved positive ions				
Calcium Ca^{2+}		5.20	4.60	$mmol.L^{-1}$
Magnesium Mg^{2+}		3.60	3.10	$mmol.L^{-1}$
Sodium $^{+}$		0.98	0.85	$mmol.L^{-1}$
Potassium $^{+}$		0.63	0.58	$mmol.L^{-1}$
Dissolved negative ions				
Carbonates CO_3^{2-}		Nill	Nill	$mmol.L^{-1}$
Bicarbonate HCO_3^{-}		1.78	1.40	$mmol.L^{-1}$
Sulfates SO_4^{2-}		1.38	0.91	$mmol.L^{-1}$
Chloride Cl^{-}		14.20	11.40	$mmol.L^{-1}$
Available Nitrogen		28.31	20.81	$mg.kg^{-1}$
Available Phosphorus		16.36	16.08	$mg.kg^{-1}$
Available Potassium		76.71	69.33	$mg.kg^{-1}$
Field capacity		0.33	0.24	$Cm^3 Cm^3$
Wilt point		0.19	0.10	$Cm^3 Cm^3$
Analysis of minute volumes	Sand	146.00	392.00	$gm.kg^{-1}$
	Silt	554.00	400.00	$gm.kg^{-1}$
	Clay	300.00	208.00	$gm.kg^{-1}$
Texture		Silty Clay Loam	Loam	——

Table 3: Concentrations of total and available heavy elements ($mg.kg^{-1}$).

Elementmg.kg^{-1}	Element Pictures	Class of texture	
		SiCL	L
Lead	total	32.55	26.32
	available	4.33	2.14
Zinc	total	25.24	20.18
	available	3.47	1.72
Nickel	total	21.34	13.91
	available	0.87	0.62
Cadmium	total	1.01	0.92
	available	0.21	0.81

Table 4: Concentrations added from heavy metals to soil during plant growth.

Class of texture	Amount of water added (L)	Type element and concentration $mg.kg^{-1}$			
		Pb	Zn	Ni	Cd
SiCL	12.5	6.25			
L	11.50	5.50			

lowest value for all elements reached 2.42, 2.01, 1.20 $mg.Kg^{-1}$ soil respectively, while the control treatment (M_0) gave highest value of the above previous elements was 4.07, 3.84, 2.12 $mg.Kg^{-1}$ soil respectively, cause of superiority of the treatment of active charcoal by giving it the lowest average effect of elements in the form of available in the study soil compared to other treatments to contain charcoal cavities or internal pores have a high surface area that has the ability to adsorb more pollutants than other materials, This results corresponds to his findings Gyaath (2012) and Lucas and Cocero (2003) Who showed that's to increase the number of negative charges on active surfaces of active charcoal, this makes it more adsorbent to the elements from the other material this is agree with what he brought Mohammed *et al.,* (2018) when studying use the active carbon surface and Bentonite metal to adsorb Lead ions show that the negative values of "G° considered as indicator to natural adsorption of Lead ions. "G° value on the surface of active charcoal it was higher than the value ("G°) on the bentonite metal surface.

The effect of soil texture (T) on the values of concentrations of available heavy metals (Lead, Zinc, Nickel and Cadmium), showed the results of tables (5, 6, 7 and 8) that there was a significant effect in reducing the concentrations of all heavy metals due to the difference in soil texture except available Nickel soil texture did not significantly affect its values, given the texture (T_1) category SiCL less value for available heavy metals, while gave the texture (T_2) type (L) highest value of the available heavy metals in the soil, the low values of available heavy metals in the soil of Abu Ghraib (T_1) for the type (SiCL) because The adsorption process is a superficial phenomenon and a characteristic for solid surface qualities, and increases the surface area for mineral colloids increases the adsorption process and

Table 5: Effect of soil materials and texture on available Lead concentrations in(mg Pb. Kg^{-1}).

	Texture class	Processing materials (M)					Average effect (T)
		M_0	M_1	M_2	M_3	M_4	
Soil texture (T)	T_1 (SiCL)	14.12	11.60	10.12	9.15	11.80	11.35
	T_2 (L)	14.41	13.34	10.63	9.86	12.10	12.06
$LSD_{0.05}$		0.01					0.007
Average effect (M)		14.26	12.47	10.37	9.50	11.95	
$LSD_{0.05}$		0.01					

Table 6: Effect of soil texture and processing materials on available Zinc concentrations in soil. ($mg.kg^{-1}$).

