

Natural Radioactivity and Radon Exhalation in the Sediment River Used in Sulaymaniyah Governorate, Iraq, Dwellings

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Abstract—The samples, sand and pebble (Sediment River), were collected from their selling places in Sulaymaniyah city, Iraq. Sand and pebble emanate each of ^{238}U , ^{232}Th , ^{40}K , and radon naturally. The radon concentration in the air (C_0) and gamma-specific activity (A_s) of the samples were measured using passive detectors (CR-39) and gamma spectrum analyzer (3×3 NaI (TI) connected to digital spectrum analyzer), respectively. The mean value of the $C_0 \pm$ deviation of the investigated samples was 176.5 ± 6.9 Bq/m³, which is lower than the worldwide value of 300 Bq m⁻³ reported by the IAEA. However, an anomaly in C_0 value was found in Tanjaro (Sand) sample. The mean value radon mass exhalation rate (E_M) was 10.9 ± 0.5 $\mu\text{Bq/kg/h}$. The second part of the measuring is the gamma-specific activities (A_s) of each ^{226}Ra , ^{232}Th , and ^{40}K . The A_s values were ranged (4.3 ± 0.2 – 22.6 ± 0.2), (1.9 ± 0.1 – 4.2 ± 0.1), and (39.8 ± 0.2 – 86.0 ± 0.4) Bq/kg, respectively. The mean calculated value of radium equivalent (Ra_{eq}) was 20.2 ± 0.2 Bq/kg. The annual effective dose rate of gamma (H_{ann}) was 0.13 mSv/y. Fortunately, the H_{ann} value of the samples is lower than the world average value of 0.48 mSv/y.

Index Terms—Building Materials, Gamma-Specific Activity, Gamma/Radon Radiological Parameters, Radon Mass Exhalation, Sediment River (Sand and Pebble).

I. INTRODUCTION

Sediment river provides valuable information about radiological contamination in the environment. Sand and pebble particles depose on the bottom of lakes, but currents continue to move the relict sediment along riverbank or stream. Sediment river (sand and pebble) is the raw materials used mainly in building construction in Iraq. These materials emanate naturally occurring radioactivity, since they contain long-lived radionuclides such as ^{238}U , ^{235}U , ^{232}Th , with their

progenies, and ^{40}K . The precursors of ^{226}Ra in the ^{238}U series are generally ignored because 98.5% of the radiological effects of the ^{238}U series are produced by the ^{226}Ra and its daughter products (Turhan and Varinliog, 2012).

The natural radioactive nuclides such as radon contribute mainly in radiation exposure of the human population. Radon is a decay product of ^{226}Ra from the natural series decay of ^{238}U with half-life of 3.82 days (Gulfink, 2008). In general, unlike the gas radon itself, radon daughters are solids and stick to surfaces of dust particles in the air. If such contaminated dust inhaled, these particles can stick to the airways of the lung and increase the risk of developing lung cancer (Mass, 2011). Previous studies (Poschl, 2007; Turhan, 2008) reported that natural radioactivity and the terrestrial gamma dose depend effectively on geological and geographical conditions, due to this radon and its progenies depend on the geological and geographical condition too (more information about geographical parameters are required). Much information about the radon emanating from the materials has been recorded in academic studies. High levels of radon are associated with granite igneous rocks, shale, dirty quartz sedimentary rocks, phosphate deposits, and some beach sands, which may contain high levels of radon progenitors (Brill, 1994). All types of building materials such as sand, gravel, concrete, brick, and granite contain potassium, uranium, thorium, and their progenies; they generate a direct radiation exposure (Cetin, et al., 2012). Previous studies reported that the radon and other natural radionuclide traces emanate of sand and gravel (raw building materials) (Banman, et al., 1982; Mansoor, et al., 2005).

