Measurement and Spatial Analysis of Uranium-238 and Radon-222 of Soil in Seoul

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ABSTRACT

Identification of radon in soil provides information on the areas at risk for high radon exposure. In this study, we measured uranium-238 and radon-222 concentrations in soil to assess their approximate levels in Seoul. A total of 246 soil samples were taken to analyze uranium with ICP-MS, and 120 measurements of radon in soil were conducted with an in-situ radon detector, Rad7 at a depth of 1-1.5 m. The data were statistically analyzed and mapped, layered with geological classification. The range of uranium in soil was from 0.0 to 8.5 mg/kg with a mean value of 2.2 mg/kg, and the range of radon in soil was from 1,887 to 87,320 Bq/m³ with a mean value of 18,271 Bq/m³. The geology had a distinctive relationship to the uranium and radon levels in soil, with the uranium and radon concentrations in soils overlying granite more than double those of soils overlying metamorphic rocks.

Key words: Radon in soil, Uranium in soil, Radon prone area, Radon in Seoul

1. Introduction

Radon is a radioactive, inert gas that is ubiquitously present in nature, and it exists mostly as 222Rn, which is the 6th progeny in the uranium-238 decay series. In daily life, radon is known to contribute to 42% of the total source of radiation (World Nuclear Association, 2016), and radon and its progenies are considered to be the second leading cause of lung cancer when delivered to lung tissues (WHO, 2009).

Soil and bedrock have been recognized as the main sources of radon in nature (Nazaroff and Nero, 1988). The concentration of radon in soil is thousands of times more than that in the atmosphere (Wilkening, 1990). It moves up to the top of the ground through pores of soil or fractures of rock and seeps into cracks of buildings through diffusion and pressure gradient (Tanner, 1980). Therefore, the radon in soil has been measured in numerous studies in order to estimate the radon potential or to identify the radon risk area (Kemski et al., 2001; Lara et al., 2011; Barnet and Pacheroa, 2011, 2013; Demoury et al., 2013; Duggal et al., 2014; Szabo et al., 2014). In Europe and in the USA, this kind of research has been carried out for decades, and the majority of European countries have produced radon risk maps by measuring radon in soil at different periods and scopes (Dubois, 2005). As for some guidelines, Åkerblom (1999) suggested the following classification for radon in soil: low risk (<10,000 Bq/m³), normal risk (10,000-50,000 Bq/m³) and high-risk (>50,000 Bq/m³), and Kemski (2001) in Germany made four categories for geogenic radon distribution: low (<10,000 Bq/m³), medium (10,000-100,000 Bq/m³), increased (100,000-500,000 Bq/m³) and high (>500,000 Bq/m³).

Radon in soil depends on many variables, including bed rock type, fragmentation of bed rock, soil characteristics and permeability, radium content, moisture content, pressure differential, climate, etc. (Tanner, 1980; Nazaroff et al., 1985; Stranden et al., 1984; Cohen and Gromicko, 1988; Miles, 1998; Kojima and Nagano, 1999). Therefore, recent studies have focused on GRP (geogenic radon potential) mapping by statistical methods using certain parameters, including geology, topography, climate, permeability, radium or uranium concentration, indoor radon concentration, radon concentration in soil, etc. (Demoury et al., 2013;
Szabo et al., 2014; Drolet and Martel, 2016; Pásztor et al., 2016). Of those parameters, geology and uranium concentrations have been reported to have a strong correlation with radon in soil in many studies (Gundersen et al., 1992; Ball and Miles, 1993; Clavensjo et al., 1999). Ielsch et al. (2010) produced a radon potential map from uranium concentrations; Kakati et al. (2013) indicated that the correlation coefficient between uranium concentration and radon exhalation rate was 0.98; and Demoury et al. (2013) made a GRP map for which the uranium concentration map was used as the first source map.

In South Korea, the guideline for radon is established only for indoor radon. Hence, almost all radon research in Seoul has focused on atmospheric radon. Most of all, radon measured in air in underground stations has been used in some studies to analyze the prevalence of radon in Seoul (Jeon, 2007; Baek, 2007). In contrast, radon in soil has been investigated in a few studies that showed that the concentration of radon in some locations with granite was quite high (Je, 2003; Kim et al., 2012). Therefore general radon concentration measurement in soil in this study is the first case in Seoul and it can make a contribution to a future plan for soil radon policy.

In this study, uranium concentrations were pre-measured in order to predict the areas with a high risk for radon, and then radon concentrations in soil were spatially measured and analyzed. The uranium map and the radon map with geological classification were made using Arcview 3.3, and then we verified their correspondence with geology.

