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Assessment of heavy metal content in rice and soil samples from rainfed paddy fields in Yangon Division, Myanmar

（ミャンマー・ヤンゴン管区の天水水田から採取した米および土壌試料中の重金属含有量評価）

Division of Environmental Science Development
Course in Global Environmental Management
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Finally, I would like to express my apology for not mentioning every personal name here.

Abstract

Much research focused on arsenic concentration in rice arising from irrigation water than the rainfed area in Southeast Asia. However, limited information exists on the major rice-growing area, Ayeyarwaddy, Myanmar. In this study, the heavy metal analysis of rice grains and soil was performed to determine the concentration of rice due to the geological background.

A total of 120 polished rice, 200 unpolished rice, and 82 soil were collected from the most rice-growing active townships near Yangon on the eastern edge of the Ayeyarwady Delta. A total of 96 polished rice (n = 50, factories) (n = 46, markets) were also collected from factories in the delta and trading market at Yangon. Heavy metals (As, Pb, Cd, Cr, Mn, Fe, Zn, Cu, Ni, Mo, and Co) were determined by triple quadrupole inductively coupled plasma mass spectrometry (ICP–QQQ). The total–As (0.16 ± 0.05) and Ni (0.39 ± 0.19) were found in Yangon polished rice and mean concentrations of Pb, Cd, and Cr (0.010, 0.0056, and 0.056) mg/kg in this rice were observed which are lower than the maximum allowable levels by over one magnitude.

The mean and range of soil As in TK, DL and KM were 1.14 (0.67−1.52), 4.90 (3.41−8.50), and 5.26 (3.48−7.26) mg/kg. The range of As concentration in soil (0.67–8.50) and Cd (0.017–0.03) of this study fell into typical rainfed soil As and Cd range (0.8 – 18) (0.06–1.1) mg/kg which can be assumed there are no manmade activities. The positive linear relationships was found between soil As and unpolished rice As (r^2 = 0.8, P <0.05). Total soil As in this study had a significantly straightforward relationship with grain As. Therefore, the amount of grain As in flooded rice paddies is more heavily influenced by soil chemical conditions. The moderate negative relationship was found between soil Ni and unpolished Ni (r^2 = –0.4, P <0.05). It can reveal that paddy grain Ni was not due to paddy soil and it might be due to other agricultural activities.

Health risks associated with rice consumption by the inhabitants were estimated, assuming that inorganic arsenic was 30% of the total. Arsenic was the main contributor (30%) to the total value of the non–cancer risk (HI) of each element, which was 4.5 times the reference value (< 1), followed by Mn, Zn, Cu, Mo, Co and Ni (15–7%) and Pb, Cd, Cr and Fe (below 4%). The total cancer risk (TCR) for each element was around 17 times higher than the upper limit of cancer risk for an environmental carcinogen (< 0.0001): Nickel accounts for two–thirds of the contribution (66%), followed by Cd (16%) and As (13%). This study suggests that consumers of Yangon rice from paddy fields without groundwater irrigation may need to be concerned about the potential risk of Ni intake, besides arsenic.

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List of abbreviations

BW…….Body Weight

CRMs ……Certified reference materials

EDI ……Estimated daily intake

HI ……Hazard index

HQ…… Hazard quotient

ICP–AES …….Inductively Coupled Plasma Atomic Emission Spectrometry

ICP–QQQ…… ……Inductively Coupled Plasma Mass Spectrometry Triple quadrupole

IR ……Ingestion rate

NMIJ……National Metrology Institute of Japan

MAC…….Maximum allowable concentration

PTWI……...…… Permissible Tolerable Weekly Intake

TDI……...……Tolerable Daily Intake

RDA…… Recommended daily allowance

RfD …… Oral Reference Dose

SF…… Slope factor

TCR…… Total cancer risk

WHO…….....…. World Health Organization

USEPA…….. United States Environmental Protection Agency

Chapter 1- General Introduction

1.1 Heavy metal in the environment

Heavy metals are metallic elements and their densities are at least five times higher than water. Heaviness and toxicity are assumed to be interrelated, some metalloids can induce toxicity at a low level of exposure (Tchounwou et al., 2012). Heavy metals are found in the biosphere, such as in water, soils, and rocks, and are also released into the environment through natural and human activities. They are composed in the earth's crust and come into the environment naturally during the physical and chemical weathering of rocks. Natural phenomena like volcanic eruptions may release heavy metals. Human activities such as agriculture, industrial, domestic, medical, mining, and technological applications discharge them into the environment.

The concentrations of heavy metals released into soil systems by natural processes from the background are low and depend largely on the origin and nature of the parent materials (Kunhikrishnan et al., 2015). Heavy metals are vital for survival, but they may become toxic when they accumulate in organisms. Ni, Cu, and Zn are essential elements that are widespread, and manganese, the most plentiful element, is formed in various oxidation states and is found all around the world about 0.1 percent of the earth’s crust. Co is found in abundance across the environment, such as vegetation, soils, rocks, and water (Mitra et al., 2022). Cd is widely distributed in the earth's crust at an average concentration of about 0.1 mg/kg. The highest level of cadmium compounds in the environment is accumulated in sedimentary rocks (WHO, 1987). Cr is a naturally occurring element present in the earth’s crust, in the form of ores, such as ferrochrome. Pb can be formed naturally occurring bluish–gray metal present in small amounts in the earth’s crust (Tchounwou et al., 2012).

1.2 Heavy metal concentration in rice

Rice is a staple food for half of the world's population. Approximately 480 million metric tons of milled rice are produced annually in the world (Tyagi et al., 2020). Rice contains high content of fibers and carbohydrates, a medium content of minerals and vitamins, and low content of proteins. Rice is the most important staple food for half of the world's population and it supports 30% of the dietary energy and 20% of the dietary protein in Asia (WHO, 2003).

Rice consumption as a daily intake of 0.5 kg in Asian countries and it is the primary staple food in southeast Asian countries. Figure 1.1 shows the rice consumption rates from higher to lower by the country according to FAO (2004). The highest consumption rate is found in Myanmar and most of the south and east Asian countries like Laos, Viet Nam, Cambodia, Bangladesh and Indonesia have the highest rice consumption (Fig. 1.1)

The concentrations of heavy metals in food kinds of stuff becomes a major concern in the world. Rice, cereal, and vegetables are the main diet for humans and it contributes to the intake of essential elements like Cr, Co, Cu, Fe, Mn, Ni, and Zn (Sharafi et al., 2019). It may contain toxic metals such as As, Pb, and Cd and they are non–essential elements however excessive intake of essential and toxic metals may lead to health problems.

