Distribution of Arsenic in Fresh and Weathered Rocks in Sri Lanka

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Research Article

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Abstract

This study was carried out to determine the distribution of arsenic, which may slowly harm human health, in the weathered rocks of different parent rocks in the country. 293 samples were collected from different crystalline rocks and in-situ weathered formations above the particular parent rock in 50 localities. Selected minor elements (including arsenic) were analysed by X-ray fluorescence spectrometry on RIGAKU KG-X system (Japan). Results indicated that the maximum arsenic amount in the fresh rock was 12 and 48 ppm in completely weathered rocks. About 86.9% of fresh rocks showed less than 5 ppm of arsenic, while 89.8% of their weathered grades showed the arsenic concentration to be less than 10 ppm. Average arsenic in all fresh rock samples was 3.5ppm (lowest); it was 7.6 ppm (highest) in residual soils. This is the normal condition of arsenic distribution worldwide. Under this condition, the arsenic concentration in natural groundwater in the residual soil areas should be below the acceptable limit. Therefore, the amount of arsenic released from parent rocks and their weathered products due to natural geological processes is very low in Sri Lanka.

Introduction

Many patients with chronic kidney disease of unknown etiology (CKDu) were reported from rural Sri Lanka, especially from the North Central Province (NCP) after 1993. CKDu has gradually emerged as a major health problem in the Dry Zone of Sri Lanka for the last two decades (Fig.1). Arsenic (in drinking groundwater) has already been identified as one of the major etiological factors for the disease (Jayasumana et al. 2013). However, Jayasumana et al. (2013) also mentioned that they arrived at this conclusion despite a lack of scientific evidence on the occurrence of arsenic in bedrock and other natural environments in Sri Lanka. Therefore, a study on the distribution of arsenic in the parent rocks and their weathered materials are very important.

The author carried out a geochemical study (via XRF) to find out the variation of major and minor elements for fresh rocks and differently graded weathered materials of major parent rocks of Sri Lanka (Jayawardena and Izawa, 1993 and Jayawardena, 2000). Concentrations of major elements were used in his studies but minor element concentrations were noted as for further scientific studies. After the recognition of the CKDu problem in rural areas of Sri Lanka, the author decided to analyze the occurrence and distribution of arsenic (one of the minor elements detected) in fresh and weathered materials to describe its natural concentration in the country. The objective of this paper is to provide scientific evidence on the distribution of arsenic concentration in different parent rocks and their weathered forms in Sri Lanka. To identify the effect of external sources on groundwater quality, some knowledge and understanding of the concentration of elements in parent and weathered rocks are necessary. These findings may be a source for scientists who research arsenic in groundwater in Sri Lankan soil and create solutions to reduce the spread of CKDu.

Literature Review
General geography and geology of Sri Lanka

Sri Lanka is an island in the Indian Ocean. The total land area measures 65,610 square kilometres. Physiographically, Sri Lanka consists of a central mountainous mass or central highland surrounded by a low, flat plain on all sides that extend to the sea. The Island can be divided into three main morphological regions as coastal lowlands (less than 305 m MSL), uplands (305–915m MSL), and highlands (915–2420m MSL) (Vitanage, 1970). Most of the rivers start from this central highland and flow in a radial pattern towards the sea. Sri Lanka is considered to have a humid tropical climate.

Geologically, 90% of Sri Lanka is made up of high-grade metamorphic rocks from the Precambrian age. These rocks have been formed under the granulite and amphibolite facies of regional metamorphism and re-metamorphism. Also, it is believed that the major types of metamorphosed sedimentary rocks from the Precambrian age consist of Quartzite, gneisses (different mineral combinations), migmatite, marble, dolomite, amphibolite, and charnockite/charnockitic gneiss. However, the original nature before the metamorphism of charnockite/charnockitic gneiss is uncertain (Cooray, 1994). The remaining rocks are sedimentary rocks of predominantly Miocene age (limestone) in the northwest (and sandstone in very few places in the southeast) with some Jurassic sediments (shale and sandstone) preserved in small faulted basins (Fig. 2). There are recent sedimentary formations, which have been identified as Pleistocene and iron oxide deposits in a few locations. There are plutonic-type igneous formations (about 1% of the country) but no evidence for volcanic igneous formations such as solidified lava flows and volcanic dust deposits has been found (Cooray, 1994). Pegmatite, granites, apatite-rich carbonatite, vein quartz, dolerite, copper-magnetite body, and serpentinite bodies are the plutonic igneous intrusions. Also, there are a few small magnetite iron ore deposits in several places within the crystalline complex.