A major part of concrete and brick consists of sand and pebble. These materials are commonly used for developing buildings in Iraq. This research work is to assess the inhalation and exhalation doses of the sand and pebble samples. These samples were collected from screen factories established near the rivers and streams of different areas of Sulaymaniyah Governorate. The main objective of the current work is to study the natural radionuclide concentrations for 12 samples. Furthermore, radon concentration \pm deviation in the air (C_0) and the specific activity of gamma (A_s) of ^{238}U (^{226}Ra), ^{232}Th , and ^{40}K was measured using two techniques (CR-39 and NaI (TI) detector - digital spectrum analyzer [DSA]) for

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the collected samples. The potential radiological hazards of radon associated with those materials (sand and pebble) were calculated such as annual radon effective dose ($D_{ann(Rn)}$) and radon mass exhalation rate (E_M). In addition, the gamma dose parameters were calculated as such the radium equivalent (Ra_{eq}), the gamma dose rate (H), the annual effective dose rate (H_{ann}), and the gamma hazard index (I). Unfortunately, according to our knowledge, there are no sufficient data in the literature on natural radioactivity levels (specific activity of gamma, radon concentration) in sand and pebble used in building materials in the studied area.

II. EXPERIMENTAL METHOD

The samples (sand and pebble) were collected from their selling places of Sulaymaniyah city, Iraq. The selling places bring sand and pebble from their screening factories, these screening factories are established near the different rivers and streams, covered the area of about 3300 km². The rivers and stream discharge in a two main lakes Dukan and Darbandikhan located in Sulaymaniyah Governorate.

A. Geological description

radon concentration in the sediment river can be predicted by studying the lithology and deposing the weathering part of the sample. The previous study classified the samples (Table I) used in this work as can be seen elsewhere (Kamal, et al., 2011).

B. Radon concentration and exhalation measurement

the passive (SSNTD) detector uses in a wide range of long-term measuring of radon in the worldwide, the previous researchers used it for different purposes as mentioned in Nisha, et al., 2016; Ajay, 2015; and Nguyen, et al., 2005. Eight different types of sediment samples, sand and pebble, were collected from their selling places in Sulaymaniyah city - Iraq to study their radionuclide concentrations. At the laboratory, samples were grinded and sieved with size 1 mm to homogenize their particles and then dried at 110°C temperature in an oven to eliminate the water content. Equal amount of samples (100 g) was placed in the Can – chamber (cylinder chamber), of 7.2 cm diameter by 28.5 cm height as shown in Fig. 1. CR-39 plastic track detector (1 cm × 1.5 cm) was suspended to the selling of Can – chamber, CR-39 made by Track Analysis Systems Ltd - Bristol, United Kingdom. Sufficient pieces of CR-39 detectors were calibrated using standard alpha emitter source (²⁴¹Am) to obtain a linearity response process, fully detail can be seen elsewhere (Kamal, 2013).

The Can_ chambers were sealed tightly with a thick tape to prevent gas escape and then stored for 60 days to obtain radioactive equilibrium between ²²⁶Ra and its decay products of short half-life, and to reduce statistical error while the alpha particles track the detector, can be seen elsewhere (Al-Sharkawy, et al., 2012). The detectors were etched chemically using 6.25 M of NaOH at 70°C for 6 h, to enlarge the alpha tracks. Etched detectors were washed and then dried. The radon

TABLE I
LITHOLOGY DESCRIPTION OF SAND AND PEBBLE SAMPLES (KAMAL, ET AL., 2011)

Samples	Geological composite
Chwarqurna (Pebble)	Dolomite (limestone), some chert (silica)
Darbandikhan (Pebble)	Chert, limestone, igneous
Darbandikhan (Sand)	Chert, limestone, minor igneous
Goptapa (Pebble)	Chert, limestone, some igneous
Goptapa (Sand)	Chert, limestone, minor igneous
Qaladize (Sand)	Igneous, metamorphic, limestone, few chert
Qaladize (Pebble)	Few igneous, chert, limestone
Kani Bee (Sand)	Some igneous, chert, limestone
Said Sadiq (Sand)	Organic-rich limestone, some chert
Sharbazher (Sand)	Chert, limestone, few igneous
Tanjaro (Sand)	Limestone, chert, shale
Tanjaro (Pebble)	Limestone, chert, some shale