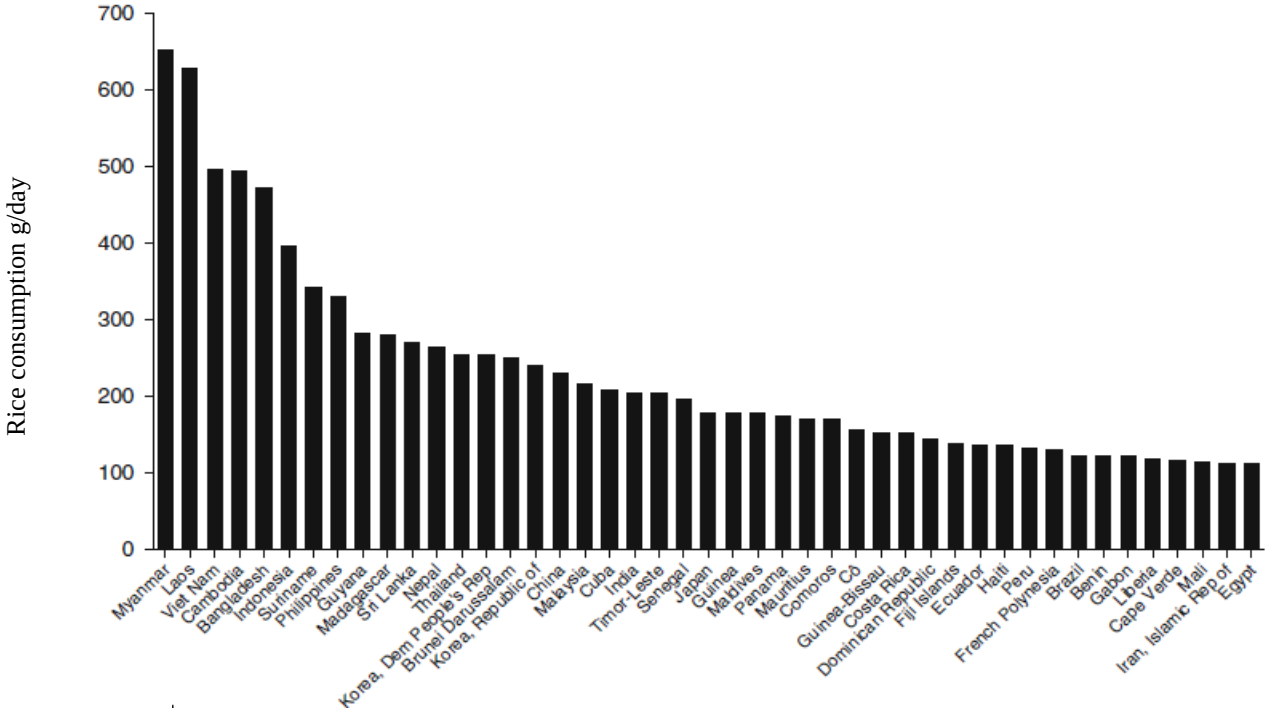


Fig. 1.1. Rice consumption rates by countries (FAO, 2004).

1.2.1 Heavy metal concentration in rice due to irrigation water

The heavy metal concentration in rice is a global issue and irrigation with the groundwater resulted in severe arsenic contamination in paddy soils and rice grains. However, the consumption of rice can cause the intake of heavy metals into the human body and it has been a serious concern. Many rice types of research had been done in many countries like Bangladesh, India, China, and the United States and there is limited attention given to some developing southeast Asian countries.

The researches mainly focused on the problem associated with irrigation rice and they had been explored the elemental concentration of domestic and export rice in many countries. Arsenic has been recorded as a toxic metal. Zavala & Duxbury, (2008) reported the global normal range of As concentration in rice is 0.08–0.20 mg/kg. They survey the concentration of As from some countries. The range of As from the market rice grain is

0.31–0.70 mg/kg found in China, <0.10–0.76 mg/kg for Taiwan, 0.11–0.66 mg/kg for the U.S., and 0.08–0.38 mg/kg for Italy and Spain. Ojinnaka study As concentration in rice of Bangladesh is <0.01–2.05 mg/ kg and 0.044–0.03 mg kg in India, and 0.03–0.47mg/kg in Vietnam, (Zavala & Duxbury, 2008). The average As concentration in rice grain is 0.2 mg/kg and ranged from 0.1 to 0.37 mg/kg in Cambodia (Seyfferth et al., 2014).

The Cd and Pb concentrations in rice grains from different countries are in the range of 0.01 to 0.05 and 0.001 to 0.5 mg/kg. Falahi et al., (2010) revealed that the mean concentration of Cd, Cu, Pb, and Zn in the USA were 0.06, 4.15, 0.250, and 43.4 mg/kg and the concentrations of Cd, Cu, Pb, Ca, Fe, and Zn were 0.021, 1.85, 0.206, 94.8, 10.2, and 17.0 mg/kg in South Korea. Pb and Cd of North Iran rice grain were 0.962 and 0.079 mg/kg (Falahi et al., 2010). Ni (0.81±0.72), Pb (0.94±0.70), Zn (8.22±2.97), Cu (0.59±0.42), and Cd (0.36±0.07) were found in Nigerian rice (Ezeofor et al., 2019). The range of the metals detected in rice grain samples was 0.67–0.8 mg/kg for cadmium, 2.14–7.0 mg/kg for Cu, 1.85–2.68 mg/kg for Ni, and 0.94–3.28 mg/kg for Pb in Malaysia (Rashid et al., 2018).

1.2.2 Heavy metal concentration in rice due to soil

Researchers have focused on the heavy metal concentration of rice from irrigation water or anthropogenic activities, but they have less attention to report on geogenic sources. The research related to rainfed rice is discussed as follows.

Ying Lu et al. conducted on rice from a rainfed area of Bangladesh in which the mean concentration of As in rainfed rice grain is 0.186 mg/kg and soil samples were collected in nonirrigated soil nearby paddy fields and the baseline soil arsenic level ranged from 0.8–21 mg/kg. The lower concentrations found on the Pleistocene Terrace around Gazipur, it was from 0.8 to 2.4 mg/kg. The higher levels of concentrations were found at a range of 3.4 to 21 mg/kg (Y. Lu et al., 2009). Rice produced even from low soil arsenic paddies not irrigated with groundwater is a major source of arsenic to the population. Consumption of 0.5 kg of this rice would be a daily intake of 130 μg/d of arsenic, most of which is inorganic and it suggested rice was a main As source for their population.

Angelia L. et al. (2014), studied an assessment of As in unpolished rice and within the paddy soil in Cambodia where groundwater is seldom used for rice irrigation. The total soil As concentrations ranging 0.8 to 18 mg/kg, and total grain As concentrations averaged 0.2 mg/kg and ranged from 0.1 to 0.37 mg/kg. Regression analyses were performed to find potential relationships between grain and soil As. However, none of the soil parameters obtained were significantly correlated with grain As.

In the study of Wang H. et al. (2020), they assessed the elemental analysis of soil samples and rice grains in Xuyi, China. The topsoil showed relatively high levels of Cr (86–279 mg/kg), Ni (45–269 mg/kg), and Zn (57–127 mg/kg), moderate levels of Cu (27–80 mg/kg) and Pb (12.20–38.20 mg/kg), and low levels of As (3.59–19.14 mg/kg) and Cd (0.05–0.21 mg/kg). Rice grain expressed relatively high levels of Zn, Cu, Ni, and Cr, and low levels of Pb, Cd, and As. There is no significant linear correlation between total soil Ni concentration and rice grains. A risk assessment analysis was done and the HI value of rice grain is 3.09 times the reference value of 1. Rice ingestion would cause a potential non–carcinogenic health risk to residents. HQ (Ni) (24.9%) and HQ (Cr) (21.7%) are the main contributors to the HI value of rice grain, followed by HQ (Cu) (14.2%) and HQ (As) (14.2%). The TCR value of rice grain is 7.54×10^{-3}.

1.3 Arsenic in the rice fields

Arsenic is classified as metalloid and it has metal and nonmetal properties. Arsenic can be organic and inorganic form in the environment and within human body. Arsenic combine with other elements like sulfur, oxygen and chlorine are inorganic arsenic and arsenic combine with carbon and hydrogen are organic arsenic. Arsenic found in marine organisms are well known as organic arsenic which has less toxicity. Inorganic arsenic are arsenite (As III) and arsenate (As V) and organic arsenic are monomethyl arsenic acid (MMA), dimethylarsonic acid (DMA), trimethylarsine oxide (TMAO), trtramethylarsonium ion (TeMA), arseno betaine (AB), arseno choline (AC), dimethylarsinylriboside, trimethylarsonioriboside, glyceryl phosphorylarsenocholine, phosphatidylarsenocholine, diphenylarsenic acid (DPAA) and methylphenylarsenic acid (MPAA) (Suriyagoda et al., 2018). Organic arsenic are not common type in soil

composition but inorganic arsenic are found as dominant type (Fig.1.2) (Suriyagoda et al., 2018). As (III) and DMA are much more soluble type and they can be more bioavailable than other arsenic forms. The alteration between organic and inorganic As can depend on many soil physical factors.