The land surface of Sri Lanka has been subjected to a prolonged period of weathering and erosion under different climatic conditions. The secondary formations arising from weathering such as clay minerals (mainly kaolinite and halloysite) and lateritic soil are found throughout the Island; laterite (goethite, gibbsite, boehmite, dias pore) is found mainly in the southwestern part of the country. In a few locations, some iron ore (hematite, limonite, and goethite) minerals occur as surface deposits. Recent deposits include both residual and alluvial formations. Residual deposits include deep weathered zones of soils that can be found in the central hill country and the intermediate slopes. These deposits are not uniform in character and contain fragments of un-decomposed rocks (Herath 1963 &1963a). The weathering is not uniform in any place in the country and the thickness changes drastically from place to place. There are thick alluvial deposits, flood plain deposits, and small deltas in the coastal lowland areas near to the sea and wind-blown dune sand and silt deposits along the coastal zones.

General distribution of arsenic in parent rocks, weathered rocks, and soils

Arsenic in rocks

The concentration of arsenic in the rocks of the lithosphere varies with the abundance of arsenic-carrying minerals. These minerals may contain arsenic at concentrations reaching 6000 mg/kg. Smedley and
Kinniburgh, (2001) and Takeshi (1998) described the concentration of arsenic in different parent rocks and minerals in the earth's crust (Table 1).

*Arsenic in natural soils*

The rocks are gradually transformed into loose or dense soils during the process of weathering. It passes various stages before it ultimately is reduced to products of residual soils. The rocks in this weathering stage can be grouped on the degree of weathering either chemically, physically, or with any other explanation (Fookes and Horswill, 1970). These processes of chemical weathering alter the parent minerals and create secondary minerals, iron oxides and hydroxides, and some other formations in different chemical compositions that keep some ions within the new products and release others to either surface or groundwater.

Soils near arseniferous deposits worldwide may range from 20 to 2400 ppm. In general, heavy metals in ore deposits are dispersed in soils or weathered zones of rocks mainly by the action of water. The arsenic concentration tends to increase during the weathering process with the lowest concentration in the unweathered rock and the highest in soils. The arsenic concentration range in normal soils varies from 0.1 to 55 ppm with an average of 7.2 ppm. It may vary in different soil horizons such as A, B, and C (Takeshi, 1988). The soils formed from different rock types in Japan are given in Table 2. It shows that the soils formed from sedimentary rocks have higher arsenic concentrations than igneous and metamorphic rocks. Arsenic concentrations in soils are not correlated with soil character or clay content and are not identified as the chemical forms of arsenic in soils. However, arsenic is generally enriched in the B horizon of most normal soils.

The average concentration of arsenic in unconsolidated sediments such as sand, clay, silt, and gravel etc. does not show much higher values but the ranges may be different as in alluvial

Table 1. Arsenic concentrations in different rocks (Takeshi, 1988 and Smedley and Kinniburgh, 2002)
| Rock type                             | Range, ppm | Mean, ppm |
|--------------------------------------|------------|-----------|
| Igneous rocks                         |            |           |
| Ultra basic rocks                     | 0.03-15.8  | 1.5       |
| Basic extrusive                      | 0.18-11.3  | 2.3       |
| Basic intrusive                       | 0.06-28.0  | 1.5       |
| Intermediate extrusive                | 0.5-5.8    | 2.7       |
| Intermediate intrusive                | 0.09-13.4  | 1.03      |
| Acid extrusive                       | 3.2-5.4    | 4.3       |
| Acid intrusive                        | 0.18-15.0  | 1.29      |
| Sedimentary rocks                     |            |           |
| Recent sediments                      | 1-13,000   | 14.1      |
| Ocean sediments                       | 0.4-455    | 33.7      |
| Shale, argillite                      | 0.3-500    | 14.5      |
| Sandstone, arkose, conglomerate       | 0.6-120    | 4.1       |
| Limestone, dolomite                   | 0.1-20.1   | 2.6       |
| Iron rich sediments                   | 1-2900     | -         |
| Gypsum                                | 0.1-10     | 3.5       |
| Phosphorite                           | 3.4-100    | 14.6      |
| Metamorphic rocks                     |            |           |
| Sedimentary origin quartzite          | 2.2-7.6    | 5.5       |
| Regional metamorphism gneiss          | 0.5-4.1    | 1.5       |
| Amphibolite, greenstone               | 0.4-45     | 6.3       |
| Contact metamorphism rocks            | 0.7-11     | 5.9       |

