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RADON CONCENTRATION AND ITS INDICES IN BULAK (MENCILIS) CAVE

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ABSTRACT

The Bulak cave is in the village of Bulak near Safranbolu city. It's one of the longest caves in Türkiye, which goes 6 kilometers into the inside, but approximately 400 meters are accessible to visitors. You require a professional guide and special equipment to travel further into the cave with lakes, a waterfall, and rivers which are essential for cavers, tourists, and researchers. However, there are possible several health problems for cave visitors. This study mainly aims to measure the radon concentrations with the nuclear track CR-39 detector in the cave environment. Twenty-two detectors were distributed inside the tourist's area, and another six detectors were in the cave's deeper region. The exposure time was one month on 24 September 2020, in addition to two soil samples collected from the cave used for medical purposes. These detectors are collected after 30 days and then etched with a chemical solution. The radon concentrations were calculated, which ranged between 16.437 (Bq/m^3) and 48.652 (Bq/m^3) using an optical microscope from the track density in detectors. The minimum and maximum values of radiation indices AED (mSv/y), LCR (WLM), PAEC (mWL), D_{soft} (nGy/h), D_{lung} (nGy/h) and H_{eff} (nSv/h) are presented, and all results for the radon and radiation indices are within the global limit. Radon concentrations for two soil samples were 26.956 (Bq/m^3) and 59.172 (Bq/m^3), and all the results were within the acceptable limits recommended by ICRP and UNSCEAR. The XRF examination was performed, which indicated the presence of Fe, Cu, Zn, As, and Mn minerals with high concentrations of 101607, 552, 1337, 237, and 1601 ppm, respectively, which are all more than the world permissible limits. The XRD analyses for the soil sample indicated the presence of clay and non-clay minerals such as Feldspar, Quartz, Gypsum, Calcite, Palygorskite, Kaolinite, and Montmorillonite.

1 INTRODUCTION

Radon (Rn-222) and its daughters (Po-218, Po-214, and Bi-214) radiate in the air as alpha particle that contributes more than 50% of public natural exposure [1]. An electrically charged nucleus (Po-218) sticks to atoms of dust or to dust and water droplets in the atmosphere and materials on the surface of the earth. So, the air in which radon is present contains dust particles loaded with the decomposition products of radon with high radioactivity. Radon gas residues stick to all surfaces in nature, including air and soil [2], and there are two ways that radon and its disintegration products can enter the human body, especially the respiratory and digestive systems. The latter does not represent a danger, because the presence of food in the stomach, even if its thickness is within a millimetre, can stop most of the alpha particles resulting from the disintegration of radon and its offspring. In the case of inhalation of radon offspring suspended in the air, if they enter the respiratory system they stick to the wall of the lungs [3]. The radiation may lead to DNA damage directly by displacing electrons from the DNA molecule or by changing the composition of other molecules in the cell. They may interact with the DNA and lead to cell damage or change the growth of the cell and the occurrence of a genetic mutation [3,4]. As stated in the committee's fourth report composed to study the biological effect of ionizing radiation, about 3-14% of cancer injuries result from radon gas [5]. Many studies have classified radon as a human carcinogen, which was done in Europe, North America, and China [6-8]. Caves are natural sites between rocks that extend in such a way as to allow human entry. It is found in most mountainous regions, and it was formed over thousands of years and has a unique landscape. It has great importance as a site for recreational, tourism, and scientific activities, especially in Türkiye [9-15]. The radon concentrations of Mencilis (Bulak) cave have been determined by using passive nuclear track detectors [16]. Radon measurements in houses in the provinces of Bingöl and Muş (Türkiye) and their neighbouring villages have been carried out by means of CR-39 nuclear track detectors [17]. Radon concentration has been examined in surface water samples taken from Van Lake in spring and autumn by using CR-39 solid nuclear track detectors and RadoSYS radon measurement system [18]. The radon concentration levels in the spring and summer seasons in two rooms (the kitchen and the living room) of a building in the city centre of Manisa have been determined by means of CR-39 nuclear track detectors [19]. Radon concentrations can be measured by two methods, the active method (by means of electronic devices) and the passive method (by means of solid-state nuclear track detectors, SSNTDs). In the second method, it is common to use CR-39, known as (Columbia Resin-39), which is a poly allyl diglycol carbonate (PADC). It has a cross-linked lattice structure. It is partially crystalline. Amorphous with ~20% crystallinity (and contains carbon bonds in its monomer, and these bonds can be easily broken as they are relatively weak when exposed to radiation or ionizing particles [20].

In this study, radon concentrations in the air and soil of the Bulak cave were measured with CR-39 nuclear track detectors. At the same time, mineral analysis was performed in the selected soil sample. This work will contribute to the many studies investigating the influence of indoor air quality in caves and other interior environments [21-23]. The paper is organized as follows: Section 1 presents shortly literature and general information about the paper. Section 2 describes the experimental part used in the calculation, and Section 3 presents the study's findings. Finally, concluding remarks are presented in Section 4.

2 EXPERIMENTAL PART

The map in Figure 1 shows the location of Bulak (Mencilis) Cave, which has the coordinates of the entrance to the cave: 41 16′28.60N, 32 37′28.58E and elevation 753.2 m [24]. This cave which forms a large ecosystem is still alive today. The Jurassic Cretaceous period was 65-200 million years ago and occurred in limestone. The entrance level of the Bulak Cave is 291 m above the water source entrance, where the underground stream is exposed. The cave's temperature is 12-14°C, and the humidity is relatively high (60-70%) [25]. The Mencilis Cave in the village of Bulak near Safranbolu is a tourist attraction. It is the fourth-largest cave in Türkiye with a length of 5250 meters [24], and its natural beauty attracts the attention of domestic and foreign tourists. The first 350 m of the cave has been opened to tourism since 2003 [24]. The cave consists of three interconnected floors. It has an underground river at the main entrance on the ground floor, and there is a siphon directly in front of the entrance. The floor above it is 200 m northeast of the main entrance fossil layer. The total length of the fossil floor is 1200 m, and it contains beautiful stalactites, stalagmites, columns, and walls.

