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Potential Source of Heavy Metals in the Geophagic Clay (Marl) and Its Implication on Human Health in NE Iraq: A Pilot Study

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Abstract

Geophagy is the eating of non-food earthy matters practice (such as clay), which is performed especially in humans, to augment a scanty or mineral-deficient diet or as part of a cultural tradition. Geophagy, the intentional ingestion of clay or soil, is a prevalent practice among animals and humans, especially is more common in young ages, such as children and among pregnant women with a young age. In spite of some well documented cases, the motivations of this practice and its consequences on the status of health of the consumer are still unclear. This study focused on the source of heavy metals and its health impact because of the importance of the topic in the environmental hazards, especially as this addictive phenomenon may not be well known among families in our societies. During this study, twenty-four samples of marl were collected within the rock sequences of the Fatha Formation in the Kirkuk and Diyala, which are used in eating (mud eating disease) in these regions. The samples were subjected to laboratory studies including chemical analyses, to determine their contents of trace elements. The laboratory analytical data showed that these muds (marl) contain different concentrations of chemical elements such as (Co, Cr, Ni, Cu, and Zn) which are among the heavy toxic elements and harmful to human health. A close look at the analytical data and results of the statistical and environmental factors, it is found that the marl (geophagia) consumption in terms of its content of trace elements, poses health risks to the consumer. However, it is worth mentioning to say that Geophagia practice may contribute large levels of the most beneficial elements such as Ca, Fe, K, Na, Cu, and Zn, supplying in demand mineral nutrients in the human body.

Keywords: Geophagia; Marl; Iraq; Mineralogy; Environmental indices; Health impact

1. Introduction

The term geophagy (or Geophagia) has been applied in 1850 to humans and animals' practice of soil consuming, it is a type of pica which refers when a person craves or eats nonfood items, such as earth eating conduct and has been accounted for among pre-school kids, pregnant ladies, and it rarely occurs in adults (Reid, 1992; Henry and Cring, 2012; Gupta, 2019; Al-Hamdani et al., 2016). Even though this practice is found in all continents, it is most trendy seen in the tropics, particularly in tropical Africa (Dissanayake & Chandrajith, 2009). The evidence suggested that the practice is not limited to poor societies but cuts across socio-economic, religious, ethnic and racial divides (Ekosse & Jumbam, 2010). Typically, only specific soils are consumed and are chosen for wanted characteristics for instance

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color, flavor, odor, plasticity and texture (Vutchkov et al., 2013). There is a relationship between geophagia and pregnancy, including medical causes, beliefs and traditions. In southern Africa, a pregnant woman thinks that eating clay is beneficial for the fetus because it makes their skin smooth and beautiful (Ekosse & Jumbam, 2010). In Iraq, some pregnant women eat mud to have a healthy baby. Clay is not only eaten by pregnant women; it is also eaten by some people to treat stomach diseases and ulcers and is used as a treatment for hair loss and clear skin (Monaco et al., 2019).

The main reasons for geophagia are to detoxify harmful or unpalatable compounds in the diet, relieve gastrointestinal disturbances such as diarrhea, supplement the diet with mineral nutrients, and relieve excess acidity in the digestive system (Wilson, 2003) and it can be a good source of calcium, manganese and magnesium (Hooda et al., 2004). However, it may also be harmful, by causing intestinal effects, reducing the bio-availability of beneficial micronutrients, and introducing toxic substances such as lead or by acting as a carrier of geohelminth infection (Pain et al., 2018). Depending on the amount and type of substance being eaten, complications such as malnutrition, developmental delay, parasitic infections and poisoning can be observed (Çakar and Şener, 2009). In addition to that it causes a decrease in iron absorption thus leads to a decrease in hemoglobin and severe anemia (Hooda et al., 2002; Alanazi et al., 2018). It was observed by visiting the health centers and meeting with a group of doctors in these centers in Kirkuk and Diyala governorates, that there are sick cases related to the practice of eating mud among some of the patients, especially women and children. Accordingly, we decided to conduct this study to show the extent of the impact of the chemical content of these clays and the problems resulting from eating them on public health. Whereas, there are no local studies about this topic at the present time, so this is the first paper dealing with the study of this field. The objectives of this study are determining the chemical compositions of marl and mud rocks of the Fatha Formation that are used for eating (geophagia); and evaluate the trace elements contamination levels based on geo-accumulation index (Igeo) and calculation of oral intake of trace elements from geophagic marl by using health risk index (HRI).