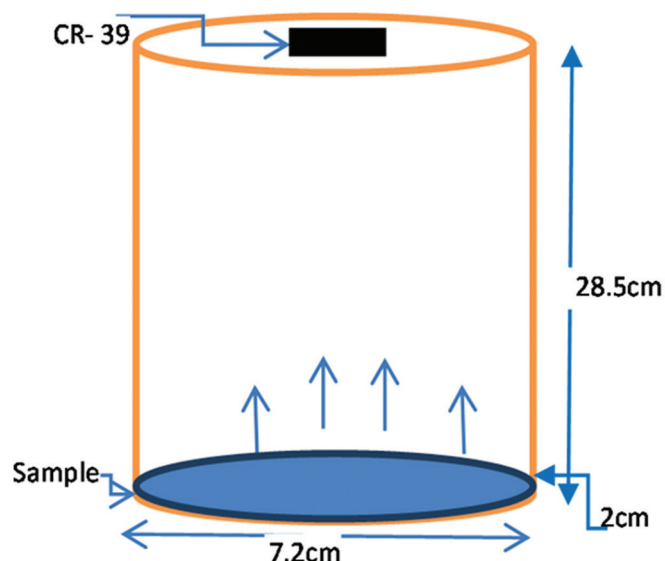


Fig. 1. Schematic diagram of closed cup tube, CR-39 detector was suspended in the upper part of it.

track density (track/cm²) on the CR-39 detectors was counted using an optical microscope (Olympus) with magnification power (×400) that is similar to the work of Kamal, 2013.

The integrated radon concentration in the air was calculated due to the measured track density of the samples of sand and pebble as given in Azam, et al., 1995,

$$\rho = K C_0 T \quad (1)$$

Where, ρ is the track density Tr/cm^2 , K is the calibration factor for cylindrical shape that equals to 0.0577 ($Tr \text{ cm}^{-2}/d/Bq \text{ m}^{-3}$). Calculation of K same as used in Kamal, 2013 and Barillon, et al., 1993. C_0 is integrated radon exposure inside the closed cup tube ($Bq \text{ cm}^{-3} \text{ h}^{-1}$) and T -radiate time (60 days).

C. Gamma emitters' measurement

the samples of sand and pebble were prepared for gamma spectroscopy measurements. The collected samples were crushed and sieved to <1 mm particle size. Each sample was

homogenized and dried for 24 h in an air circulating oven at 110°C to ensure that moisture was completely removed. About 1000 g of each sample was stored in polyethylene containers (Marinelli beaker) and sealed for 3 weeks before analyzing for ⁴⁰K, ²²⁶Ra, and ²³²Th spectrums.

The gamma spectrum analyzer (NaI (Tl) with DSA 1000) has been used as a suitable radiation measuring technique found in Ramasamy, et al., 2011; Ali, et al., 2015; and Laith, et al., 2011. The NaI (Tl) detector specified by model (3 in × 3 in) produced by Canberra industries INC., with an energy resolution of about 7.5% at the 662 keV peak of ¹³⁷Cs, the detector was interfaced to the DSA-1000 made by Canberra industries INC. The detector was surrounded by a cylindrical lead shield (10.0 cm thick and 40 cm height) fixed on steel holder and covered by a thick (5 cm) lead to reduce the background radiation. The energy calibration of the gamma spectrometry systems was carried out using standard radionuclides ¹³⁷Cs and ⁶⁰Co. Furthermore, the counting and signal process has been done for each sample in a 21,000 second period. The spectroscopy information includes count versus channel, peak searching, net peak evaluation, energy/efficiency calibration, nuclide identification, and net count area. The photopeak at 1460 KeV was used for the measurement of ⁴⁰K, whereas those at 1760 KeV peak from ²¹⁴Bi and 2614 KeV from ²⁰⁸Tl were used for the measurement of ²²⁶Ra and ²³²Th, respectively. The activity concentration (*A_s*) in Bq/kg of ²²⁶Ra, ²³²Th, and ⁴⁰K radionuclides is given by (El-Taher, 2012),

$$A_s = N / [\varepsilon_{(E_\gamma)} * P_\gamma * t * M] \quad (2)$$

Where, *N* is the net peak area of γ -ray energy (*E_γ*), $\varepsilon_{(E_\gamma)}$ is the full-energy peak efficiency for γ -rays, *P_γ* is the γ -ray yield per decay, *t* is the counting live time, and *M* is the dried sample mass in terms of kilogram.