	Texture class	Processing materials (M)					Average effect (T)
		M_0	M_1	M_2	M_3	M_4	
Soil texture (T)	T_1 (SiCL)	3.97	3.24	2.36	2.41	3.01	2.83
	T_2 (L)	4.17	3.15	2.49	2.66	3.41	3.34
$LSD_{0.05}$		0.01					0.007
Average effect (M)		4.07	3.19	2.42	3.53	3.21	
$LSD_{0.05}$		0.01					

Table 7: Effect of soil texture and processing materials on available Nickel concentrations in soil. ($mg.kg^{-1}$).

	Texture class	Processing materials (M)					Average effect (T)
		M_0	M_1	M_2	M_3	M_4	
Soil texture (T)	T_1 (SiCL)	3.70	2.62	1.89	2.08	2.62	2.58
	T_2 (L)	3.98	2.86	2.13	2.27	2.86	2.82
$LSD_{0.05}$		0.31					N.s
Average effect (M)		3.48	2.74	2.01	2.17	2.74	
$LSD_{0.05}$		0.22					

Table 8: Effect of soil texture and processing materials on available Cadmium concentrations in soil. ($mg.kg^{-1}$).

	Texture class	Processing materials (M)					Average effect (T)
		M_0	M_1	M_2	M_3	M_4	
Soil texture (T)	T_1 (SiCL)	2.10	1.68	1.18	1.24	1.80	1.60
	T_2 (L)	2.15	1.89	1.23	1.32	1.93	1.70
$LSD_{0.05}$		0.01					0.007
Average effect (M)		2.12	1.78	1.20	1.28	1.86	
$LSD_{0.05}$		0.01					

Table 9: Effect of soil texture and processing materials on total Lead concentrations in soil (mg Pb. kg^{-1}).

	Texture class	Processing materials (M)					Average effect (T)
		M_0	M_1	M_2	M_3	M_4	
Soil texture (T)	T_1 (SiCL)	20.28	21.84	27.67	25.72	22.12	23.52
	T_2 (L)	15.21	16.94	22.37	21.10	17.41	18.60
$LSD_{0.05}$		0.08					0.03
Average effect (M)		17.74	19.39	25.02	23.41	19.76	
$LSD_{0.05}$		0.05					

according to this concept the clay with a high surface area increases the adsorption process and the binding strength of the ions (Elzing and Sparks, 1999), and the current study results explains the higher removal rate in soil with texture (T_2) compared with soil (T_1) because the cations exchange capacity of positive ions of soil (T_1) is larger than the cations exchange capacity of positive ions (T_2), which reduces the binding strength of Lead ions in the texture (T_2) thus increasing the amount of ions removed, This is consistent with AL-gawwam and Al-Robai (2016) whom showed in their study when Lead ions were removed for two calcareous soils after their contamination at different concentrations of heavy metals, after removal of Lead the study showed that the Lead ion removal rate was higher in sandy texture soil compared with clay loam soil at all pollutant concentrations. The results of tables 5, 6, 7 and 8 showed that the bilateral interference between T × M had a significant effect on the values of concentrations of available heavy metals (Lead, Zinc, Nickel and Cadmium). the treatment (T_1M_3) gave the lowest value, while control treatment for T_2 gave concentration of metals higher values for all elements compared to other treatments.

Total concentration of heavy metals

The results of tables 9, 10, 11 and 12 showed that the processing materials (M) had a significant effect on increasing the concentrations of the total heavy metals in the soil (Lead, Zinc, Nickel and Cadmium), The active charcoal treatment (M_2) exceeded by giving the highest values compared to other treatments and for all elements (25.02, 24.63, 20.12 and 4.72 mg. kg^{-1} soil) respectively. While the control treatment (M_0) gave the lowest concentrations 17.74, 17.11, 14.48 and 2.85 mg. kg^{-1} soil respectively. We show from the results of tables 9, 10,

Table 10: Effect of soil texture and processing materials on total Zinc concentrations in soil (mg Zn. kg^{-1}).