2. Location and Geological Setting

The study area is located in northeastern Iraq within the low folded zone of the unstable shelf of the tectonic division of Iraq at Kirkuk and Diyala (Fig 1). It is represented by marl outcrops of the Fatha Formation. The study area includes specific locations from the Laylan area, which is 19 km southeast of Kirkuk, and the Kiwan region, 10 km northwest of Kirkuk, and includes specific sites from the Kifri region, which is about 100 km north of Diyala. Kirkuk is about 9679 km, and it is located geographically between the longitude (44°18' 0") and (44°26' 30") and latitude (35°31' 30") and (35°22' 30"), its average height above the sea level is about 300 m. Diyala is about 17685 km, and it is located geographically between the longitude (44°30'00") and (46°12'00") and latitude (33°12'30") and (35°46'15") and its average height above the sea level is about 232 m.

The area consists of several geological formations, the most important of which are the Fatha, Injana, Mukdadiya and Bai-Hassan formations (Fig. 1). The Fatha Formation consists of limestone, marl, anhydrite, gypsum, and fine-grained clastic, with 900 m thickness in the center of the depositional basin, (Al-Naqib, 1960). It is deposited in the environment of high salinity coastal lakes, and this is reflected by the presence of gypsum layers and the presence of fossils in the limestone rocks, whereas, the abundance of shell fragments that form the barriers and ridges reflects a calm (low energy) marine environment, while the soil crumb material increases and evaporation decreases when the formation is deposited near the coast. The Injana Formation is characterized by the heterogeneity of lithology, but the basic units consist largely of marlstone or mudstones and siltstones of red or gray color, as well as sandstone layers with granular sizes ranging from medium to coarse. The Mukdadiya Formation consists mainly of fine gravel deposits and coarse-grained sandstone and siltstone, pebbly sandstone and

conglomerate as thin lenses in the upper part of the formation. Bai Hassan Formation is very similar to the deposition of the Mukdadiya Formation, but they differ in the granular size of the deposit, as the deposition of the Bai Hassan Formation in general, have large sizes.

Fig.1. Geological map of northern Iraq showing the location of the studied area

3. Materials and Methods

3.1 Sampling and Chemical Analysis

Twenty-four samples of exposed marl were collected from the Fatha Formation, including five samples from Laylan region, five samples from the Kiwan region belonging to Kirkuk, six samples from the Kifri region of Diyala and eight samples from the markets, five of them from Dibs and three from Kifri (Table 1). The geophagy marl was analyzed for Co, Ni, Cu, Cr and Zn using the energy dispersive X-ray fluorescence (PEDXRF) techniques in Turkey. Samples were dried by the sun and then ground into fine powder in agate mortar. Samples were sieved to pass through of 200 μm , and then pressed into thick pellets of 32 mm diameter using watch as blinder. USGS standards, GBW 7109, GEOL and GBW-7309 Sediment equally pressed into pellets in a similar manner as the samples, and these used for quality assurance (Timothy and La, 1989; Johnson et al., 1999).

3.2 Assessment Indices

3.2.1. Geoaccumulation Index (I_{geo})

This indicator is used to determine the pollution of trace elements in sediment samples (Haris et al., 2017). The equation was presented by Muller (1979) as a measure of the severity of pollution, by comparing the current concentration with background levels. It can be calculated according to the following equation (Muller, 1979):

$$I_{geo} = \log_2 \frac{C_n}{B_n \times 1.5} \quad (1)$$

Where C_n is the measured concentration of trace element in the marl sample; B_n is the reference value or geochemical background concentration of the trace element (Mason, 1966), and the factor 1.5

in this equation is to reduce the effect of possible differences in background values Bn (Lar et al., 2014). The index of Geo-accumulation (Igeo) is classified into seven classes by Muller (1969) (Table 2).