D. Radon exhalation and dose assessment

the rectangular chamber is suitable for measuring the surface exhalation rate (*E_A*). The cylindrical chamber is suitable for measuring the radon mass exhalation rate (*E_M*) (Predrag, et al., 2010 and Sharma, et al., 2003), cylindrical chamber was used in this work; therefore, we focused on *E_M* calculation only. The *E_M* is derived from the *E_A* equation, which found in Barillon, et al., 1993, and the modified formula of *E_M* can be found in Imme, et al., 2014; Kovler, 2006; and Khan, et al., 1992 as,

$$E_M = \frac{C_0 V \lambda}{M [T + 1 / \lambda (e^{-\lambda T} - 1)]} \quad (3)$$

Here, *E_M* is the radon exhalation rate in terms of mass (Bq/kg/h), *V* is the effective volume of the Can container in m³, *C₀* is integrated radon exposure, *T* is the exposure time in hours (h), λ is the decay constant for radon (h⁻¹), and *M* is the mass of sand or pebble sample (100 g).

E. Estimation of gamma dose rate

a common radiological index has been introduced to represent the *A_s* level of ²²⁶Ra, ²³²Th, and ⁴⁰K, which is usually known as radium equivalent activity (*Ra_{eq}*) (Beretka and Mathew, 1985).

$$Ra_{eq} = A_{Ra} + 1.43 A_{Th} + 0.077 A_K \quad (4)$$

Where, *A_{Ra}*, *A_{Th}*, and *A_K* are the *A_s* of ²²⁶Ra, ²³²Th, and ⁴⁰K, respectively, in Bq/kg. In the definition of *Ra_{eq}*, it is assumed that 10 Bq/kg of ²²⁶Ra, 7 Bq/kg of ²³²Th, and 130 Bq/kg of ⁴⁰K produce equal γ -ray dose rate. The *A_s* of ²²⁶Ra, ²³²Th, and ⁴⁰K through conversion factor in units (nGy/h per Bq/kg) results *H* for different materials. According to EC-112, 1999 and Baykara, et al., 2011, the *H* is formulated for the sand and sandstone as,

$$H_{nGy/h} = 0.430 A_{Ra} + 0.666 A_{Th} + 0.042 A_K \quad (5)$$

The annual effective dose rate (*H_{ann}*) in unit (mSv/y) is an important radiation risk parameter, can be formulated from the *H* as (UNSCEAR- annexb, 2000; Caridi, et al., 2015),

$$H_{ann} (mSv / y) = H (nGy / h) * \frac{8760h}{yr} * 0.7 Sv / Gy * 10^{-6} = 0.006132 * H (nGy / h) \quad (6)$$

Where, the number 0.7 (Sv/Gy) is a conversion factor.

Radiation hazard was determined by a hazard index (*I_γ*) of gamma, the maximum range of them was 1 mSv/y. *I_γ* is expressed in terms of the specific activities of ²²⁶Ra, ²³²Th, and ⁴⁰K, and their conversion factors were evaluated in EC,1999 and UNSCEAR, 1993,

$$I_g = \frac{A_{Ra}}{300 B_q / k_g} + \frac{A_{Th}}{200 B_q / k_g} + \frac{A_K}{3000 B_q / k} \quad (7)$$

TABLE II
RADON CONCENTRATIONS IN AIR (C0) IN UNIT , RADON MASS EXHALATION (EM) IN UNIT (BQ/Kg/h), THE RADON EFFECTIVE DOSE (D_{EFF (RS)}) IN UNIT nGY/H, AND ANNUAL EFFECTIVE DOSE (D_{EFF (RS)}) OF RADON IN UNIT MSV/Y OF THE SAND AND PEBBLE SAMPLES WERE CALCULATED

Samples	ρ (Track/cm ²)	C ₀ (Bq/m ³)	E _M (μBq/kg/h)
Chwarqurma (Pebble)	503.04	138.27±6.16	8.6±0.4
Darbandikhan (Pebble)	503.04	138.27±6.16	8.6±0.4
Darbandikhan (Sand)	786	216.06±7.70	13.5±0.5
Goptapa (Pebble)	848.88	233.34±8.00	14.5±0.5
Goptapa (Sand)	345.84	95.06±5.11	5.9±0.3
Qaladize (Sand)	786	216.06±7.70	13.5±0.5
Qaladize (Pebble)	314.4	86.42±4.87	5.4±0.3
Kani Bee (Sand)	848.88	233.34±8.01	14.5±0.5
Said Sadiq (Sand)	534.48	146.92±6.35	9.2±0.4
Sharbazheer (Sand)	597.36	164.20±6.71	10.2±0.4
Tanjaro (Sand)	943.2	259.27±8.44	16.2±0.5
Tanjaro (Pebble)	691.68	190.13±7.23	11.9±0.5
Mean value	641.9	176.45±6.87	10.9±0.46