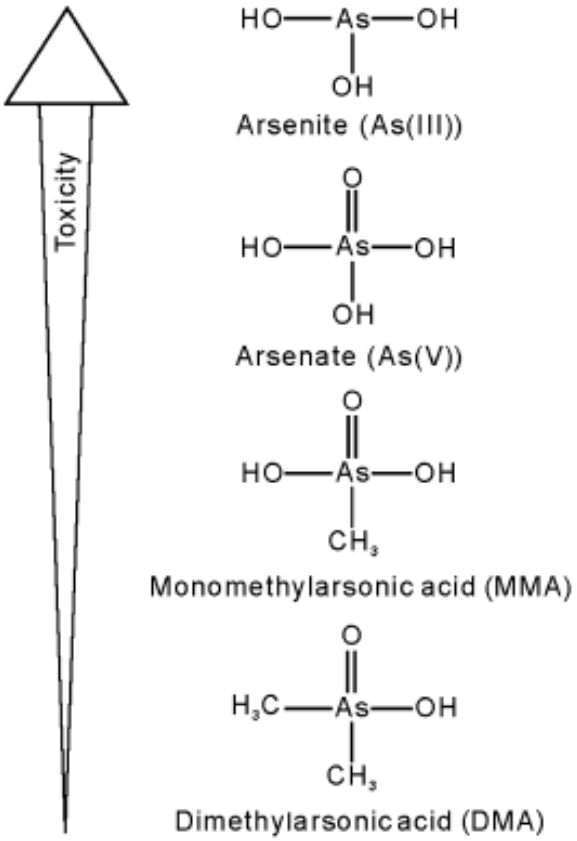


Fig 1.2. Most common form of As chemical structure (Suriyagoda et al., 2018).

In the flooding ricefields, arsenite (As III) is the most dominant arsenic species, Fig. 1.3 shows As situation in rice field. As mainly occurs in aquatic system as inorganic As species (As (III) and As (V)). Under anaerobic condition, the sediment, organic carbon and metal oxides located in the minerals of aquifers or As bearing iron hydroxides release As (III) with the help of anaerobic bacteria. In semi-arid or arid environment, the weathering of minerals and aerobic bacteria leads to desorption of adsorbed As (V). Inorganic As (III) dominants in flooded water or anaerobic condition (Herath et al., 2016) comprising 63% of total arsenic in soil, followed by arsenate As(V) at 36%, and

methylated arsenic species (Sinha & Bhattacharyya, 2020). Some studies said As(III) is 87%–94% of the total As in flooded soils, whereas As(V) covers 73%–96% of the total As in aerobic soil conditions (Das et al., 2008) (Yamaguchi et al., 2011).

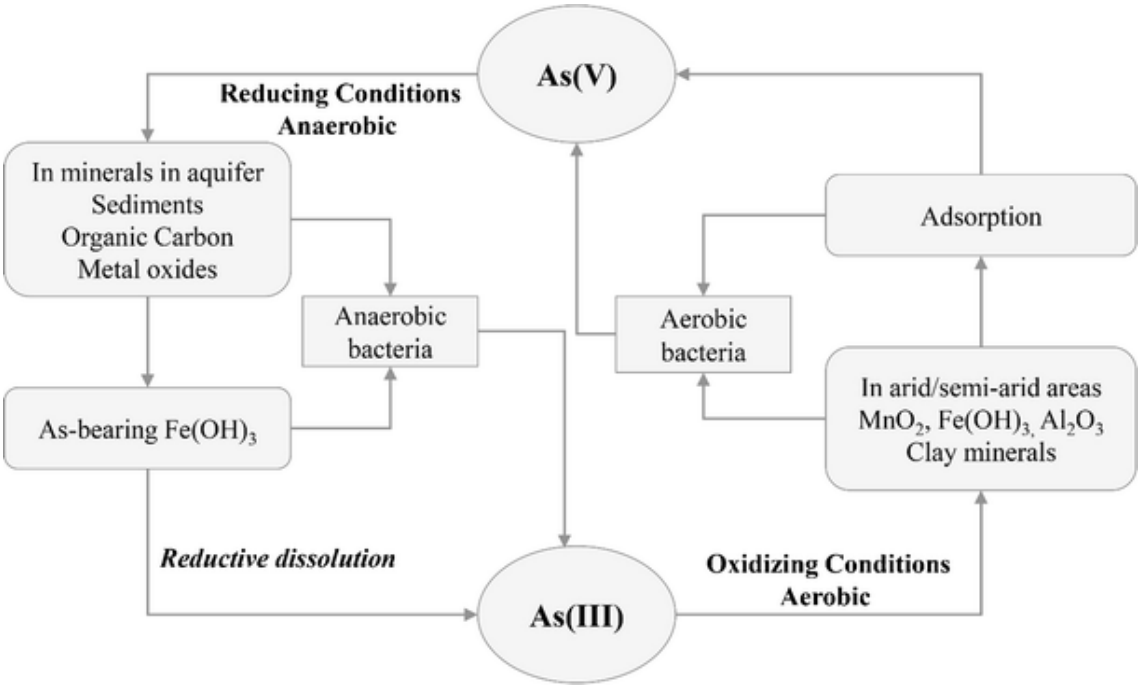


Fig. 1.3. Arsenic situation in rice field under reducing and oxidizing conditions.

Arsenic speciation accumulated in rice is important. Rice is a major dietary staple for large populations, especially in Asian countries, and elevated arsenic levels are found in rice and rice products available in marketplaces around the world (Nookabkaew et al., 2013). The main component species of total arsenic in rice grain are As (III) and As (V) which causes a range of cancers: lung bladder and skin being the most prominent as shown in Fig. 1.4 (Hassan et al., 2023). Chronic exposure to inorganic arsenic species is also implicated in a range of other negative health impacts such as hypertension, diabetes, and premature births (Meharg & Zhao, 2012).

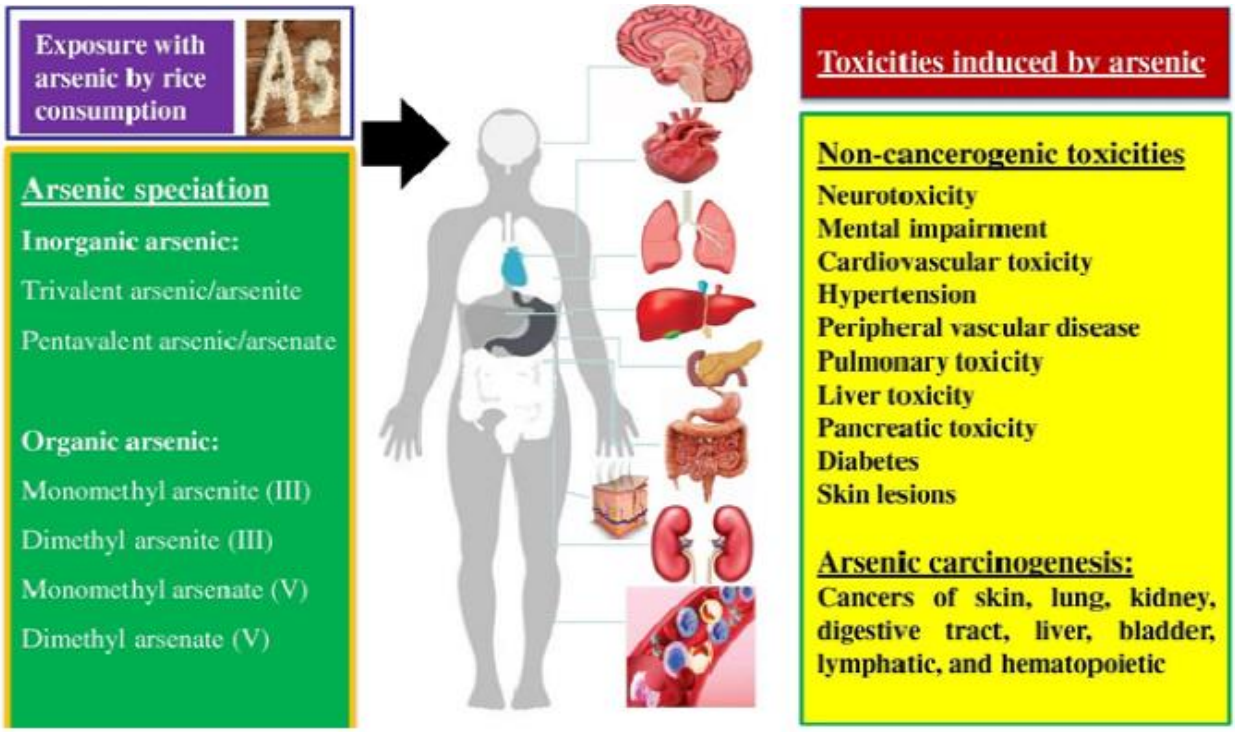


Fig. 1.4. Toxicities induced by arsenic in human body (Hassan et al., 2023).