2. Material and Methods

2.1. Geology of Seoul

Seoul is the study area, and it is located in the mid-west region of the Korean peninsula in East Asia. The city covers about 605 km², and its population is of about 10.3 million people (Seoul Census, 2016). Topographically, Seoul has some high mountains in the north, and the Han River divides the city into north and the south, flowing westward.

The bedrock of Seoul consists of granite and metamorphic rocks, including gneiss and some schist. The pre-Cambrian banded gneiss (Pcbngn) and pre-Cambrian granite gneiss (Peggn) with an age of about 2,500-1,800 million years lie under most hills and plains, and some of them are intercalated with pre-Cambrian mica schist (Pcms). Granite exists as Jurassic granite (Jgr) or Porphyritic granite (Pgr), with an age of about 160 million years in and around northern mountains as a type of domes. Along the Han River and its 8 sub-flows, Quaternary alluvium (Qa) covers the granite and gneiss bedrocks with an age of about 1.8 million years, and it is composed of gravel, sand, silt and mud with a depth of 15 m at most (Korea Institute of Geoscience and Mineral Resources, 1999; Grapes and Jeong, 2008). The geological classification is shown in Fig. 3 and 4.

2.2. Measurement of Uranium-238 in soil

246 soil samples were randomly collected using Geoprobe, a type of boring equipment, at a depth of 1.5 m from June to October in 2014. An EPA method 200.8 was referred to analyze the uranium concentrations using acid digestion for pretreatment. The soils were dried and crushed, and then screened with a 100-mesh sieve in the laboratory and acid digestion using aqua regia (hydrochloric acid : nitric acid = 3 : 1) was conducted by official protocol on soil pollution. The samples were diluted to measure uranium-238 using ICP-MS (Spectro MS, Germany) in the department of drinking-water analysis in Seoul Metropolitan Government Institute of Public Health and Environment. For quality control, uranium 0.5 ppb was measured 5 times with the result of that the accuracy was 97.93% and the precision was 0.12%, furthermore, IDL (Instrument Detection Limit) was 0.02 ppb and LOQ (Limit of Quantification) was 0.08 ppb, respectively.

2.3. Measurement of Radon in soil

Radon-222 was measured in-situ for 120 sites from July to mid-September in 2015, which is that measuring Radon In-Situ is quite complex and time-consuming and limitation of time and instrument made us hard to conduct measuring radon and uranium in the same year. Although radon in soil as a representative value has been generally measured using grid methods through which the area is divided at some intervals as 1 sample per 100 x 100 meter squares or 1 x 1 kilometer squares in European radon soil maps (Dubois, 2005), boring in privately-owned lands requires adequate administrative procedures to gain permission in advance.
Therefore, the sampling sites of this study were selected from about 350 Annual Soil Contamination Monitoring sites that were previously legally appointed. Instead, the sites were randomly selected, and these also covered the entire area of study at some intervals. After removing the cement or concrete top coverings, Rad7 (Durridge Company, USA) was used to measure radon. Rad7 is an alpha-ray detector with a 0.7 litter hemispheric electrical conductor where Polonium-218 decayed from Radon-222 is attached to, and Rad7 uses its own air pump to draw out air from soil. A stainless steel probe with holes at the end of it, supplied by Durridge Company, was inserted into the soil at a depth of 1-1.5 m, and the hole on top was sealed well enough not to disturb the radon gas in the soil. The instrument was purged for 5 minutes, and a Sniff Protocol recommended by the company for the measurement in soil was carried out. The protocol is set to inhale air from soil 5 minutes at a flow of 1 L/min, and then measure the radon concentration four times.

3. Results and Discussion

3.1. Uranium-238 in soil

Uranium-238, commonly found in nature, produces radon-222 in its course of decay. Regarding the relation between uranium and radon, the uranium-238 in soil was investigated prior to measuring radon-222. General statistics for uranium-238 are shown in Table 1 and Fig. 1. For 246 measurements, the concentrations are from 0.0 to 8.5 mg/kg with a mean value of 2.2 mg/kg and a median value of 2.3 mg/kg. The data is skewed-right, and the majority of the data points are at around 2.3 mg/kg, which is a little higher than the 1.8 mg/kg as the normal concentration of uranium-238 in soil produced by Wilkening (1990).

The spatial distribution of uranium-238 in soil layered with bedrocks is displayed in Fig. 2 using a format corresponding to a map. The map was made using Arcview3.3, and the geological base layer files (shp, dbf, shx, etc.) were provided by the Seoul Development Institute and KIGAM (Korea Institute of Geoscience and Mineral Resources). And basement rock in each sampling spot was double-checked on the 1:250,000 geographical map provided on-line by KIGAM (https://mgeo.kigam.re.kr/map/map.jsp?mode=geology_250k).