III. RESULTS AND DISCUSSION

A. Radon measurements

The integrated radon concentration in the air (C_0) \pm deviation can be calculated via Eq. 1, and radon mass exhalation rate (E_M) from Eq. 3. The C_0 value was ranged between 86.4 ± 4.9 and 259.3 ± 8.4 Bq/m³ as shown in Table II. The mean value of C_0 is lower than the worldwide value of 300 Bq/m³, which is given in IAEA, 2018. Furthermore, the C_0 value of the sand samples from the locations Tanjaro, Kani Bee, Qaladize, and Darbandikhan with Goptapa (pebble) was close to the worldwide value. However, the mentioned samples are widely used in construction materials of our governorate (area). The maximum exhaled value of radon (E_M) was 16.2 ± 0.5 (μ Bq/k_g/h) recorded in the Tanjaro (Sand) sample, and the minimum was 5.4 ± 0.3 (μ Bq/k_g/h) returned to Qaladize (Pebble), whereas the E_M mean value was 10.9 ± 0.5 (μ Bq/k_g/h). The high radon dose rate is a result of discharging sewage of Sulaymaniyah city into the Tanjaro stream, which causes to sediment the sand near the Tanjaro factory of screening sand (Rebwar, et al., 2016). This result differs from the E_M value of Tanjaro (pebble), which was much less than Tanjaro (sand) sample. The reason is that the Tanjaro screening factory (sand and pebble) established in a position of collecting most sewage of the Sulaymaniyah city. The sewage containing organic materials, these materials labeled sand particle more than the pebble one. In addition, the heavy metals Cr, Mn, Cu, and Pb were detected in the wastewater of Qalyasan stream (Salih, et al., 2014). These materials are the main sources in increasing radon concentration (Andrew, 2014 and Lookman and Ayser, 2016) because the surface area of the sand covered totally by the organic material, the ratio (surface area/mass) of the sand particle is more than the pebble one.

The mentioned radiology parameters are lower than the values recorded in most previous studies (Zakariya, et al., 2013; Mansour, 2005; and Zakariya, 2017).

B. Γ -ray measurements

table III shows that the A_s value of ²²⁶Ra, ²³²Th, and ⁴⁰K ranged (4.3 ± 0.2 – 22.6 ± 0.2) B_q/K_g, (1.9 ± 0.1 – 4.2 ± 0.1)

B_q/k_g, and (39.8 ± 0.2 – 86.0 ± 0.4) B_q/k_g, respectively. These comparisons based on the specific activities of ²²⁶Ra, ²³²Th, and ⁴⁰K values of 50, 50, and 500 B_q/k_g, respectively, reported by the UNSCEAR, 2008.

The important radiological parameters of gamma were derived from the measured A_s such as the radium equivalent (Ra_{eq}), the gamma dose rate (H), the annual effective dose rate (H_{ann}), and the gamma hazard index (I) were tabulated in Table III. Equations (2, 4-7) were used for calculating the mentioned parameters, respectively. The mean Ra_{eq} value was 20.2 ± 0.2 B_q/k_g, which was lower than the worldwide one 200 B_q/k_g (UNSCEAR, 2008). Fortunately, the minimum value of Ra_{eq} was 7.4 ± 0.2 B_q/k_g recorded in Darbandikhan (pebble); furthermore, these raw materials are widely used in construction building in Sulaymaniyah Governorate. In addition, the mean value of I was 0.17 which is less than the unity (EC, 1999). The I value of Darbandikhan (pebble) was 0.08, whereas in Darbandikhan (sand) was 0.19, which is greater than the calculated mean value. The reason is that the heavy metals (⁴⁰K, Ti, Mn, Fe, Mg, Si, P, and Ca) were detected in the Darbandikhan dam as reported in Adeeb, et al., 2017, another fact is that the fine particles (sand) deposit faster than the coarse one (Rebwar, et al., 2016).