Nookabkaew et al., (2013) studied arsenic speciation in Thailand, Vietnam, Pakistan, India, Sri Lanka, and Malaysia white rice and unpolished or brown rice. The result was As(III) and As(V) as inorganic As in the present study accounted for 53.1 and 67.2% of the total As in all types of rice. All rice samples can be recognized as the inorganic As type due to the high predominance of inorganic As in these rice samples (Nookabkaew et al., 2013).

Some research on the arsenic speciation of rice from the neighboring country is reviewed as follows. In the study of Sinha B. et al., (2020), the rice samples were collected from farmer fields in West Bengal, India, and arsenic speciation of rice was analyzed. Most of the As species in rice were noticed to be inorganic [Arsenite (As–III), Arsenate As(V)]. The order of As species in rice grain observed in this study was As(III) (54.5–65.4 %) > As(V) (21.2–28.3%) > DMA (5.2%) (Sinha & Bhattacharyya, 2020).

1.4 Nickel in the ricefields

Nickel can be found in earth crust, volcanoes and the soil naturally. Nickel and nickel compounds have many industrial and commercial uses and they are used in many kinds of human applications as shown in Table 1.1 (Genchi et al., 2020). Nickel is abundantly found everywhere in the earth. So the exposure of nickel to human can occur easily. It can be from the headaches to lung cancer.

Table 1.1. Nickel in human application and its toxic effect (Genchi et al., 2020).

Nickel Containing Foods	Hazelnuts; cocoa and dark chocolate; fruits (almonds, dates, figs, pineapple, plums, raspberries); grains (bran, buckwheat, millet, whole grain bread, oats, brown rice, sesame seeds, sunflower seeds); seafood (shrimps, mussels, oysters, crab, salmon); vegetables (beans, savoy cabbage, leeks, lettuce, lentils, peas, spinach, cabbage), tea from drinks dispensers; soya and soya products; peanuts; licorice; baking powder.
Nickel Containing Items	Inexpensive jewelry; cosmetics; keys; cell phones; eyeglass frames; paper clips; orthodontic braces; stainless steel articles; nickel plated articles; clothing fasteners (zippers, snap buttons, belt buckles); electrical equipment; armaments; alloy; metallurgical and food processing industries; pigments; catalysts.
Nickel Toxic Effects	Contact dermatitis; headaches; gastrointestinal manifestations; respiratory manifestations; lung fibrosis; cardiovascular diseases; lung cancer; nasal cancer; epigenetic effects.

In recent years, Ni contamination in agriculture soil caused by pesticides, fertilizers and animal manure usage have reported the increased Ni in the rice grains (Duan et al., 2012). According to da Rosa Couto et al., (2018) , the application of cattle manure in agriculture soil increased Ni concentration to the grains of corns and wheats. The agrochemical usages such as fertilizers and pesticides in agricultural land is a common practice and the usage of phosphate fertilizers can cause Pb, Co, Fe, Cd and Ni as impurities (Arunakumara et al., 2013).

1.5 Impacts of heavy metal on human health

A small number of trace metals in the environment may enter into the food chains through bio–magnification and their cumulative concentration under long–term exposure may

become toxic to both living organisms and human beings. The toxicity, persistence, and bioaccumulation of heavy metals are associated with health problems. Human exposure to heavy metals is in different routes such as inhaling dust, consumption of contaminated drinking water, ingestion of soil, and consumption of food crops grown on contaminated land (Salehipour et al., 2015). Essential trace elements of the human body include Zn, Cu, Cr, Co, Mn, and Mo, however, the excessive amount brings health problems. The human health problem associated with As, Pb, Cd, Cr, Mn, Fe, Zn, Cu, Ni, Mo, and Co is as follows.

1.5.1 Arsenic

Arsenic is a ubiquitous element at low concentrations in the Earth's crust. It can be found in soil and water that have interacted with arsenic–rich rocks and it can enter the environment through volcanic eruptions and soil erosion or weathering processes of rocks. It can also be discharged into the groundwater naturally or anthropogenically through the application of arsenic–rich herbicides and pesticides such as insecticides, herbicides, fungicides, algicides, sheep dips, wood preservatives, and dyestuffs (Tchounwou et al., 2012). Arsenic can also be discharged from the chemical, paints and tannery and other activities such as metal mining, sewage, smelting and municipal solid waste to the environment (Fig. 1.5.) (Sahoo & Kim, 2013). Exposure to arsenic occurs via ingestion, inhalation, dermal contact, and the parenteral route. High concentrations of As can cause various acute and chronic diseases such as skin lesions, cardiovascular diseases, neurological disorders, or even cancer.

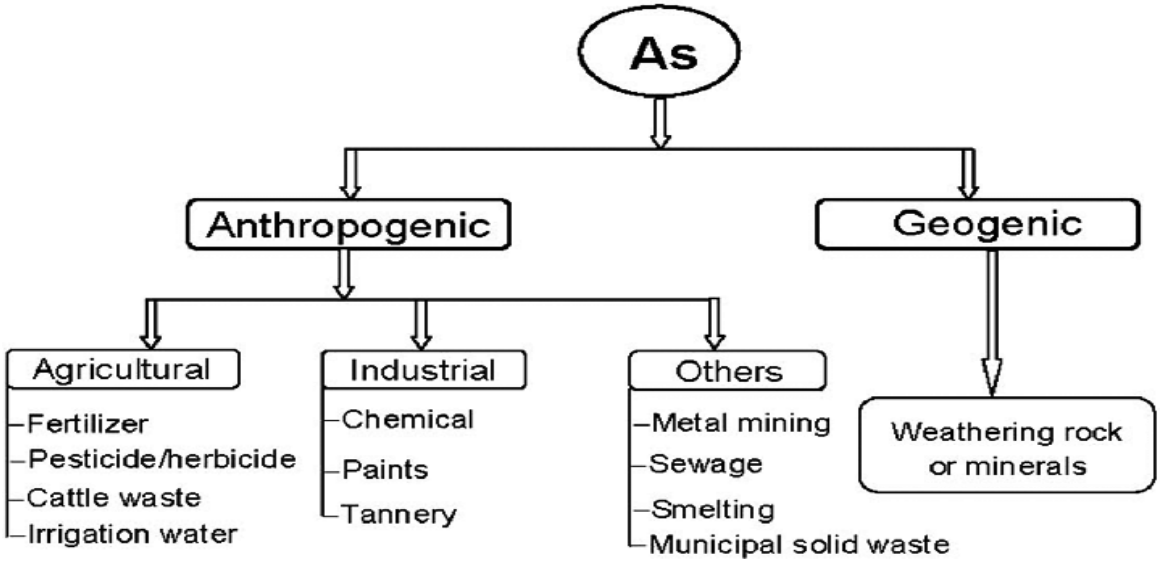


Fig. 1.5. The ways of As come into the environment (Sahoo & Kim, 2013).

1.5.2 Lead

Lead is a toxic metal at the lowest concentration and is naturally non–degradable in the environment. Exposure to lead occurs in various ways like leaded gasoline, industries such as lead smelting and coal combustion, lead–based paints, lead–containing pipes or lead–based solder in water supply systems, battery recycling, and grids (Fashola et al., 2016). The adverse health problem caused by Pb is lead poisoning and it is one of the most common and best–recognized childhood effects of the toxic element. Acute and chronic exposure are lethargy, vomiting, irritability, loss of appetite, dizziness, ataxia, and consciousness, which may lead to coma and death, weakness in fingers, wrists, ankles, and miscarriage for pregnant women (Salehipour et al., 2015).