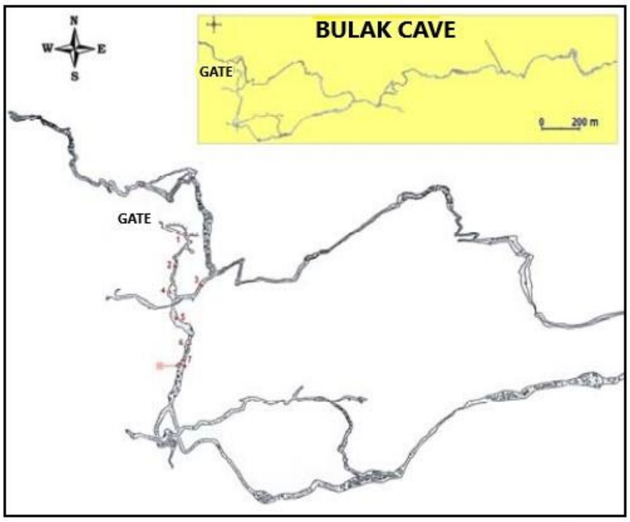

Figure 1. *Location map of Bulak cave. (Taken from ref. [16] with permission of the authors.)*

At the bottom of the cave, a water source contributes to Safranbolu's drinking water. The flow rate of the underground river on the lower floor varies between 0.55 – 2.20 m^3/s depending on the rainy seasons. The underground river has formed lakes of different sizes, reaching 5 meters in depth. An image of the interior view of the cave is given in Figure 2. Considering all the floors of the cave, it forms a large ecosystem with its living and non-living assets [25].

Figure 2. *Interior view of Bulak cave.*

Twenty-two detectors were distributed within the cave at 20 meters intervals from each other in the walking area of the tourists. These detectors were kept in a sponge as shown in Figure 3 and placed in a wide and deep area (group B consisting of 6 detectors) inside the cave as well as in the area under the walking path reserved for tourists. The detectors were collected after a period of one month, and the detectors were transferred after the exposure process to a chemical etching process using a sodium hydroxide solution with a normality of 6.25 at a temperature of 60ºC for 6 hours. The detectors transferred to the process of calculating the tracks using a Nikon type-168 optical microscope.

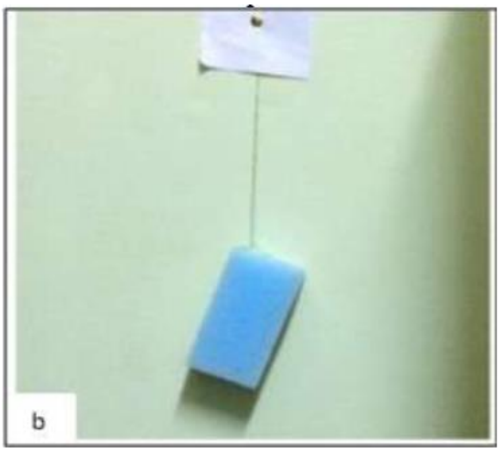

Figure 3. *The nuclear track CR-39 detectors inside the sponge.*

The radon concentration (C_{Rn}) is calculated after comparing it with the standard source in Figure 4 by using the following equations [26].

$$C_{\text{Rn}} = (\rho/t)(E_s/\rho_s) \quad \textbf{(1)}$$

where ρ is the track density given by $\rho = N_{\text{avg}}/A$, where N_{avg} is the number of tracks, A is the area of the view of the microscope, t is the exposure time (days), E_s is the radon exposure of standard source (Bq.day.m^{-3}), and ρ_s is the track density (number of tracks per mm^2) of the standard source. Equation (1) can be rewritten as

$$C_{\text{Rn}} = \rho k/t \quad \textbf{(2)}$$

where k is the calibration factor. It gives the linear relationship between E_s and ρ_s. The value of k is dependent on the radius and the effective height of the measuring can. The radiation indices are calculated, like the annual effective dose (mSv/y) by using the following equation [27]

$$D(\frac{\mathrm{mSv}}{\mathrm{y}}) = C\ E\ h\ t\ f \quad \textbf{(3)}$$

where E is the equilibrium factor (0.4), h is the occupancy factor (0.8), t is the conversion factor for year 8760 (h/y), f is the dose factor of conversion (9×10^{-6} $mSv/Bq.h.m^{-3}$) [26].

For millions of lung cancer patients every year, the lung cancer compact panel (LCCP) value is calculated using the following equation [26]

$$LCCP = D \times 18 \times 10^{-6} \quad \textbf{(4)}$$

Also, the concentration of potential alpha energy (WL) is obtained by equation [24]:

$$PAE(\mathrm{WL}) = E\frac{C_{\mathrm{Rn}}}{3700} \quad \textbf{(5)}$$

The dose rate related to soft tissues of inhaled radon that partly dissolved with a factor of (0.4) is given by the following relationship [29]:

$$\dot{D}_{\text{soft tissues}}(\mathrm{nGyh^{-1}}) = 0.005\chi_{\mathrm{Rn,\ air}}(\mathrm{Bqm^{-3}}) \quad \textbf{(6)}$$

Assuming that the air volume in the lungs is (3.2×10^{-3} m^3) for the reference man, the dose rate for the lungs and the effective equivalent dose rate are defined by [29].

$$\dot{D}_{\mathrm{lung}}(nGyh^{-1}) = 0.004\chi_{\mathrm{Rn,\ air}}(Bqm^{-3}) \quad \textbf{(7)}$$

and

$$\dot{H}_{\mathrm{eff}}(nSvh^{-1}) = 0.18\chi_{\mathrm{Rn,\ air}}(Bqm^{-3}) \quad \textbf{(8)}$$

where the quality factor for alpha-radiation equals 20, and the weighting factor for the lungs and other tissues are 0.12 and 0.88, respectively.