In addition, the mean values of the H and the H_{ann} were 21.9 nGy/h and 0.13 mS_v/y, respectively. The H_{ann} values of the samples are low than the worldwide average value of 0.48 mS_v/y (UNSCEAR, 2000 and Darwish, et al., 2015). The measured specific activity of the ²²⁶Ra value is close to the ²²⁶Ra value found in Zakariya, 2017; Kamal, et al., 2014; and Imme, et al., 2014. The measured H and H_{ann} values are lower than the values of the previous studies (Ramasamy, et al., 2011 and Ramasamy, et al., 2014). The reason is that the measured natural radionuclides dose with NaI (3X3) detector is always less than the actual value, because only three energy peaks (1460 KeV ⁴⁰K, 1760 KeV ²¹⁴Bi, and 2614 KeV ²⁰⁸Tl) of the sample spectra were calculated based on the method that allows to calculate just three energy peak of the measured spectra of the sample.

TABLE III

THE RADIUM EQUIVALENT (Ra_{eq}) ACTIVITIES IN BQ/K_g AND THE GAMMA DOSE RATE (H) IN nGy/h, THE ANNUAL EFFECTIVE DOSE RATE (HANN) IN mS_v/y AND THE HAZARD INDEX (I) OF GAMMA-RAY WERE CALCULATED BASED ON THE SPECIFIC ACTIVITIES (A_s) OF ²²⁶RA, ²³²TH, AND ⁴⁰K, FOR THE SAND AND PEBBLE SAMPLES

Sample	Specific activities (A) in B _q / k _g			²²⁶ Ra Equivalent B _q /k _g	Hazard index (I)	H (nGy/h)	H _{ann} (mSv/y)
	⁴⁰ K	²³² Th	²²⁶ Ra				
Chwarqurna (Pebble)	41.95±0.25	2.41±0.04	22.59±0.2	26.37±0.26	0.20	26.80	0.16
Darbandikhan (Pebble)	49.47±0.27	1.85±0.05	4.34±0.16	7.36±0.23	0.08	9.98	0.06
Darbandikhan (Sand)	72.62±0.31	3.57±0.05	16.33±0.17	22.00±0.25	0.19	24.76	0.15
Goptapa (Pebble)	48.74±0.27	3.02±0.05	14.61±0.16	19.31±0.23	0.16	20.67	0.13
Goptapa (Sand)	62.37±0.30	3.56±0.05	15.03±0.16	20.61±0.24	0.18	22.74	0.14
Qaladize (Sand)	85.99±0.36	4.18±0.06	17.57±0.17	24.22±0.25	0.22	27.65	0.17
Qaladize (Pebble)	52.24±0.28	3.07±0.05	14.17±0.15	18.96±0.22	0.16	20.59	0.13
Kani Bee (Sand)	67.32±0.32	3.72±0.05	16.04±0.16	21.88±0.24	0.19	24.23	0.15
Said Sadiq (Sand)	41.59±0.25	2.44±0.04	15.94±0.17	19.75±0.23	0.16	20.68	0.13
Sharbazheer (Sand)	53.88±0.28	2.99±0.05	14.63±0.16	19.31±0.24	0.16	21.05	0.13
Tanjaro (Sand)	41.15±0.25	2.62±0.04	19.48±0.19	23.55±0.25	0.18	24.10	0.15
Tanjaro (Pebble)	39.83±0.24	2.51±0.04	14.84±0.16	18.73±0.23	0.15	19.60	0.12
Mean value	54.76±0.28	2.99±0.05	15.47±0.17	20.17±0.24	0.17	21.90	0.13

IV. CONCLUSIONS

The radium isotope contributed mainly in specific activity of gamma (A_γ) in comparison with the thorium isotope. The C_o values of half of the samples were close to the worldwide value. These samples specified by high count rate are commonly used in construction materials for building in the Sulaymaniyah Governorate, Dwelling (Alsaedi, et al., 2013).

Most dwelling areas of the northern region of Iraq (Kurdistan region) are bounded by mountains. Usually, high gamma (radon) dose emanates from the houses floor of the mentioned area. Therefore, the building should contract with raw materials containing a low radon/gamma dose. Fortunately, the lowest value of H_{ann} was clarified in Table III that encourages the author to suggest that the best locations for establishing the construction material factory including Goptapa, Qaladize, and Darbandikhan areas. An anomaly in C_o value was found in Tanjaro (Sand) sample, this means that the sand sample of this Tanjaro sand screening factory is not appropriate for buildings. Therefore, the author suggests that the authorities should prevent to establish the (sand and pebble) screening factory near a stream draining of a city.

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