Table 1. Location sites in the studied area

samples No.	Site	Samples Name	Latitude	Longitude
1	Kirkuk	Lay1	35° 23'45.50"	44°29'01.80"
2		Lay2	35° 23'46.90"	44° 29'01.00"
3		Lay3	35°23'45.30"	44° 29'04.30"
4		Lay4	35° 23'45.50"	44° 29'04.40"
5		Lay5	35° 23'43.38"	44° 29'04.71"
6		KO1	35° 35'25.50"	44° 14'45.30"
7		KO2	35° 35'27.30"	44° 14'38.40"
8		KO3	35°35 '26.80"	44° 14'37.10"
9		KO4	35° 35'27.52"	44° 14'42.39"
10		KO5	35° 35'24.22"	44° 14'40.51"
11	Diyala	Kif1	34°36'48.00"	45°0'31.10"
12		Kif2	34° 36'49.02"	45° 0'37.30"
13		Kif3	34° 40'49.34"	45° 0'11.21"
14		Kif4	34° 40'48.45"	45° 0'09.22"
15		Kif5	34° 40'07.66"	45° 0'30.38"
16		Kif6	34° 39'37.78"	45° 0'15.47"

Table 2. Classification of Geo-accumulation index (Muller, 1979)

Class	I _{geo} value	Description
0	$0 \geq I_{geo}$	Unpolluted
1	$1 \geq I_{geo} > 0$	Unpolluted to moderately polluted
2	$2 \geq I_{geo} > 1$	moderately polluted
3	$3 \geq I_{geo} > 2$	moderately to strongly polluted
4	$4 \geq I_{geo} > 3$	strongly polluted
5	$5 \geq I_{geo} > 4$	strongly to extremely polluted
6	$5 < I_{geo}$	extremely polluted

3.2.2. Health Risk Index Determination (HRI)

The Evaluation of health risk potential was determined by consuming geophagy marl using the Health Risk Index (HRI) (US EPA 1989). The health risk index is the ratio between exposure to specific trace element and the Oral Reference Dose (RfD) for such an element, which provides an indication of the level of health risks from exposure to pollutants (Lar et al., 2014). In this case, it is obtained by the equation:

$$HRI = \frac{DIC * C}{RD * BW} \quad (2)$$

Where DIC is considered to be 0.02 kg/day; C is the measured trace element concentration in the geophagical marl (in ppm); RD is the oral reference dose for the trace element (mg/kg of body weight/day) and BW is the average human body weight (kg).

4. Results and Discussion

4.1. Concentration of Geochemical Elements and their Assessment Indices

The chemical elements that are naturally present in the human body help it to perform the vital functions which are necessary for the body and its health (Aydogan and Incekara, 2017; Al-Maadhidi, 2019). Cu and Zn, but have toxic effects at elevated concentrations (Wong et al., 2006). Heavy metals are the more toxic trace element, they include: As, Pb, Hg, Cr and Cu (Albayati, 2017). All these elements are derived through ingestion, inhalation and dermal. The effects of regional geology and processes related with soil-forming of an area have a direct impact on the chemical composition of the geophagic marl (Lar et al., 2014). In addition, human activities are among the most common sources of trace elements, such as urban road construction, agriculture, waste burning (Abaha et al., 2014). The results of the chemical contents of the studied geophagy marl are shown in Table 3, the geo-accumulation index (Igeo) values are illustrated in Table 4 and HRI values for the trace elements are presented in Table 5.

Table 3. The concentration of major and trace elements in studied samples.

ppm	Al	Ca	Mg	Fe	Na	Co	Ni	Cu	Cr	Zn
Max	60175.73	170598.9	49876.37	52701.62	3635.11	17.34	115.38	235.43	89.17	112.41
Min.	37523.83	142797.1	26657.02	30470.55	1261.16	12.02	110.01	28.92	49.84	73.49
Ave.	46360.09	156248.3	38824.56	38862.26	2203.94	13.97	112.81	137.12	64.423	84.81