1.5.3 Cadmium

Cadmium is a natural element and is widely distributed in the earth's crust at a concentration of 0.1–0.5 ppm, the high concentration can be found in an abandoned metal mine and various industries activities. The half–life of Cd entered into the human body is

26 years or over and the main source of Cd exposure is food intake (Ahn et al., 2017). Chronic Cd exposure damage kidney function and bones, renal failure, osteoporosis, and increased cancer risks (Hezbullah et al., 2016).

1.5.4 Chromium

Chromium is found naturally and widely distributed in soils and rocks. It enters the environment from natural and anthropogenic sources such as metal processing, tannery facilities, chromate production, stainless steel welding, and ferrochrome and chrome pigment production. It causes renal damage, allergy and asthma, and cancer of the respiratory tract in humans (Tchounwou et al., 2012).

1.5.5 Manganese

Manganese can be found abundantly in the environment in many types of rock and soil. It is ubiquitous in the environment and found in low levels in the water, air, soil, and food. The major anthropogenic sources of environmental Mn are Mn ore mining, Mn alloy production, welding, coke ovens, dry alkaline battery manufacturing, Mn salt production, traffic, and agriculture. The most common intake of Mn in humans is via inhalation or ingestion, which may result in central nervous system pathology, and irreversible brain disease, to some extent similar to Parkinson's disease (Cima, 2019).

1.5.6 Iron

Iron is abundant in the earth's crust. It is essential for the biological system in the hemoglobin synthesis and functioning of cells. The toxicity of iron is the liver, kidneys, and cardiovascular systems failure and a higher concentration causes acute toxicity in newborn babies and young children (Gautam et al., 2014).

1.5.7 Zinc

Zinc is an essential element and natural component in the earth's crust. Zinc toxicity in children is nausea and vomiting. Mining, zinc ores, coal burning, and their industrial

application are the major sources of zinc in the air, soil, and water. A higher concentration of zinc may cause anemia, pancreas damage, and cholesterol problems in human beings (Salehipour et al., 2015).

1.5.8 Copper

Copper is an essential trace element, and is important for metabolism. Copper can be found in rock and soil as a pure metal and in combination with others. Mining, metallurgy, and industrial applications are the major anthropogenic sources of copper in the environment. but an excessive amount of copper causes harmful effects on the human body such as nose, eyes, and mouth irritation, nausea, diarrhea, kidney damage, and even death (Gautam et al., 2014).

1.5.9 Nickle

Nickel can be formed from natural sources and human activities. The natural processes are volcanic eruption and weathering of rocks and soil in soils and volcanic rocks. Nickel and its compositions are used in several industrial applications such as electroplating, batteries, automobile and aircraft parts, batteries, coins, spark plugs, cosmetics, and stainless steel (Gautam et al., 2014). The health effects due to Ni exposure include fatal cardiac arrest, skin rashes, fatigue, headache, kidney heart problems, dizziness, respiratory illness, lung function disorder, and carcinogenic to humans (Salehipour et al., 2015).

1.5.10 Molybdenum

Molybdenum is found in minerals, rocks, and soil. Toxicity in humans is rare but acute toxicity is hallucination and seizure (Novotny & Peterson, 2018).

1.5.11 Cobalt

Cobalt can be found both natural and anthropogenic. The source of cobalt in the environment is the hard metal industry, electronic waste, paint, and construction. The

higher concentration of cobalt causes hematological and endocrine dysfunctions (Leyssens et al., 2017).

1.6 Country Background

Myanmar lies *22° 00′ N and 98° 00′ E*. The high–altitude northern part of Myanmar connects to the Tibetan Plateau, while the southern part has a low altitude and faces the sea. The Ayeyarwady River flows in the sedimentary basin's central zone and forms the Ayeyarwady Delta by the Quaternary sediment at the southern end. Some Cu–Au mines distribute in the northern half of the central zone, and tertiary intermediate to acidic volcanic rocks and Cenozoic limestones scatter the zone's center.

Myanmar is an agricultural country and rice (*Oryza sativa* L.) has been a major crop. The total cultivable area is almost 18.3 million ha. The total cultivated area in 2009 was around 12.1 million ha of which 11.0 million ha or 91 percent was for annual crops and 1.1 million ha or 9 percent for permanent crops. The cultivated areas are concentrated in the Ayeyarwady river basin, while the potential for further expansion lies mainly in upper Myanmar, in the Chin, Kachin, and Shan states (*Country profile – Myanmar*, FAO, 2011).

1.6.1 Description of the study area, Yangon Division, Ayeyarwady Delta

Ayeyarwady Delta is one of the most productive rice–growing areas in the world (Bhattacharya et al., 2010). It covers an area of 20570 km^2, of which 60% is used for rice cultivation. Ayeyarwady Delta is comprised of three divisions, namely Ayeyarwady, Yangon, and Bago (Besset et al., 2017). The location map of Ayeyarwady delta was shown in Fig.1.6 (USDA 2008). Rice production in Ayeyarwady Delta accounts for approximately 50% of the total country's production (Atsuda, 2009). Ayeyarwady River, which flows from the north covering 61% of the country's territory has five major tributaries in the delta and many distributaries to the Andaman Sea in the south (Brakenridge et al., 2017). The river flows from the eastern Himalaya ranges which are known as the source of arsenic origin, which caused the highest erosion into the Ayeyarwady River during the Late Cenozoic (Robinson et al., 2014). Annual rainfall

ranges across the Ayeyarwady delta areas is 2000–3000 mm. However, paddy irrigation with groundwater is quite rare in the Ayeyarwady Delta (20570 km^2), which accounts for half of the total rice production of Myanmar (Atsuda, 2009). The heavy monsoonal rains allow rainfed rice fields with the usual continuous flooding irrigation in the Ayeyarwady delta area, of which 60% is used for rice yield (Besset et al., 2017; Brakenridge et al., 2017). Rice is not cultivated in the dry season, and farm–pond water may be used for irrigation in Myanmar.

The five townships TK (Taik Kyi) and HTP (Htan Ta Pin), DL (Dala), KM (Kaw Mhu), and KCK (Kon Chan Kone), 25 sampling sites, are located in the Yangon division, an eastern part of the Ayeyarwady Delta (Fig 3.1). The altitudes of the northern two townships, TK (Taik Kyi) and HTP (Htan Ta Pin), are almost 11–18 m on the upstream side; The altitude of the southern three townships, DL (Dala), KM (Kaw Mhu), and KCK (Kon Chan Kone), is almost in 4–9 m on the downstream side. Since these five townships are the most agriculturally active areas and have no non–farm dwellings, the samples in this study can be regarded as the least affected by human activities. In these areas, the irrigation water source is the rainwater on the ground, including river water, but farm pond water is also available only in TK township Because the study area is a lower floodplain area, rice paddies in this region remain constantly flooded from sowing to earning. Rice harvesting is usually done after the water naturally recedes at the end of the rainy season.