3 RESULTS AND DISCUSSION

The radon concentrations and radiation indices for Bulak cave and soil sample taken from the cave and the mineral concentrations in this soil sample were measured using the nuclear track CR-39 detectors and presented in Tables 1-3.

Table 1. *Radon concentrations and radiation indices for Bulak cave. The global limit values are 100-200 (Bq/m^3) [35], 3-10 (mSv/y) [35], 170-230 [35], 53.33 (mWL) [26], and 1.15 (mSv/yr) [36] for the radon concentration (Bq/m^3), the AED (mSv/y), the LCR (WLM) per 10^6 persons, the PAEC (mWL), and H_{eff} (nSv/h), respectively.*

SN	Track density (ρ) (track/mm^2)	Radon concentration (Bq/m^3)	AED (mSv/y)	LCR (WLM) per 10^6 persons	PAEC (mWL)	D_{soft} (nGy/h)	D_{lung} (nGy/h)	H_{eff} (nSv/h)
1B	996	65.483	1.644	29.592	7.963	0.328	2.619	11.786
2B	952	62.590	1.572	28.296	7.655	0.316	2.518	11.330
3B	950	62.459	1.568	28.224	7.596	0.313	2.498	11.242
4B	735	48.323	1.213	21.834	5.877	0.242	1.933	8.697
5B	886	58.251	1.463	26.334	7.084	0.292	2.330	10.484
6B	760	49.967	1.255	22.590	6.076	0.250	1.998	8.993
1	300	19.724	0.498	7.200	2.399	0.099	0.789	3.550
2	270	17.751	0.448	8.064	2.159	0.089	0.710	3.195
3	260	17.094	0.431	7.758	2.079	0.085	0.684	3.077
4	370	24.326	0.614	11.052	2.959	0.122	0.973	4.379
5	610	40.105	1.012	18.216	4.878	0.201	1.604	7.219
6	630	41.420	1.045	18.810	5.038	0.207	1.657	7.456
7	570	37.475	0.945	17.010	4.558	0.187	1.499	6.746
8	530	34.845	0.879	15.822	4.238	0.174	1.394	6.272
9	640	42.078	1.062	19.116	5.118	0.210	1.683	7.574
10	600	39.448	0.995	17.910	4.798	0.197	1.578	7.101
11	250	16.437	0.415	7.470	1.999	0.082	0.657	2.959
12	580	38.133	0.962	17.316	4.638	0.191	1.525	6.864
13	410	26.956	0.680	12.240	3.278	0.135	1.078	4.852
14	350	23.011	0.581	10.458	2.799	0.115	0.920	4.142
15	370	24.326	0.614	11.052	2.959	0.122	0.973	4.379
16	640	42.078	1.062	19.116	5.118	0.210	1.683	7.574
17	740	48.652	1.227	22.086	5.917	0.243	1.946	8.757
19	400	26.298	0.663	11.934	3.198	0.131	1.052	4.734
20	410	26.956	0.680	12.240	3.278	0.135	1.078	4.852
21	420	27.613	0.697	12.456	0.003358	0.138	1.105	4.970
22	410	26.956	0.680	12.240	0.003278	0.135	1.078	4.852

As can be seen from Table 1, it can be noted that the highest value of radon concentration is in sample-17 (48.652 Bq/m^3) and the lowest value in sample-11 (16.437 Bq/m^3). In group B, the highest value of radon concentration is (65.483 Bq/m^3) in sample-1B and the lowest value is (48.323 Bq/m^3) in sample-4B which represents a region deeper than the first region and through the observation of radon concentrations.

It has been measured that they are different because the cave areas are sometimes open and wide, and others are narrow. This also affects the results of radon concentration in two soil samples. According to the procedure in reference [30], the measured results for the black and red samples are 58.298 Bq/m^3 and 127.972 Bq/m^3, as shown in Table 2. These soil samples contain some minerals that contribute to increasing the concentration of uranium in them. Since they are rich in minerals, they can be used in the treatment of certain skin diseases.

Table 2. *Radon concentrations and radiation indices for soil sample of Bulak cave.*

	Sample		
	Red	Black	Global Limit
Radon concentration (Bq/m^3)	127.972	58.298	(100-200 Bq/m^3) [35]
Radium equivalent content (Bq/kg)	3.361	1.531	(370 Bq/kg) [36]
Area exhalation rate (Bq/m^2.h)	0.160	0.073	(2.5 Bq/m^2.h) [26]
Mass exhalation rate (Bq/kg.h)	0.012	0.012	-

The minimum values of radiation indices AED (mSv/y), LCR (WLM), PAEC (mWL), D_{soft} (nGy/h), D_{lung} (nGy/h) and H_{eff} (nSv/h) are 0.415, 7.470, 1.499, 0.082, 0.657, and 2.959. The maximum values of these radiation indices are 1.227, 22.086, 5.917, 0.243, 1.946, and 8.757. All results for the radon and radiation indices are within the global limit [26, 35-37].

There are approximately 40.000 caves in Türkiye, most of which are protected to preserve geological formations. Only 1% of them are for tourism purposes because they are dangerous to human health due to the quality of the air and the concentration of radon in it. There are limited studies on the nature of the atmosphere inside the caves in Türkiye [15-24]. The mineral concentrations obtained from the XRF examination of the soil samples are given in Table 3. We find that all concentrations of minerals are higher than the acceptable limit given by some science institutions.