Sediments (3–10 ppm), lake sediments (0.9–44 ppm), glacial till (1.9–170 ppm), and aeolian deposits (5.4–18 ppm), etc. Arsenic in placer deposits may be much higher in some localities if there are considerable amounts of sulfide minerals such as pyrite (Smedley and Kinniburgh, 2001). The arsenic content in recent and old alluvial sediments in Bangladesh is much higher than the normal concentration due to the occurrence of arsenic-enriched pyrites at different levels below the ground surface (Alam et al. 2002).

Table 2. Arsenic concentrations in different soils (Takeshi, 1988)
Soil | Range, ppm | Mean, ppm
--- | --- | ---
Soils worldwide | 0.1-55 | 7.2
Soils formed from different rocks
From igneous rocks
Extrusive | 8-31.9 | 20
Intrusive | 13.9-16.9 | 15.4
From sedimentary rocks
volcanic ash | 20.3-31.6 | 24.8
clastic rocks | 14-51.3 | 25.6
From metamorphic rocks | 10.9—25.8 | 16.9

**Arsenic minerals in Sri Lankan rocks and soils**

No records of the occurrence of minerals that contain arsenic include arsenopyrite (FeAsS), realgar (AsS), orpiment (As$_2$S$_3$), and arsenolite (As$_2$O$_3$) in the parent rocks, especially in the plutonic igneous origin rocks (1%) in Sri Lanka currently exist. However, there are many accessory minerals in crystalline rocks (metamorphic and igneous rocks) such as sulphide-group metallic minerals; namely pyrite (FeS$_2$), chalcopyrite (CuFeS$_2$), pyrrhotite (Fe$_5$S$_6$), Molybdenum (MoS$_2$), and Galena (PbS). These minerals occur as minor accessory minerals in the crystalline rocks. Magnetite, ilmenite, rutile, zircon, apatite, sphene, and graphite are the other accessory minerals in Sri Lankan rocks. The magnetite in iron ore deposits is associated with these sulfide minerals. Higher arsenic content may be higher in these small magnetic iron ore deposits but only in few localities. The sedimentary rocks (nearly 10%) in the NW and north are mainly limestone. In some localities, the limestone is covered by ferruginous gravels and red earth (Cooray, 1994). There are Jurassic age sedimentary rocks, namely shale and sandstone in two locations. Those are also very small deposits. Graphite, mica, feldspar, apatite (carbonatite rock), limestone, marble and dolomite, river sand, mineral sand, and gems are the major mining sites in Sri Lanka. Except for apatite (carbonatite), others are not arsenic-releasing mines or metal-extracting mines. There are several hot water springs in the eastern part of the country. All hot springs occur in the flat terrains in the Dry Zone of Sri Lanka (Fig.1). The reasons for the increase in water temperature have not been identified yet (Jayawardena, 1988, Premasiri et al., 2006). There are no very clear deposits of chemical sediments around these hot springs. Ferruginous gravel deposits and red earth deposits are the major quaternary sediments above the limestone bed along the northwest coastal zone (Fig.2).

**Method Of Study**

**Sample collection**
Limestone is the major sedimentary rock in the country but there are no good weathering profiles in this carbonate rocky terrain. Hence, it is not selected for the investigation. The distribution of igneous rocks (carbonatite, granite, dolerite, vein quartz, and pegmatite) is about 1% or less in the total land of Sri Lanka. Only carbonatite (apatite rock) was selected to collect samples for this investigation. Samples were collected from metamorphic rocks (about 90% of the country) and their weathered grades in different localities. Most of the samples were collected from the hilly region where the thick weathering profiles can be seen. A few samples were collected from some other locations, which are nearly flat with isolated hills and uneven terrains.