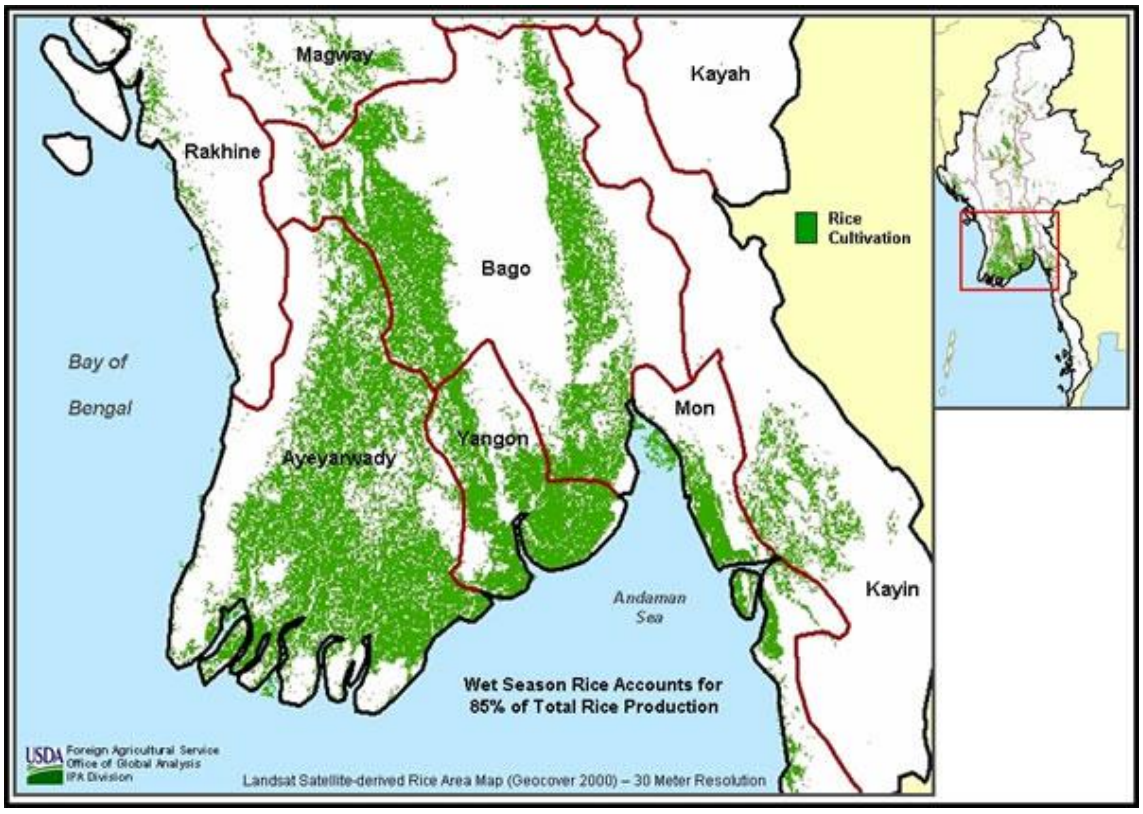


Fig. 1.6. The location map of Ayeyarwady Delta.

1.6.2 Heavy metal concentration in rice, water, and soil of Ayeyarwady Delta, Myanmar

Some domestic research conducted on the elemental concentration of rice in Myanmar. In one domestic study, the elemental concentration of polished rice and brown (unpolished) rice was done in rainfed paddy fields in Tharpaung Township, Ayeyarwady delta. Ten elements were determined by ICP–OES and Cd, As, Cr, and Co in white rice samples were below their MAC as in brown rice samples. In white rice samples, Pb, Zn, Cu, and Mn were above their respective MAC. However, these concentrations were lower than those of brown rice samples. The range of Ni, Zn, Cu, Mn and Fe concentration in white rice is 0.55–2.69 mg/kg, 50.53–158.3 mg/kg, 20.69–121.0 mg/kg, and 9.61–25.89 mg/kg 0.67–6.16±0.34 mg/kg. (Phyo Wai Zin et al., 2018).

Khin Phyu Phyu et al. (2017) studied As, Cd, Pb, Cr, Zn, Cu, Ni, Fe, and Mn in rice grains, their soil, and tube well water by ICP–OES in Kyon Pyaw Township, Ayeyarwady Delta. This study area uses rainwater for growing the paddy. Zn, Cu, Ni, and Pb in rice grains were above MAC value. As concentration was found higher in tube well water and the correlation between Fe and As was found in tube well water. The order of elemental concentrations in rice grain samples was Zn>Pb>Mn>Cu>Fe>Ni>Cd>Cr>As. Zn in rice grains ranged from 12.10±7.5 to 306.2±5.61 μg/g and Cu ranged from 3.102±1.63 to 253.0±1.48 μg/g. Ni ranged from 0.006±0.00 to 15.09±0.27 μg/g. Pb concentrations were from 0.11±0.01 to 6.898±0.02 μg/g.

Gianfranco. et al., (2020) researched heavy metal concentration in the groundwater of Maubin township which is a part of Ayeyarwady Delta. The trace element As, Cd, Co, Pb, and Zn were measured with ICP–MS. The highest As concentrations were examined in groundwaters from the Alluvial aquifer in Kyaunggon and Maubin townships. The arsenic speciation was analyzed and the results showed that 80% of the studied samples had As(III) which performed as the predominant species.

1.7 Problem Statement

Exposure to heavy metals in human beings via ingestion is a major concern in developing countries. Rice is the staple food in Myanmar and the average consumption of rice per person is about 496 g per day. Assessment of heavy metal concentration in the rice is important and the consumption of rice to be safe is a growing concern. Many researchers have focused on the heavy metal concentration of rice from irrigation water or anthropogenic activities, but they have less attention to report on heavy metal concentration of rainfed rice .

Ayeyarwady delta is an important agricultural province with most rice yield ranking first among all Myanmar's provinces. It plays a critical role in ensuring the national rice grain production safety of Myanmar. However, little data on rice has been conducted and the corresponding data on soil is rare. The potential health risk assessment on rice

consumption is a need and the present study has to assess the heavy metal concentration in rice of Ayeyarwady Delta and domestic market rice of Myanmar and to check the water and soil condition of the delta as a geological effect on the rice.

1.8 Main Objective

- To determine the health risk assessment of rice in study area
- To determine the concentration of rice in rainfed paddy fields due to the geological background

Chapter 2- Arsenic and heavy metal contents in white rice samples from rainfed paddy fields in Yangon Division, Myanmar-Natural background levels?

2.1 Introduction

Arsenic (As) is a well-known toxic element. Low-level arsenic poisoning symptoms include skin lesions, cardiovascular diseases, diabetes, and various organ cancers (Halder et al., 2020). The natural arsenic release mechanism to groundwater is a microbial reduction of arsenic bearing iron oxides in organic-rich sediment (Drahota et al., 2013), mainly in the shallow reducing aquifer at the early stage of arsenic contamination of groundwater (Andrew et al., 2008). The poisoning areas' characteristics are rapidly buried, Holocene sediments with low hydraulic gradients in deltaic areas with rivers flowing from the Himalayas ranges (Winkel et al., 2008), including Ayeyarwady River Delta in Myanmar (Fig 2.1). In the groundwater from below 20 m depth in the Ayeyarwady Delta area, elevated As concentrations (up to ca. 0.6 mg/L), higher than the WHO (World Health Organization) standard for drinking water (0.01 mg/L) (Herschy, 2012), were reported (Van Geen et al., 2014).

Rice (O*ryza sativa* L.) is the primary staple food in Southeast Asian countries. Paddy irrigation with the groundwater resulted in severe arsenic contamination in paddy soils and rice grains, inducing health concerns associated with rice consumption (Meharg, 2004). Arsenic-accumulated rice samples are reported from these countries, e.g., 0.49 mg/kg of average As concentration in rice from Gazipur, Western Bangladesh (Meharg & Rahman, 2003).

Recently, more serious pollution in rice grains was reviewed (Halder et al., 2020). However, paddy irrigation with groundwater is quite rare in the Ayeyarwady Delta (20570 km^2), which accounts for half of the total rice production in Myanmar (Atsuda, 2009). The heavy monsoonal rains allow rainfed rice (*Oryza sativa* L.) fields with the usual continuous flooding irrigation in the Ayeyarwady Delta area, of which 60% is used for rice yield (Besset et al., 2017). Therefore, the As concentration in the rainfed rice from the delta areas is expected to be a background level anterior to the groundwater irrigation.

Recently, it was reported that inorganic arsenic contents from markets in the Ayeyarwady Delta area (including the Yangon deltaic region) were 0.04–0.07 mg/kg (Nwe Ni Tun, 2019) below the maximum contamination level (Codex standards) (0.2 mg/kg) (FAO, 2006). The accumulation of heavy metal contents in rice samples has also received tremendous attention to the health impacts in Asian countries and was comprehensively reviewed (Bhattacharya et al., 2010).