***Table 3**. Mineral concentrations in a soil sample of Bulak cave.*

Minerals	Sample		
	Red	Black (ITU) [31]	Permissible Limits (ppm) [32-34]
Fe	132400	8800	3800
Cu	550	489	30
Zn	1393	0.044	50
As	260	-	10
Mn	1905	-	300
Pb	-	-	10
Cr	-	2	70
Co	-	-	8
Cd	-	-	0.06
Ni	-	-	40
Hg	-	-	-

The relationship between E_s and ρ_s is shown in Figure 4. The value of *k* is equal to 0.169 Track. m^3/Bq.Day.mm^2.

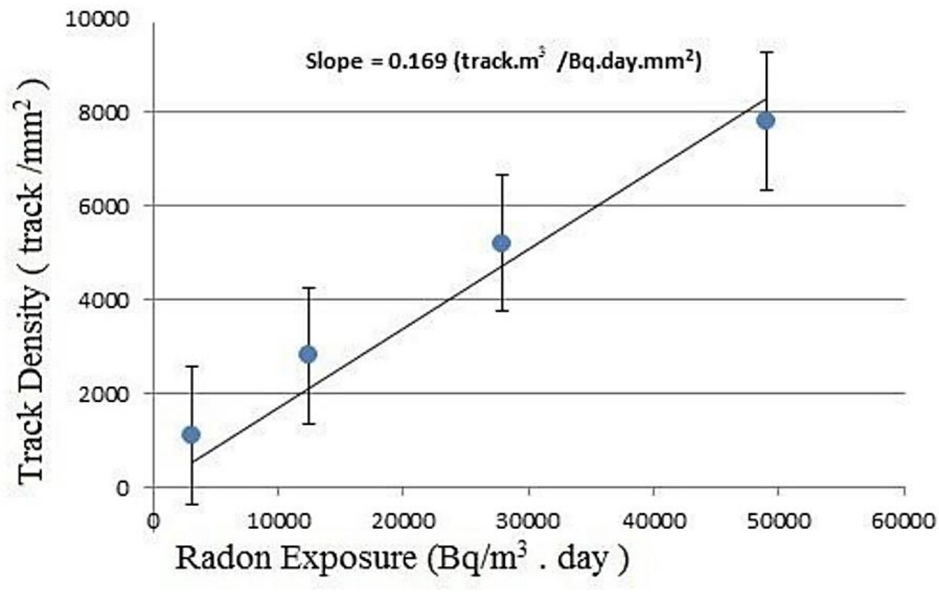

***Figure 4**. The linear relationship between E_s and ρ_s.*

The clay and the non-clay minerals such as Kaolinite, Feldspar, Montmorillonite, Calcite, Palygorskite, Gypsum, and Quartz in the soil sample (the red one) detected with the XRD analyses are presented in Appendix 1 (a-g).

4 CONCLUSIONS

There are different types of soil due to the narrow and wide areas inside the cave, so we conclude that the variation of radon concentrations according to the different regions is normal. The reason for all this is probably the geological composition of the rocks of this cave. The XRF and XRD analyses show that the soil samples taken from the cave are rich with minerals. The mineral concentrations are above acceptable limits set by some scientific institutions. Radon concentrations and radiation indices obtained in this study are within the global limit by UNSCEAR (2000), ICRP (2009), ICRP (2010), and OECD (1979). To reduce radon concentration and improve air quality by reducing carbon dioxide, we recommend at least the construction of ventilation corridors in the area reserved for tourism.

This study will be useful for the next measurements of the concentration of radon in the cave. It is important to make regular radon measurements at least every five years to reduce the risk of exposure to radon gas and the possibility of cancer from this gas. Most of the studies around the world agree that radon risk increases by ~10% with the increase in radon concentration of 100 Bq/m^3 [3]. The present work is a support to the study conducted by [38] on the quality of air and carbon dioxide concentrations inside the cave.

Conflict of interest

There is no conflict of interest among the authors.

Authors contributions

Necla ÇAKMAK and Ulvi KANBUR participated in design and coordination of the study; helped to draft the manuscript; performed the statistical analysis. Khalid Hadi Mahdi AAL-SHABEEB and Ahmet Mustafa ERER participated in the experimental studies. All authors read and approved the final manuscript.

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Research and Publication Ethics

The study is complied with research and publication ethics.