Representative locations were selected to collect samples from different rock types for testing. 49 out of 50 locations show metamorphic rock types and the other shows an igneous rock. Charnockite/charnockitic gneiss, garnet sillimanite gneiss, hornblende biotite gneiss, migmatite, biotite gneiss, and quartzite are the major metamorphic rocks types in Sri Lanka. Pink granitic/microcline gneiss, calc gneiss, and amphibolite are not the major rock types but samples were collected. Carbonatite (apatite rock) was the only igneous rock. Before obtaining the samples, the exposed surfaces were cleared to avoid the mixing of other materials. Fresh, irregular samples, similar to the same size were obtained from rock quarries and outcrops. Generally, the sample block size was approximately 10 × 10 × 10 cm$^3$. The degree of weathering of rocks was identified according to the standard field methods (Fookes and Horswill, 1970). Weathered samples were collected from in-situ weathered formations above the particular parent rock at the same place or an adjacent place very close to the fresh rock. The samples represented their grade of weathering according to the field classification. The weathering grades are slightly weathered (SW), moderately weathered (MW), highly weathered (HW), completely weathered (CW), and residual soil (RS). CW samples and residual soils were collected in polyethylene bags and each bag was sealed and numbered to indicate the field.

Table 3. Total number of samples collected from different rock types and different grades of weathering
| Rock Type                | Climatic Zone | Number of Locations | FR | SW | MW | HW | CW | RS |
|-------------------------|---------------|---------------------|----|----|----|----|----|----|
| Ch/ChGn                 | WET           | 15                  | 15 | 15 | 15 | 15 | 15 | 15 |
|                         | DRY           |                     | 8  | 8  | 8  | 8  | 8  | 8  |
| HbBtGn*                 | WET           | 8                   | 8  | 8  | 8  | 8  | 8  | 8  |
|                         | DRY           |                     | 5  | 5  | 5  | 5  | 5  | 5  |
| GtSilGn                 | WET           | 8                   | 4  | 5  | 8  | 8  | 8  | 8  |
| Pink GrGn               | DRY           |                     | 1  | 1  | 1  | 1  | 1  | 1  |
| Calc Gneiss             | WET           | 2                   | 2  | 2  | 2  | 2  | 2  | 2  |
| Amphibolite             | WET           | 1                   | 1  | 1  | 1  | 1  | 1  | 1  |
| Quartzite               | WET           | 1                   | 1  | 1  | 1  | 1  | 1  | 1  |
| Apatite rock            | DRY           |                     | 1  | 1  | 1  | 1  | 1  | 1  |
| All Rocks               | Both          | 50                  | 46 | 47 | 50 | 50 | 50 | 50 |
| **TOTAL**               | **50**        | **293 Samples**     |

*Ch/ChGn = Charnockite/ Charnockitic gneiss, HbBtGn* = Hornblende biotite gneiss (*including Migmatite and garnet bearing biotite gneiss), GtSilGn = Garnet sillimanite gneiss, Pink GrGn = Pink Granitic Gneiss, Apatite rock = Carbonatite.

The uppermost materials (about 1 m below surface level) were not considered as residual soils but were treated as transported surface materials. Black soils near the residual soil level were also rejected assuming that the soils were mixed with organic materials. The localities of the 50 sampling points are shown in Fig. 1. The sample collection was carried out in two stages. The total number of samples collected was 293 (Table 3).

**Preparation of samples**

About 10 grams of each sample were pulverized by employing a sample vibration mill (HEIKO, Model No T1-100). Every attempt was made to avoid contamination of the powdered material. After preparation, the powder sample was inserted into a separate small polythene bag and sealed. This powdered sample of the particular specimen was used for its entire dry analysis.