Cadmium (Cd) is a toxic element having a high risk of accumulating in rice grain as As (e.g., (Zhao & Wang, 2020), which is the cause of an epidemic occurrence of Itai-Itai disease (1910–1970, Japan) due to mining and smelter activities. The benchmark dose as the threshold limit level of Cd concentration in rice for Itai-Itai disease development was estimated as 0.27 mg/kg (Nogawa et al., 2017). Cadmium is usually found in ores together with lead (Pb), copper (Cu), and Zinc (Zn). Nickel (Ni) is also a high-risk heavy metal accumulating in rice grain in the paddy soil derived from basalt rock (Wang et al., 2020), which is usually enriched in Ni, chromium (Cr) and cobalt (Co). These heavy metals (Cd, Pb, Cu, Zn, Ni, Cr, Co, Fe, and Mn) are widely used in industrial processes. The other symptoms of an overdose of heavy metals are Cd: damage to the kidney, liver, lungs, and the nervous system, cardiovascular diseases, development of reproductive toxicity, cancer, and mortality; Pb: renal dysfunction, damage to the central nervous system, and decrease of the intelligence quotients in children; Ni: genotoxicity, haematotoxicity, immunotoxicity, and carcinogenicity (Halder et al., 2020).

Although Yangon is the most industrialised city in Myanmar, little industrial activity has been found in paddy fields in the Ayeyarwady Delta area, far from any heavy metal mining activity in Myanmar (NATIONS, 1996). We can expect background levels of heavy metals and As in rice grain from the rainfed paddy field. However, we could find only a few data on heavy metals by ICP-OES analysis in the reports (Khin Phyu Phyu, 2017). So, we quantified As and these heavy metal contents in white (polished) rice samples (Yangon Rice, named in this paper) from the east end of Ayeyarwady Delta, five townships in Yangon Division (Fig 1A), Myanmar. The elemental content in white rice depends not only on the concentration in the soil but also on the growing conditions and the genotype of rice (Spanu et al., 2020). Assuming that the growing conditions and rice

varieties in the region will remain the same in the future, it is essential to know the concentration of harmful elements before human
activities to identify artificial contamination. By comparing the literature values worldwide, we will highlight the low level of heavy metal elements in rice in this study and consider the accumulation factors from previous reports.

2.1.1 Problem Statement

Exposure to heavy metals in human beings via ingestion is a major concern in developing countries. Rice is the staple food in Myanmar and the average consumption of rice per person is about 500 g per day. Assessment of heavy metal concentration in the rice is important and the consumption of rice to be safe is a growing concern.

Ayeyarwady delta is an important agricultural province which accounts for half of total rice production with most rice yield ranking first among all Myanmar's provinces. It plays a critical role in ensuring the national rice grain production safety of Myanmar.

2.1.2 Objective

- To confirm Yangon rice whether it is safe or not

2.2 Material and Method

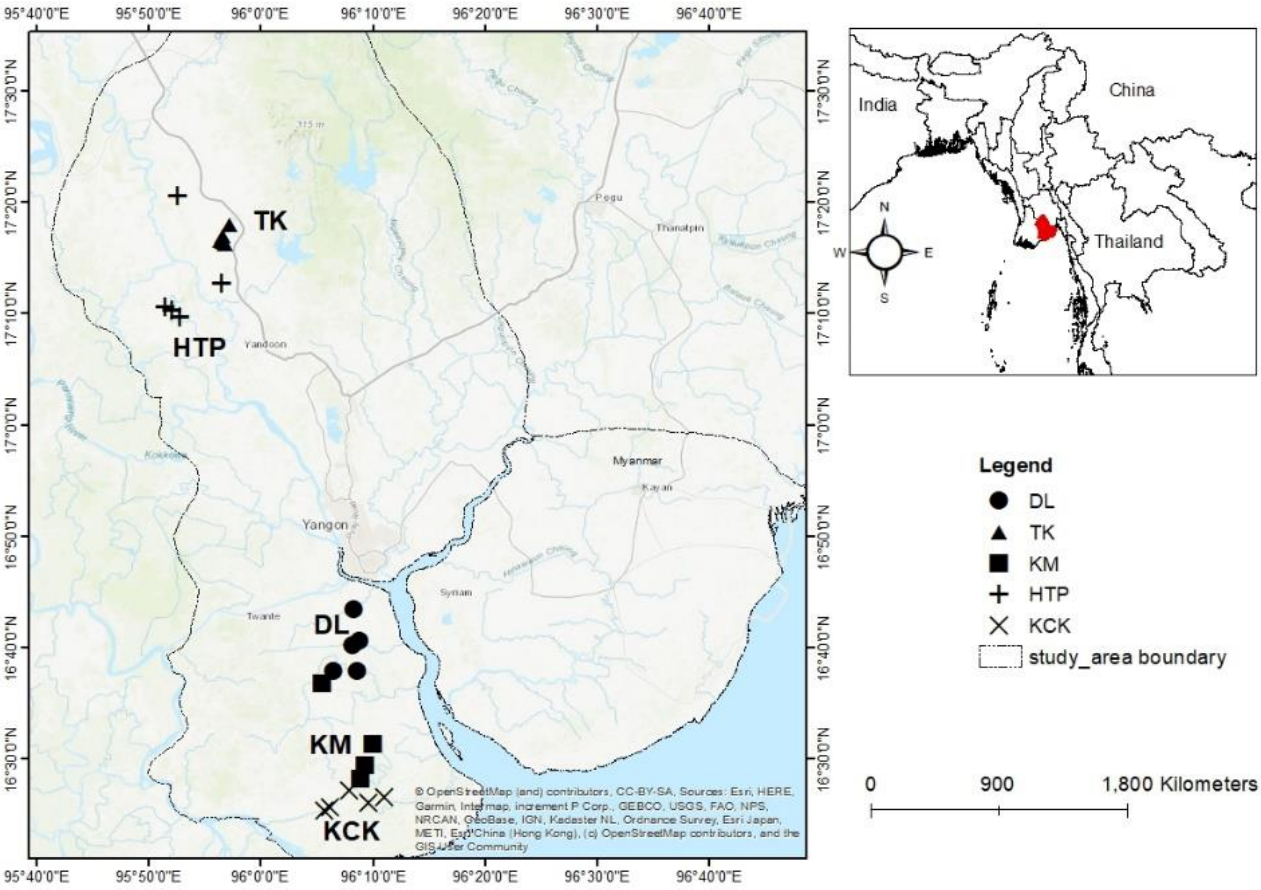


Fig. 2.1. Map of the study area and sampling points.

2.2.1 Sample collection

The first time sampling was conducted in April 2018. A total of 120 rice samples were collected from farmer houses in five different townships, TK (n = 24), HTP (n = 25), DL (n = 24), KM (n = 22), and KCK (n = 25). The latitude, longitude, and altitude data of the sampling point were obtained with the GPS tool. All the rice samples were sent for checking pests to Yangon, Myanmar Agriculture and Irrigation Minister under Plant Pest Quarantine Regulation to import to Japan for research purposes. Additionally, all the samples in this research were imported under the permit of the Minister of Agriculture, Forestry and Fisheries, Japan under the Plant Protection Law.

2.2.2 Sample preparation

The first time rice samples, April 2018 were polished rice and the second time samples December 2019 were field paddies and polished rice. The husks of the paddies were crushed and removed by rolling with the log. The first and second–time samples were treated on a clean bench and digested with certified reference materials (CRMs) and blank samples in runs. About 10 g of collected white (polished) rice grains were finely powdered by agate mortar, homogenized, and dried in a hot air oven (55°C for 36 h) until the weight became constant.

Fig. 2.2. Rice sample preparation at Toyoda laboratory.

2.2.3 Analytical Procedure

The powdered rice sample (30 mg) was weighed and placed in a 7 ml PFA vial (Savillex) and then mixed with 1mL of ultrapure–grade 69% (v/v) nitric acid and 0.1 mL of 30% (v/v) H_2O_2 (Kanto Chemical). After the digestion in the closed PFA vial in a hot air oven at 80°C for 12 hours, each sample solution was diluted into 30 mL with ultrapure water (milli–Q, Simplicity UV, and Gradient with Elix10, Millipore) from the cleanroom facility of the school.