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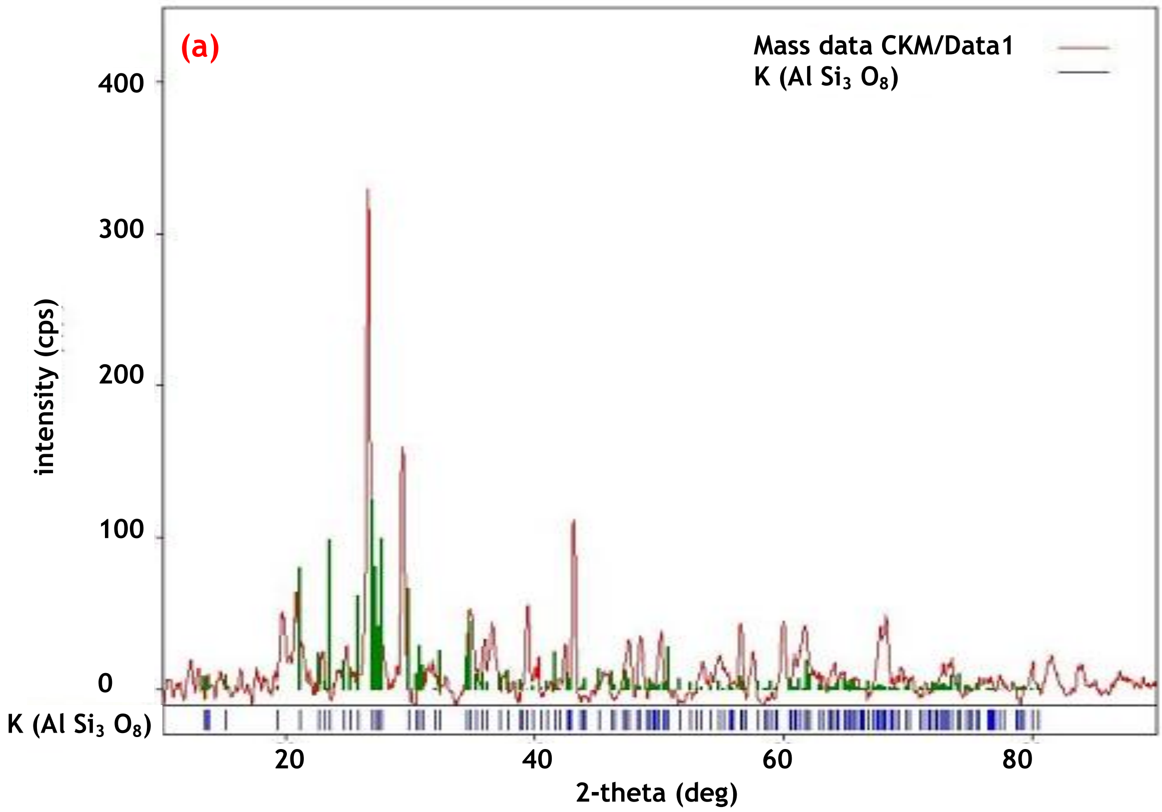