Most of the high concentrations (over 4 mg/kg) appear in the granite bedrock area on the north of Seoul. Average value of the uranium-238 concentrations in soil in granite area is 4.0 mg/kg while that of in non-granite area is 1.5 mg/kg.

Table 1. General statistics of uranium-238 concentration in soil (mg/kg)

| Measurements       | Mean | Median | SD  | Min. | Max. |
|--------------------|------|--------|-----|------|------|
| Total              | 246  | 2.2    | 2.3 | 1.7  | 8.5  |
| granite area       | 72   | 4.0    | 3.9 | 1.3  | 8.5  |
| non-granite area   | 174  | 1.5    | 0.8 | 1.2  | 6.5  |

Fig. 1. Box-plots for uranium-238 in the overall area and the granite area.
mg/kg and Fig. 1. shows that data distribution in granite area is more Gaussian than what was previously observed for the overall data. Although the soils in the sampling spots were mostly covering soils that seem to have been moved from another place before construction, it can be assumed that they were supplied from the vicinity since uranium concentrations layering on the geological map display a distinct difference in between granite and non-granite area.

3.2. Radon in soil

Radon in soil is influenced by many factors, including uranium and radium content, permeability, moisture of soil or rock, meteorological differences like temperature and air pressure (UNSCEAR, 1993), and lithological features such as faults and joints (Choubey et al., 1997). In this study, we tried to minimize the assumable influences those kinds of factors;

i) Permeability

Most of soils examined were backfilled soils, whose depths are from 0.4 to 12.7 m with an average of 2.8 m and standard deviation of 2.2 m. The data were achieved from the Borehole Status in Seoul in Geotechnical Information Database System (http://survey.cp.seoul.go.kr) and the distance between a borehole and sampling spot is within 50 m. From the acquired data, the compositions of the soils are shown in Table 2.

Table 2. Physical compositions of the soils in sampling areas

| Composition                          | Counts | Percentage (%) |
|-------------------------------------|--------|----------------|
| Sand and Gravel                     | 14     | 11.7           |
| Silty Sand with Some Sand Gravel     | 55     | 45.8           |
| Waste, Sand and Gravel              | 5      | 4.2            |
| Sand                                | 6      | 5.0            |
| Sand and Silt                       | 35     | 29.2           |
| Silt                                | 1      | 0.8            |
| Missing                             | 4      | 3.3            |
| **Total**                           | **120**| **100.0**      |

Table 2 and almost of them consist of gravel, sand, or sandy silts, therefore the permeability of each soil is expected to be quite high with no such a meaningful difference.

ii) Moisture

In the literature, the dryness of the soil resulted in a decrease in radon transport (Klausman and Jaacks 1987), and the soil moisture content up to some extent (about 15%) increased the radon concentration in soil (Kemski et al. 1996). In this study, although we could not set the moisture content of the samples identically because measuring was done in situ, the moisture content of soil was 3-25%, with an average of 13% and standard deviation of 5%, which is no drastic difference between them.
iii) Depth of soil
Radon in soil is very changeable according to soil depth, but this is known to be solvable by sampling deeper than 70 cm (Talbot et al. 1998). Therefore, the radon was measured at a depth of 1-1.5 m.

iv) Meteorological factors
Diurnal and seasonal differences were not expected to be substantial as radon was measured in summer daytime with the air temperature of 30-35°C. Generally we have lots of rain in summer but it scarcely rained in 2015.

General statistics for the radon concentrations in soils are presented in Table 3 and Fig. 3. The radon concentrations are 1,887-87,320 Bq/m$^3$ with a mean value of 18,271 Bq/m$^3$ and a median value of 13,727 Bq/m$^3$, and their distribution is biased to lower concentrations. Similar to the case of uranium-238, the radon concentration in a granite area also follows a more normal distribution. In granite area (including alluvium area overlying granite), the average radon concentration in soil is 27,415 Bq/m$^3$ while it is 11,512 Bq/m$^3$ in non-granite area (gneiss, schist, and alluvium area overlying them), which the former is more than two times than the latter.