Table 4. Geo-accumulation index in studied marl samples

	Al	Ca	Mg	Fe	Na	K	Co	Ni	Cu	P	Cr
Max	0.334	2.122	1.078	0.476	0.058	0.291	0.230	0.680	1.863	0.220	0.305
Min.	0.208	1.776	0.576	0.275	0.020	0.126	0.219	0.662	1.034	0.262	0.235
Ave.	0.258	1.943	0.839	0.351	0.035	0.189	0.238	0.663	0.372	0.206	0.227

Table 5. Health risk index in studied Marl samples for children and adults

		Co	Ni	Cu	Cr	Zn		Co	Ni	Cu	Cr	Zn
Max.		1.084	7.211	7.357	37.15	0.468		0.248	1.648	1.682	8.492	0.107
Min.	Child	0.751	6.876	0.904	20.77	0.306	Adult	0.172	1.572	0.207	4.747	0.07
Ave.		0.873	7.05	4.285	26.84	0.353		0.2	1.612	0.979	6.136	0.081

4.2. Major Elements Distribution in Geophagic Marl

Ca and Al display relatively higher concentration than the other elements with average 156248 ppm and 46360 ppm, respectively (Table 3), depending on the region that it came from (Othman et al., 2020). Fe, Mg, Na and K average concentration are 38862, 38825, 2204 and 10813 ppm respectively (Table 3). Apparently, the levels of the major element (Ca, Al, Fe, Mg, Na and K) concentrations determine their benefits and harmfulness when digested into the system of the human body. The geo-accumulation index (Igeo) for these chemical elements display values (Igeo > 1) higher than 1 for Ca, and less than 1 for others, which indicate that the studied marl samples are uncontaminated by these chemical elements except Ca was contaminated (Table 4).

4.3. Trace Elements Distribution in Geophagic Marl

The trace elements like Cr, Co, Cu, Ni and Zn display average concentration of 65.65, 14.23, 131.73, 112.99 and 85.61 ppm, respectively (Table 4). All of these chemical elements have geo-accumulation index (I_{geo}) values less than 1 and are below contamination levels except Cu which show values more than 1 and can be considered as a contamination hazard. Their trace elements concentration levels in the marl are therefor in acceptable quantities and could be considered as a significant source of benefit mineral nutrient that can complement food source but attention should be paid to the Cu high concentrations which can be harmful.

4.4. Potential Human Health Risks

Co and Zn show an HRI less than 1, which means that there are no potential problems with these two elements. Whereas, Cu, Cr and Ni values of HRI are more than 1, which mean it can cause a serious health problem for children (Table 5). For adults Co, Cu and Zn values are less than 1, while Ni and Cr values of HRI are more than 1 (Table 5). Cr tends to cause kidney failure, lung cancer, and dermatitis (Adriano, 2001; Cefalu and Hu, 2004). Also, excess Cu has adverse effect on physiological processes such as: renal insufficiency, nausea and diarrhea (Oyebanjo et al., 2020, Awadh, 2015). Petzold and Al-Hashimi (2011) explained that the Ni plays an important role in the iron absorption by the human body and is vital for building vigorous bone structures, while the toxicity of Ni can lead to miscarriages and musculoskeletal defects (Vaktskjold et al. 2008). Zinc is generally considered non-toxic, but it can cause dehydration, vomiting, abdominal pain, nausea, dizziness, decreased muscle coordination, and kidney failure (Ekosse and jumbam, 2010).

5. Conclusions

Geophagic marl consumption poses health risks to the consumer in terms of its trace elements content. The practice of geophagia contributes to large levels of beneficial elements such as Fe, Ca, K, Na, Cu and Zn, consequently providing the deficient mineral nutrients that the human body requires it. The Geo-accumulation values of Ca, Mg, and Cu are more than 1, which means the geophagic marl is contaminated with them. But geophagic marl is uncontaminated with Al, Fe, Na, K, Co, Ni, Cr and Zn, which values are less than 1. Co and Zn values of HRI are less than 1, which means they have not any adverse health effects. HRI values for Cu, Cr and Ni are more than 1 for children. For adults Co, Cu and Zn values are less than 1, Ni and Cr values are more than 1. They can cause a serious health problem for practitioners.

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