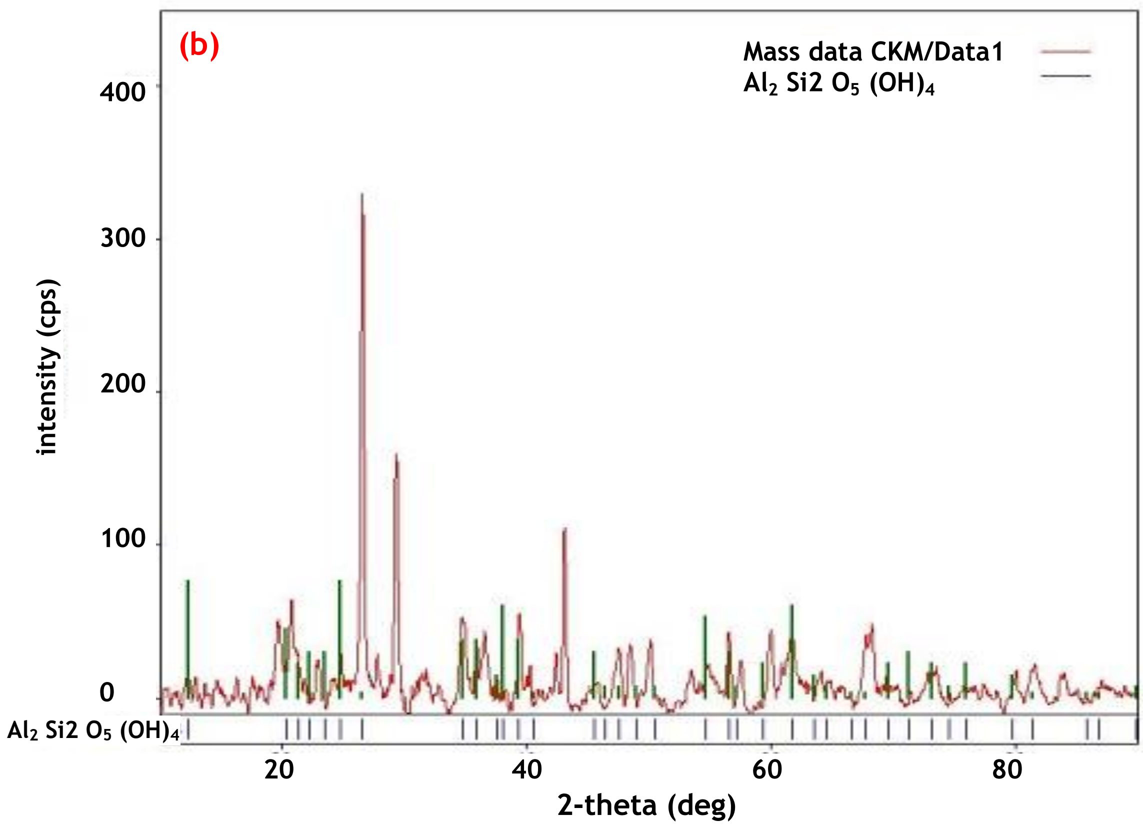
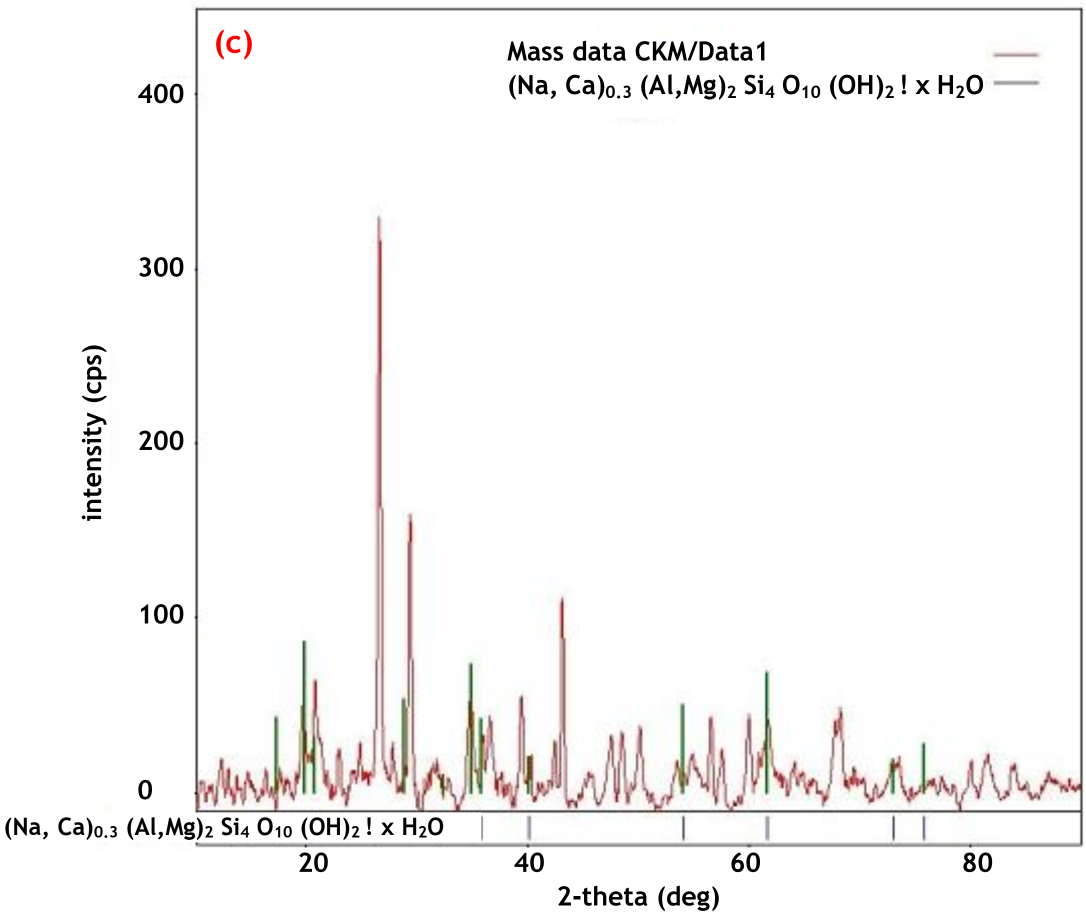
APPENDIX