The spatial distribution of the radon concentration in soil layered with their bedrocks is mapped in Fig. 4 using Arcview3.3. Each class was determined by referring to the Swedish Criteria on soil radon, which originally classifies the concentration as ‘low’ at under 10,000 Bq/m$^3$, ‘moderate’ from 10,000 to 50,000 Bq/m$^3$, and ‘high’ for over 50,000 Bq/m$^3$ (Åkerblom, 1987). However, we added the ‘medium high’ level from 25,000 to 50,000 Bq/m$^3$ to provide a more specific description. 68% of the total measurements belonged to the ‘low’ or ‘moderate’ level, 20% to the ‘medium high’ level, and 3% to the ‘high’ level (Table 4). The radon concentrations are low in the metamorphic rock area on the south of the Han River, which cuts across the middle of the city from east to west. On the other hand, they are quite high in the granite rock area around Mt. Bukhan and Mt. Dobong in the north of the city, and the top 3 of the highest, 87,320, 70,855, 70,300 Bq/m$^3$ were observed in that area.

As for the alluvium area, the average concentration is 17,704 Bq/m$^3$. However, in terms of the bedrock, it can be also divided into granite and metamorphic rock areas, and the results show that the average concentration for the alluvium area overlying the granite bedrock is 28,859 Bq/m$^3$ while that of the overlying metamorphic bedrock is 12,391 Bq/m$^3$. Therefore it can be expected that the actual

| Table 3. General Statistics of Radon222 Concentration in Soil (Bq/m$^3$) |
|-----------------|------|------|------|------|------|
| Measurements   | Mean | Median | SD.  | Min. | Max. |
| Total          | 120  | 18,271| 13,727| 15,251| 1,887| 87,320|
| granite area   | 51   | 27,415| 23,569| 17,044| 4,903| 87,320|
| non-granite area| 69   | 11,512| 7,955 | 9,185 | 1,887| 46,800|

Fig. 3. Box-plots for radon-222 in the overall area and the granite area.
4. Conclusions

In this study, we measured uranium-238 and radon-222 concentrations in soil to determine the radon prone area in Seoul and therefore to identify the current risk of a high concentration of radon to take this into account for building construction in the future.

4.1. The uranium-238 concentrations in soil were from 0.0 to 8.5 mg/kg with a mean value of 2.2 mg/kg and a median value of 2.3 mg/kg which follows the generally known average figure in soil. But we identified that the concentrations were much higher in the granite bedrock areas by geological distribution. Unfortunately, the spots for uranium and radon are not identical so that the correlative analysis could not be done.

4.2. The radon concentrations in soil were from 1,887 to 87,320 Bq/m$^3$ with a mean of 18,271 and a median value of 13,727 Bq/m$^3$, and they were biased to a lower concentration. The classifications for the radon map were set to low (<10,000 Bq/m$^3$), moderate (10,000-25,000 Bq/m$^3$), medium-high (25,000-50,000 Bq/m$^3$), and high (>50,000 Bq/m$^3$), referring to the Åkerblom Criteria, which sets 50,000 Bq/m$^3$ as the safety level in soil. Although the risk of soil radon in Seoul was mostly low or moderate, radon concentrations in some spots in the north of Seoul were over 50,000 Bq/m$^3$.

Table 4. Distribution of radon prone area in Seoul

| Risk | Low | Moderate | Medium high | High |
|------|-----|----------|-------------|------|
| Conc. (Bq/m$^3$) | <10,000 | 10,000-25,000 | 25,000-50,000 | >50,000 |
| Counts | 44 (37%) | 47 (39%) | 24 (20%) | 4 (3%) |

Table 5. Geological distribution of soil radon concentration

| Basement rocks | Granite | Metamorphic rocks | Overlying granite | Overlying Metamorphic rocks |
|----------------|---------|-------------------|------------------|--------------------------|
| Counts | 31 | 27 | 20 | 42 |
| Mean | 26,483 | 10,144 | 28,859 | 12,391 |
| Std.Dev | 14,456 | 9,452 | 20,758 | 9,013 |

Fig. 4. Spatial distribution of soil radon by geology.

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4.3. The radon distribution showed a similar tendency to the geology like the uranium did, which is that a high concentration of radon generally occurred in soil overlying granite bedrock. The average radon concentration in soil for which the bedrock consists of granite was of 27,415 Bq/m$^3$, and it is over twice as high as that of metamorphic bedrock at 11,512 Bq/m$^3$, so a more cautious investigation over the granite area is required in the future. In addition, for the alluvium area, the underlying bedrock was thought to dominate the soil radon concentration because the average radon concentration in those areas with granite bedrock is also over twice as bigger than the one with non-granite bedrock.

4.4. The in-situ measurement of radon in soil is relatively complicated, and its representativeness could be questioned due to its changeability according to various variables. Therefore, estimating the radon potential according to a model using parameters influencing the radon concentration such as uranium content, geology, temperature, etc., which is relatively easy to measure can be a better alternative. Therefore the radon potential model using specific statistical techniques as a form of principal component analysis and GIS technique is planned to be developed, and the measured concentrations in this study can be used to verify the model for further study.

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