**X-ray fluorescence analysis**
Before X-ray fluorescence analysis, a small part of each powdered sample was used to find out the H$_2$O (+) and H$_2$O (-) using the ignition loss method. Major elements and selected minor elements were analysed by X-ray fluorescence (XRF) spectrometry on a RIX 3100 system (KG-X system), manufactured by RIGAKU Denki Kogyo Company Limited, Japan at the Department of Earth Resources Engineering of Kyushu University, Japan, (Jayawardena and Izawa, 1993), (Jayawardena, 2000). The major elements are SiO$_2$, TiO$_2$, Al$_2$O$_3$, Fe$_2$O$_3$, MnO, MgO, CaO, Na$_2$O, K$_2$O, and P$_2$O$_5$. The minor elements are S, Cl, F, Cr, Ni, Zn, As, Zr, Pb, Ga, Rb, Y, Ca, Sr, Ba, Co, and Nb. The detection limits of the calibrated XRF spectrometry are in parts per million (ppm).

**Results And Method Of Analysis**

The total results of all minor elements are given in Annexure 1 as a Supplementary Table. Arsenic (As) is one of the selected minor elements. The results obtained after the testing can be summarized in different ways. Table 4 shows the maximum and minimum concentrations of arsenic in different rock types and their weathering grades without considering the climatic zones as Dry or Wet. Rather than indicating the concentrated amount for each sample, it is better to divide it into two different ranges to explain its distribution. Generally, the average arsenic concentration in metamorphic rocks is 5 ppm (Smedley and Kinniburgh, 2001). Accordingly, the number of fresh rock samples less than 5 ppm arsenic and more than 5 ppm arsenic were found and is given in Table 5a as a percentage. Takeshi (1988) found that the minimum arsenic content in soils formed from weathered metamorphic rocks was 10.9 ppm (Table 2). Therefore, for this study, 10 ppm of arsenic in weathered rocks and residual soils was considered as the upper limit. The number of weathered rocks and soil samples that show 10 ppm or less than 10 ppm of arsenic concentration is given in Table 5b.

Table 4. Arsenic concentrations as ppm in different fresh rocks and their weathering grades and total summary
| Rock Type                          | Climatic Zone (Fig.2) | As concentration (ppm) in different fresh and weathered rocks | FR  | SW  | MW  | HW  | CW  | RS  |
|-----------------------------------|-----------------------|----------------------------------------------------------------|-----|-----|-----|-----|-----|-----|
| Charnockite/Charnockitic gneiss   | Both                 | Minimum                                                        | 0   | 0   | 0   | 1   | 2   | 1   |
|                                   | Zones                | Maximum                                                        | 12  | 18  | 17  | 46  | 18  | 27  |
| Hornblende Biotite Gneiss         |                      | Minimum                                                        | 0   | 0   | 0   | 0   | 0   | 0   |
|                                   |                      | Maximum                                                        | 7   | 13  | 13  | 9   | 5   | 24  |
| Garnet Sillimanite Gneiss         |                      | Minimum                                                        | 0   | 2   | 0   | 2   | 0   | 3   |
|                                   |                      | Maximum                                                        | 6   | 9   | 9   | 14  | 48  | 31  |
| Some other rock types             |                      | Minimum                                                        | 0   | 1   | 0   | 1   | 3   | 3   |
|                                   |                      | Maximum                                                        | 7   | 9   | 9   | 9   | 10  | 10  |
| Summary                           | Both                 | Minimum                                                        | 0   | 0   | 0   | 0   | 0   | 0   |
|                                   | Zones                | Maximum                                                        | 12  | 18  | 17  | 46  | 48  | 31  |

Table 5a. Summary of Arsenic concentration more than 5ppm (>5ppm) or less than 5ppm (5ppm >) in fresh rock as a percentage
| Arsenic concentration | Charnockite/ Charnockitic Gneiss | Hornblende Biotite Gneiss | Garnet Sillimanite Gneiss | Other rock types | ALL (total) |
|-----------------------|----------------------------------|--------------------------|--------------------------|-----------------|-------------|
| Less than 5ppm        | 20                               | 12                       | 3                        | 5               | 40          |
| More than 5ppm        | 3                                | 1                        | 1                        | 1               | 06          |
| Total Samples         | 23                               | 13                       | 4                        | 6               | 46          |
| Percentage of samples | 86.9                             | 92.3                     | 75.0                     | 83.3            | 86.9        |
| Less than 5ppm        | 13.1                             | 7.7                      | 25.0                     | 16.7            | 13.1        |