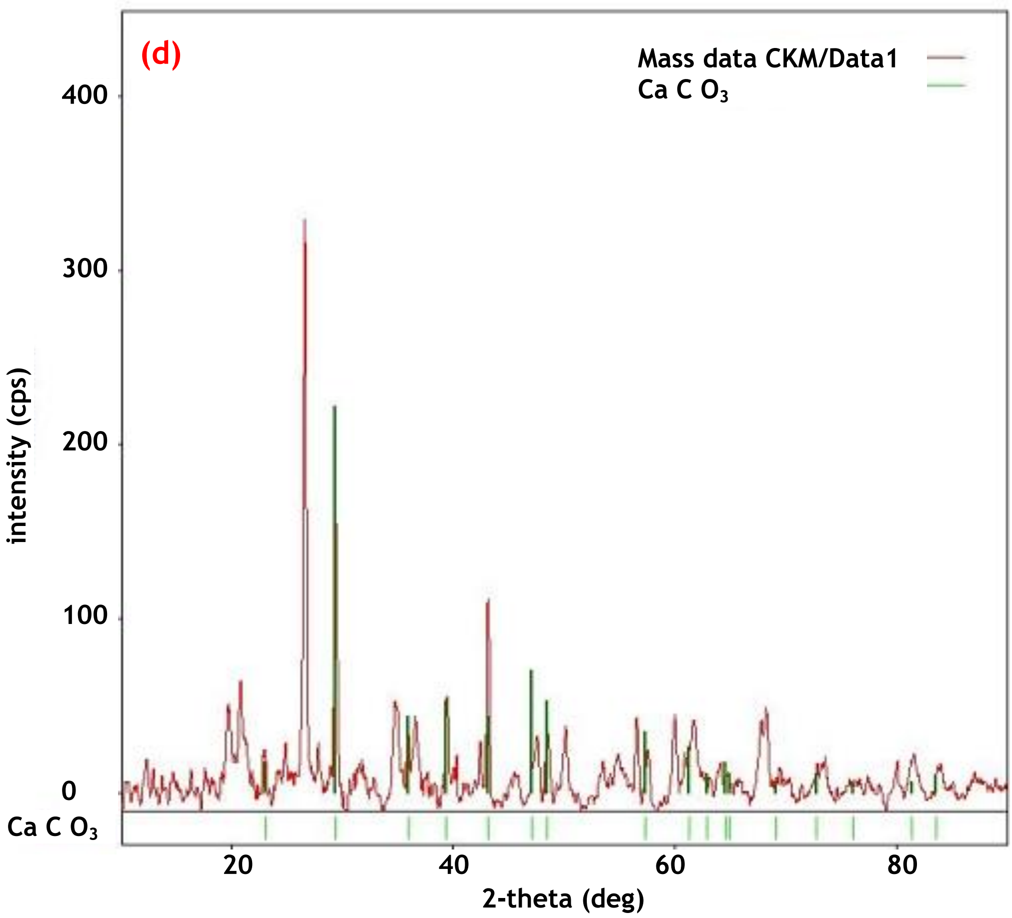


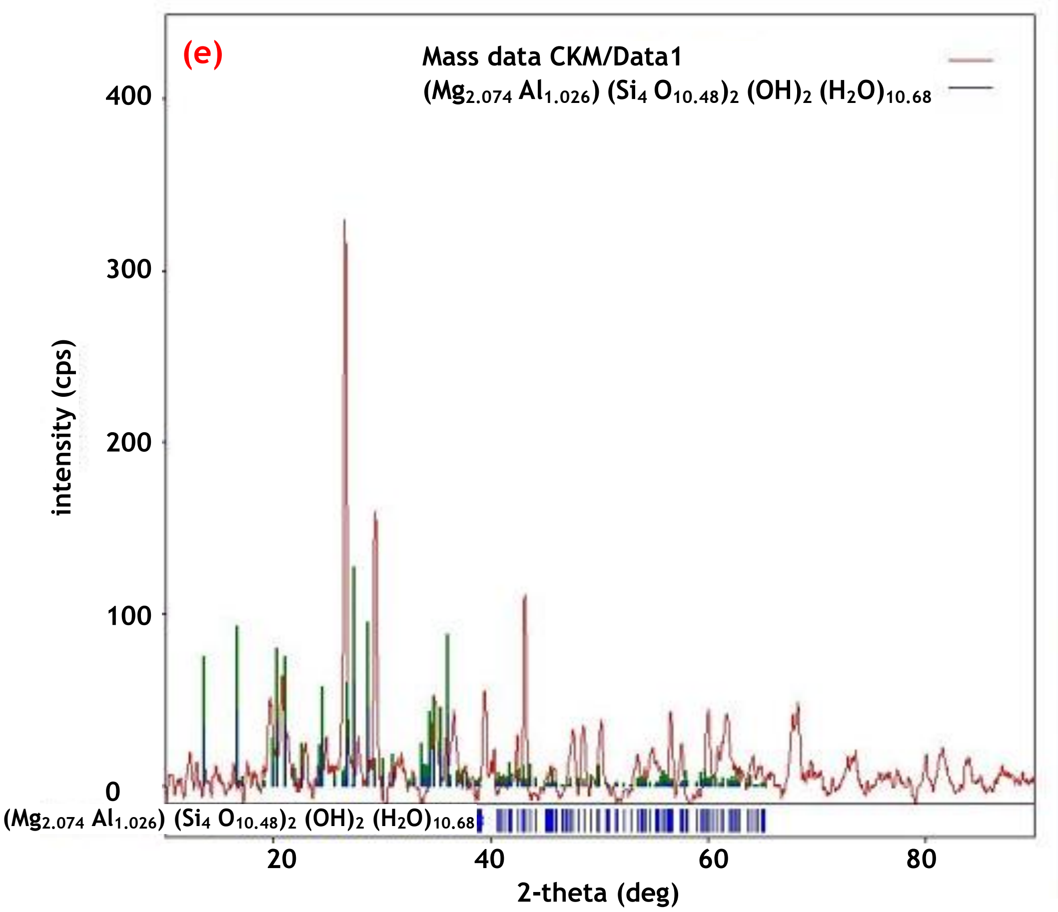
(a)
Mass data CKM/Data1
K (Al Si3 O8)
intensity (cps)
400
300
200
100
0
K (Al Si3 O8)
20
40
60
80
2-theta (deg)