	Texture class	Processing materials (M)					Average effect (T)
		M_0	M_1	M_2	M_3	M_4	
Soil texture (T)	T_1 (SiCL)	18.10	19.12	29.10	27.52	20.47	22.86
	T_2 (L)	16.12	21.00	20.16	19.60	17.73	18.92
$LSD_{0.05}$		0.07					0.03
Average effect (M)		17.11	20.06	24.63	23.56	18.92	
$LSD_{0.05}$		0.05					

Table 11: Effect of soil texture and processing materials on total Nickel concentrations in soil (mg Ni. kg^{-1}).

	Texture class	Processing materials (M)					Average effect (T)
		M_0	M_1	M_2	M_3	M_4	
Soil texture (T)	T_1 (SiCL)	16.80	20.25	21.72	20.36	17.85	19.39
	T_2 (L)	12.17	18.80	18.52	17.84	13.10	16.08
$LSD_{0.05}$		0.07					0.03
Average effect (M)		14.48	19.25	20.12	19.10	15.47	
$LSD_{0.05}$		0.05					

Table 12: Effect of soil texture and processing materials on total Cadmium concentrations in soil (mg Cd. kg^{-1}).

	Texture class	Processing materials (M)					Average effect (T)
		M_0	M_1	M_2	M_3	M_4	
Soil texture (T)	T_1 (SiCL)	3.00	3.06	5.14	3.88	3.10	3.63
	T_2 (L)	2.70	2.82	4.31	3.66	2.92	3.28
$LSD_{0.05}$		0.02					0.01
Average effect (M)		2.85	2.94	4.72	3.77	3.01	
$LSD_{0.05}$		0.01					

11 and 12 that there is an increase in the values of concentrations of the total heavy metals when adding processing materials (M) compared with do not add The reason is due to the ability of these materials in the formation of complexes with these elements and constrained compatible with Yobuet *et al.,* (2010) in their study that showed the elements are often associated with organic matter and mineral hydroxides in different proportions differently on the type of add matter, thus the addition of their materials have an active role in the stability and sedimentation of heavy metals present in the soil.

The effect of soil texture (T) on the concentrations of heavy metals in their total forms in the soil under study (Lead, Zinc, Nickel and Cadmium), Showed in tables 9, 10, 11 and 12 that there was a significant effect on the values of the concentrations of all the heavy metals due to variation in soil texture, T_1 (SiCL) gave the highest values for total heavy metals concentrations of the above mentioned, while the (T_2) type (L) gave the lowest values for the concentration of the total heavy metals in the soil, we note from the results of the tables above that the soil texture with a high content of clay was a role in increasing the concentrations of total heavy metals and this is due to the role of clay in the retention of elements and convert them from the available to non-available with the possibility of increasing the bonding energy This is consistent from Julid and Resat (2006) in their study they showed that increasing the clay a contributed to increasing the concentration of total heavy metals, Al-Obaidi (2018) explain also that the size of soil has effective role in the fixation and increased binding energy of Lead ions, he explained that both the capacity of the soil to bind these ions and energy binding it decreased with the reduction of the amount of soft minutes in the Specific reaction and this consistent with Mandznieva *et al.,* (2014).

The results of the above tables showed that the bilateral interference between T × M had a significant effect in increasing the concentrations of the total heavy metals (Lead, Zinc, Nickel and Cadmium), she gave the treatment (T_1M_2) gave highest value for all total heavy metals, while control treatment for (T_2) and all elements gave the lowest values compared to other treatments.

Table 13: Efficiency of add material in the total concentration of heavy metals in soil (%).

Sample code	Soil texture	Lead	Zinc	Nickel	Cadmium
M_0	T_1	47.73	42.52	39.10	58.67
M_1		43.71	39.28	26.60	57.82
M_2		28.68	7.58	21.27	29.20
M_3		33.71	12.60	26.20	46.55
M_4		42.98	34.99	35.30	57.30
M_0	T_2	52.19	37.22	37.30	57.94
M_1		46.76	21.22	4.14	56.07
M_2		29.69	18.49	3.58	32.86
M_3		33.68	23.67	8.08	42.99
M_4		45.28	30.95	32.50	54.51

Efficiency of usd materials in the available and total concentration of heavy metals

Table 14 shows the efficiency of the processing materials (M) in the total concentrations of heavy metals (Lead, Zinc, Nickel and Cadmium) for the study soil, the treatment of active charcoal (M_2) for texture T_1 and T_2 compared to other treatments gave it a less efficiency for the total concentration of all heavy elements and this is due to the reasons described earlier in the role of active charcoal in increasing the total of heavy metals.

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