Table 5b. Summary of Arsenic concentration more than 10ppm (>10ppm) or less than 10ppm (10ppm> ) in weathered rocks and residual soils as a percentage
| Weathering Grade          | Number of weathered rock/soil samples which show Arsenic concentration less or more than 10ppm | Charnockite/Charnockitic Gneiss | Hornblende Biotite Gneiss | Garnet Sillimanite Gneiss | Other rock types | ALL (total) |
|---------------------------|-----------------------------------------------------------------------------------------------|---------------------------------|---------------------------|--------------------------|------------------|-------------|
|                           |                                                                                               |                                 |                           |                          |                  |             |
| Slightly Weathered        | 20                                                                                           | 11                              | 5                         | 6                        |                  | 42          |
| Moderately Weathered      | 21                                                                                           | 12                              | 8                         | 6                        |                  | 47          |
| Highly Weathered          | 21                                                                                           | 13                              | 7                         | 6                        |                  | 47          |
| Completely Weathered      | 21                                                                                           | 13                              | 6                         | 6                        |                  | 46          |
| Residual Soil             | 16                                                                                           | 12                              | 6                         | 6                        |                  | 40          |
| No. of Samples less than 10 ppm | 99                                                               | 61                              | 32                        | 30                       | 222              |             |
| Total Samples             | 115                                                                                          | 65                              | 37                        | 30                       | 247              |             |
| % of samples              | 86.0                                                                                         | 93.8                            | 86.5                      | 100.0                    | 89.8             |             |
| Less than 10 ppm          | 14.0                                                                                         | 6.2                             | 13.5                      | 0.0                      | 10.2             |             |

The results indicate that there is no arsenic in 6 fresh rock samples and 19 weathered rock samples (total 25). There is no meaning to calculate an average value using zero ppm. Therefore to calculate the average, 1.0 ppm arsenic concentration was used instead of zero ppm of those 25 samples. Table 6 shows the total arsenic values in each sample and the average values after adding 1 ppm instead of zero ppm.