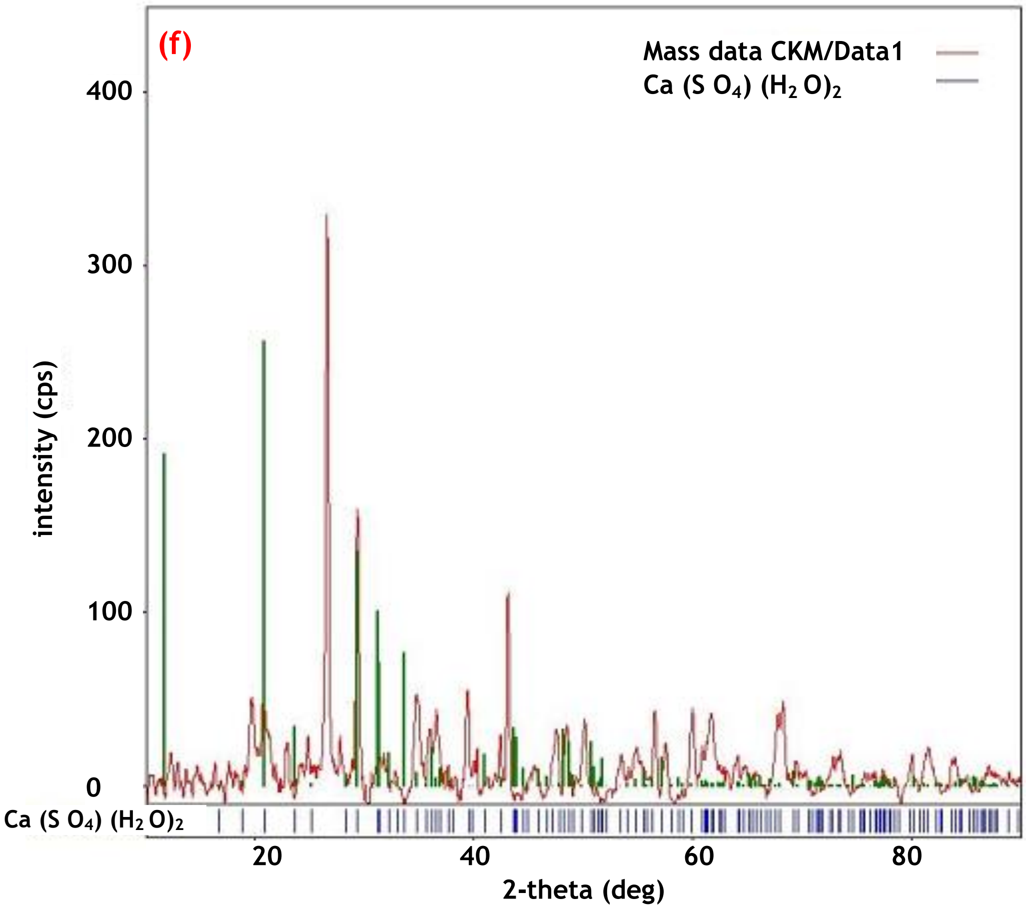


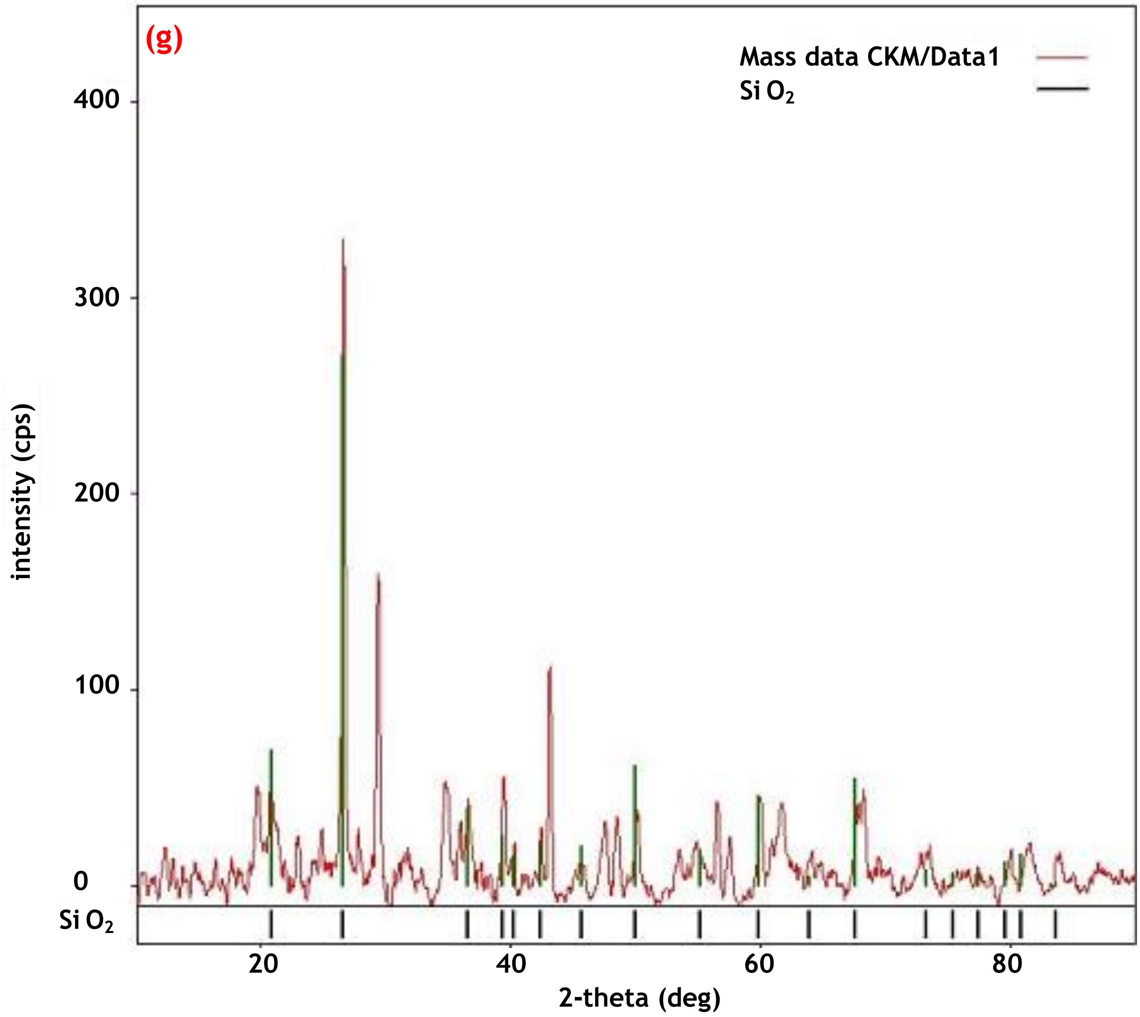
(b)
Mass data CKM/Data1
Al2 Si2 O5 (OH)4
intensity (cps)
400
300
200
100
0
Al2 Si2 O5 (OH)4
20
40
60
80
2-theta (deg)


(c)
Mass data CKM/Data1
(Na, Ca)0.3 (Al,Mg)2 Si4 O10 (OH)2 ! x H2O
400
300
200
100
0
intensity (cps)
(Na, Ca)0.3 (Al,Mg)2 Si4 O10 (OH)2 ! x H2O
20
40
60
80
2-theta (deg)


(d)
Mass data CKM/Data1
Ca C O3
400
300
200
100
0
intensity (cps)
Ca C O3
20
40
60
80
2-theta (deg)


(e)
Mass data CKM/Data1
(Mg2.074 Al1.026) (Si4 O10.48)2 (OH)2 (H2O)10.68
intensity (cps)
400
300
200
100
0
(Mg2.074 Al1.026) (Si4 O10.48)2 (OH)2 (H2O)10.68
20
40
60
80
2-theta (deg)


(f)
Mass data CKM/Data1
Ca (S O4) (H2 O)2
intensity (cps)
400
300
200
100
0
Ca (S O4) (H2 O)2
20
40
60
80
2-theta (deg)



Appendix 1. *The XRD analyses for the soil sample (the red one), and represent* ***(a)*** *Kaolinite,* ***(b)*** *Feldspar,* ***(c)*** *Montmorillonite,* ***(d)*** *Calcite,* ***(e)*** *Palygorskite,* ***(f)*** *Gypsum, and* ***(g)*** *Quartz minerals.*