Table 6. As concentrations in total 293 samples and average values after added 1 ppm for zero values.
| Sample Location No | FR | SW | Mw | Hw | CW | RS |
|--------------------|----|----|----|----|----|----|
| 1                  | 5  | 7  | 6  | 4  | 9  | 27 |
| 2                  | 8  | 9  | 10 | 9  | 9  | 13 |
| 3                  | 4  | 5  | 8  | 9  | 10 | 10 |
| 4                  | 12 | 12 | 11 | 9  | 8  | 1  |
| 5                  | 5  | 18 | 17 | 11 | 8  | 10 |
| 6                  | 1  | 5  | 2  | 0  | 3  | 2  |
| 7                  | 2  | 4  | 1  | 2  | 0  | 1  |
| 8                  | 1  | 0  | 1  | 3  | 0  | 1  |
| 9                  | 5  | 1  | 4  | 5  | 3  | 2  |
| 10                 | 2  | 4  | 4  | 8  | 6  | 7  |
| 11                 | 3  | 2  | 10 | 2  | 8  | 9  |
| 12                 | 7  | 8  | 8  | 7  | 9  | 20 |
| 13                 | 1  | 2  | 2  | 1  | 1  | 1  |
| 14                 | 4  | 1  | 2  | 2  | 4  | 18 |
| 15                 | 4  | 4  | 5  | 6  | 9  | 11 |
| 16                 | 0  | 11 | 10 | 46 | 13 | 9  |
| 17                 | 5  | 6  | 6  | 10 | 8  | 12 |
| 18                 | 4  | 1  | 1  | 1  | 5  | 2  |
| 19                 | 1  | 0  | 1  | 1  | 2  | 2  |
| 20                 | 5  | 2  | 5  | 1  | 4  | 8  |
| 21                 | 3  | 5  | 5  | 4  | 4  | 4  |
| 22                 | 0  | 0  | 0  | 0  | 10 | 14 |
| 23                 | 9  | 0  | 0  | 0  | 16 | 2  |
| 24                 | 1  | 2  | 5  | 8  | 48 | 31 |
| 25                 | 3  | 1  | 1  | 3  | 1  | 3  |
| 26                 | 2  | 1  | 2  | 2  | 2  | 3  |
| 27                 | 0  | 0  | 0  | 0  | 24 | 13 |
| 28                 | 1  | 4  | 3  | 3  | 8  | 8  |
|    | 2 | 2 | 1 | 2 | 3 | 2 |
|----|---|---|---|---|---|---|
| 30 | 7 | 6 | 7 | 2 | 5 | 8 |
| 31 | 3 | 2 | 6 | 2 | 10| 8 |
| 32 | 5 | 2 | 2 | 3 | 1 | 5 |
| 33 | 2 | 3 | 1 | 3 | 2 | 2 |
| 34 | 3 | 4 | 6 | 8 | 13| 7 |
| 35 | 3 | 12| 13| 9 | 5 | 7 |
| 36 | 2 | 3 | 1 | 2 | 3 | 2 |
| 37 | 5 | 4 | 4 | 3 | 6 | 8 |
| 38 | 0 | 9 | 0 | 0 | 25| 6 |
| 39 | 4 | 6 | 6 | 8 | 4 | 3 |
| 40 | 7 | 3 | 11| 5 | 7 | 16|
| 41 | *| 2 | 7 | 3 | 5 | 7 |
| 42 | *| *| 5 | 3 | 4 | 9 |
| 43 | *| *| 6 | 2 | 6 | 1 |
| 44 | *| *| 5 | 8 | 0 | 0 |
| 45 | 5 | 3 | 3 | 2 | 3 | 2 |
| 46 | 2 | 1 | 2 | 5 | 6 | 10|
| 47 | 3 | 1 | 4 | 5 | 5 | 9 |
| 48 | 0 | 1 | 2 | 1 | 3 | 3 |
| 49 | 0 | 9 | 9 | 10| 10| 18|
| 50 | 7 | 4 | 0 | 7 | 10| 5 |
| Added for zero ppm | 6 | 5 | 5 | 5 | 3 | 1 |
| Average separately | 3.5| 4.1| 4.7| 5.1| 7.4| 7.6|
| Average | Fresh Rock | Weathered Rocks  | 5.2 ppm |
|         | 3.5 ppm     |

*Samples were not collected

**Discussion**
Previous results by others

There may be some investigations under various objectives carried out by different scientists and organizations for agricultural soils and stream sediments from shallow depths. Those samples are not residual soils related to the parent rock and hence cannot be considered as natural residual earth material. Therefore, the arsenic concentrations in those samples carried out by previous researchers are not related to the original fresh rock. The results from the present investigation indicate the distribution of arsenic that is related to the parent rocks and their weathered grades only.

Arsenic content in general

Table 6 shows the distribution of arsenic amounting to 293 samples analysed by XRF spectrometry in different ways. The results for different rock types given in Table 4 show the maximum and minimum amount of arsenic concentration in fresh and weathered rocks in Sri Lanka. Accordingly, the highest arsenic amount (48 ppm) was found from one completely weathered (CW) rock sample of garnet sillimanite gneiss. The overall results indicate that the average arsenic concentration in fresh rocks is 3.5 ppm and 7.6 ppm in residual soils. The average in total weathered rocks is 5.2 ppm (Table 6). This is similar (or less than) to the average arsenic concentration in metamorphic rocks from other countries (Takeshi, 1988 and Smedley and Kinniburgh, 2001). With the continuation of weathering, rocks are converted into the soil and the composition of the soil may vary with the number of clay minerals and other altered primary minerals. The average results indicate that the arsenic concentration increases slightly Table 6. As concentrations in total 293 samples and average values after added 1 ppm for zero values.

with the increase of weathering and the formation of soil. This increase of arsenic in soils may be due to (a) absorption by clay minerals and other secondary minerals; (b) altered primary minerals such as ferrous and ferric oxides, aluminium oxide, goethite, limonite and bauxite, laterite, etc.; (c) concentration of non-solubility in groundwater; and (d) movement of fine sediments with the groundwater from one place to another.

Arsenic content in fresh rocks

In general, the average arsenic concentration in metamorphic rocks is 5ppm (Smedley and Kinniburgh, 2001). Considering that the fact in Table 5a was made to show the percentages of distribution of arsenic as 5 ppm or less than 5ppm (5ppm >) in fresh rocks of this study. 86.9% of chamockite (20 of 23 samples), 92.3% of horblende biotite gneiss (12 of 13 samples), 75 % of garnet sillimanite gneiss (3 of 4 samples), and 83.3% of other rock types (5 of 6 samples including apatite rock) show the arsenic concentration is less than 5ppm. 86.9% of the total samples (40 of 46 samples) show less than 5ppm arsenic content (Table 5a). These results indicate that metamorphic rocks of Sri Lanka cannot be the source rocks for the distribution of arsenic on the Island.

Arsenic content in weathered rocks
Table 2 indicates that the arsenic distributions in residual soils are formed from metamorphic rocks in Japan. It varies from 10.9 to 25.8ppm and the average is 16.9ppm. In Sri Lanka, the maximum is 31ppm but the average is 5.2ppm (Table 6), less than the minimum amount of soils in Japan. Table 5b shows a summary of the variation of arsenic content in naturally weathered rock materials and residual soils. To classify the arsenic content, 10 ppm of arsenic in a sample was considered as the upper limit in soils. The percentage of arsenic content less than or equal to 10 ppm in different weathered rock samples is different but the average for the total weathered samples including residual soils is 89.8%. Only a few samples of weathered rocks and soils have arsenic more than 10 ppm. This analysis does not indicate whether the arsenic content in Sri Lankan soils is soluble or non-soluble. However, the average arsenic content in fresh rocks is 3.5ppm and is 7.6ppm in residual soil (Table 6). This shows that the arsenic content in fresh rocks is concentrated in residual soils and is increased by weathering due to its non-soluble behaviour.

Garnet sillimanite gneiss is a fast weathering metamorphic rock in Sri Lanka. Generally, unlike other rock types, very fresh rock samples of garnet sillimanite gneiss cannot be seen at the surface but such samples that experienced weathering are available in wet zones within the country. The highest arsenic concentration (48ppm) was found in completely weathered (CW) rocks of garnet sillimanite gneiss (Table 4). Recent investigations carried out by Dissanayake and Chandrajith (2003) found that the arsenic content in river sediments in Sri Lanka is less than 7 ppm. Therefore, it can be assumed that the arsenic concentration in alluvial deposits also is similar to the recent measurements due to the same parent rock condition.

The arsenic content in fresh apatite minerals is 7 ppm and increases up to 10 ppm in highly weathered grades (Annexure 1). Jayasumana et al.(2015) claimed that the range of arsenic in apatite is from 3.4 to 21.8 ppm. The average arsenic content in other intrusive rocks may be less than 5 ppm.

**Conclusion**

The results indicate that the maximum arsenic amount in fresh rock is 12 ppm and 48 ppm in completely weathered rocks. 86.9% of the samples (total 46 samples) from different fresh rock types show less than 5 ppm arsenic content. 89.8% of their weathering grades (a total of 247 samples) showed that the arsenic concentration is less than 10 ppm. The average arsenic in all fresh rock samples is 3.5 ppm (lowest average) and is 7.6 ppm (highest average) in residual soils. This is the normal condition of arsenic distribution worldwide; under this condition, the arsenic concentration in natural groundwater in the residual soil areas should be below the acceptable limit. Therefore, the amount of arsenic released from parent rocks and their weathered products due to natural geological processes is very low in Sri Lanka.

**Declarations**

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All data used for this analysis and preparation of the manuscript are the results of my laboratory experiments at the University of Kyushu, Fukuoka, Japan. Samples were collected from various localities in Sri Lanka before my arrival to Japan. All data have been attached as a supplementary Table. These data have not been published previously in any local or international journal.

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Figure 1

Different climatic boundaries of Sri Lanka (NARESA,1991). The black dots indicate the locations of samples collected for laboratory tests. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research
Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

**Figure 2**

Subdivisions of the geology of Sri Lanka (Cooray, 1994) Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.
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