Eleven elements in all sample solutions stocked in clean polypropylene bottles were determined with an Agilent 8800 triple quadrupole ICP–MS (ICP–QQQ) at the Open Facility of Hokkaido University. Arsenic was determined by using mass–shift mode for As^+ from m/z 75 to AsO^+ at m/z 91 to eliminate the mass interference by $^{150}Nd^{2+}$ and $^{150}Sm^{2+}$ (Jackson & Punshon, 2015); and the other elements (Pb, Cd, Cr, Mn, Fe, Zn, Cu, Ni, Mo, and Co) were with helium collision mode. The sweep times were 50–100 and

integration times were 3.0 x 3 sec for As, Pb, Cd, Cr, and Co; 0.1 x 3 sec for Mn and Fe; 0.3 x 3 sec for Zn, Cu, Ni, and Mo. All working and running standard solutions (0, 5, 20, 50, and 100 µg/L) were prepared by serial dilution of ICP multi–element standard solution H (Kanto Chemical) with ultrapure water. The result of raw data was ppb and the calculation was carried out based on the weight and solution of the sample in terms of mg/kg.

2.2.4 Method of validation

Data precision and accuracy were checked by the analysis of certified reference materials: NMIJ CRM 7501–a, 7502–a, from the National Metrology Institute of Japan for the rice. The reference samples were put in each batch of digestion rice and soil. After analysis, the recovery rates were checked and the results were compared with the certified values for the respective standard sample. The blank samples were set for checking the contamination of the digestion.

2.2.5 Health Risk Assessment of polished rice

2.2.5.1 Estimated Daily Intake

The health risk assessment method is established by the Joint FAO (Food and Agriculture Organization)/WHO (World Health Organization, 2007) and Expert Committee on Food Additives, and the United States Environmental Protection Agency (USEPA, 2022). The average daily intake (mg/kg/day) of heavy metals through ingestion of rice grains can be estimated as follows:

$$EDI\ (\text{Estimated daily intake}) = Cm \times IR/BW$$

where Cm (mg/kg) is the mean heavy metal concentration in grains; IR (g/day) is the ingestion rate of rice; BW (kg) is the average body weight. Incidentally, the daily rice consumption in Myanmar was reported to be 578 g/day before 2000 (Genchi, 2020), 512 g/day in 2014, and 487 g/day in 2017 [https://www.helgilibrary.com › indicators › myanmar]; we set IR at 500 g/day as the convenient value to consider in this study. We put 58 kg of average body weight into BW since the mean body weights between 18 and

25 years of age are 61.1 kg for males and 54.7 kg for females in Myanmar [https://www.worlddata.info/average–bodyheight.php].

2.2.5.2 The hazard index of non–carcinogenic risk (HI)

The hazard index (HI) of non–carcinogenic risk by rice consumption is estimated as the sum of each toxic element's non–carcinogenic hazard quotient (HQ) values. So, the HI (hazard index) is equivalent to the total HQ (THQ). The HQ values are calculated as follows:

$$HI = THQ = \Sigma HQ = \Sigma (EDI/RfD)$$

where RfD (mg/kg/day) is the oral reference daily dose of each heavy metal. Chronic oral RfDs should be used to evaluate the potential non–carcinogenic effects associated with exposure periods greater than seven years (USEPA,2022) "HI (THQ) > 1" indicates that the level of exposure is likely to cause non–carcinogenic health risks to an individual, whereas chronic risks are assumed to unlikely happen in case HI < 1 (USEPA,2005). RfD (Oral Reference Dose) values of 0.0003, 0.003, 0.001, 0.003, 0.14, 0.7, 0.3, 0.04, 0.011, 0.005, and 0.0003 for inorganic As, Pb (phosphate), Cd, Cr(VI), Mn, Fe, Zn, Cu, Ni (hydroxide), Mo and Co, respectively (Adimalla N. et al., 2020).

2.2.5.3 Total cancer risk (TCR)

Total cancer risk (TCR) is the sum of cancer risk (CR) values for each carcinogenic metal(loid) (As, Cd, Cr, Ni, and Pb). The CR was calculated by multiplying the EDI (mg/kg/day) over a lifetime with a slope factor (SF) for an incremental risk of individual developing cancer, using the equation provided in USEPA Region III Risk–Based Concentration Table (USEPA, 2006).

$$TCR = \Sigma CR = \Sigma (ADI \times SF \times 0.371) = \Sigma \{ADI \times SF \times (EF \times ED/AT)\}$$

where EF is the exposure frequency (365 days/year), ED is the exposure duration (26 years) (USEPA, 2022), and AT is the average time for carcinogens (365 days/year × 70 years). The child exposure duration is limited to 6 years, and the adult exposure is assumed to be the entire 26 years (USEPA, 2022), so the results of this study are for middle–aged and older adults. The TCR indicates the cumulative probability of an individual developing cancer over time. For the oral slope factors (SFs) the values are 1.5, 0.038, 15.0, 0.50, and 0.91 for inorganic As, Pb (phosphate), Cd, Cr (VI), and Ni (hydroxide), respectively.

2.3 Results and Discussions

2.3.1 The geological data of the sampling sites

A total of 120 white rice grains are collected from the five townships (TK, HTP, DL, KM, and KCK). The geological data of the sampling townships are shown in table 2.1.

Table 2.1. The geological data of sampling sites.

Township name	irrigation method	Village Code	Sample No.	Altitude (m)	Latitude	Longitude
Taik Kyi (**TK**)	Rainfed Irrigation	TK 1	1–5	15.5	17°18'01.86"N	95°57'15.14"E
		TK 2	6–10	12.5	17°16'32.61"N	95°56'35.42"E
		TK 3	11–15	11.0	17°16'15.39"N	95°56'48.77"E
		TK 4	16–19	18.0	17°17'02.88"N	95°56'38.22"E
		TK 5	21–24	16.0	17°16'36.62"N	95°56'23.53"E
Htan Ta Pin (**HTP**)	Rainfed	HTP 1	1–5	11.0	17°20'40.43"N	95°52'32.58"E
		HTP 2	6–10	6.0	17°12'48.46"N	95°56'32.52"E
		HTP 3	11–15	13.0	17° 09'45.19"N	95°52'47.88"E
		HTP 4	16–20	12.0	17°10'19.98"N	95°52'07.94"E
		HTP 5	21–25	13.0	17°10'38.33"N	95°51'29.49"E
Dala (**DL**)	Rainfed	DL 1	1–5	6.0	16°37'52.99"N	96° 06'32.05"E
		DL 2	6–10	8.0	16°40'34.32"N	96° 08'50.59"E
		DL 3	11–15	5.0	16°40'13.90"N	96° 08'11.85"E
		DL 4	16–20	5.5	16°37'51.42"N	96° 08'40.26"E
		DL 5	21–24	4.0	16°43'22.36"N	96° 08'16.85"E
Kaw Mhu (**KM**)	Rainfed	KM 1	1–5	7.0	16°37'07.40"N	96° 05'23.87"E
		KM 2	6–10	4.0	16°31'15.92"N	96°10'0.62"E
		KM 3	11–15	6.0	16°28'12.83"N	96° 08'53.47"E
		KM 4	16–20	5.0	16°36'43.86"N	96° 05'28.71"E
		KM 5	21–25	6.0	16°29'24.56"N	96° 09'20.10"E
Kon Chan Kone (**KCK**)	Rainfed	KCK1	1–5	9.0	16°25'40.10"N	96° 06'13.33"E
		KCK 2	6–10	11.0	16°25'23.45"N	96° 05'40.40"E
		KCK 3	11–15	6.0	16°26'06.24"N	96° 09'38.92"E
		KCK 4	16–20	3.0	16°27'10.35"N	96° 07'57.74"E
		KCK 5	21, 22	9.0	16°26'37.35"N	96°11'5.41"E

2.3.2 Accuracy Analysis

The quality of analytical procedures checked by the analysis of certified reference materials were shown in Table 2.2. The results of the standard reference material for rice and the results were consistent.

Table 2.2: Analytical result of heavy metals in two certified reference materials (mg/kg)

	NMIJ CRM 7502–a (n = 18)					
mg/kg	Results	±	SD	literature (2)		
As	0.12	±	0.01	0.109	±	0.005
Pb	0.003	±	0.003	0.0043	±	0.0006
Cd	0.41	±	0.20	0.548	±	0.02
Mn	10.18	±	1.92	11.2	±	0.4
Fe	4.15	±	0.98	4.48	±	0.2
Zn	23.73	±	3.17	26	±	0.9
Cu	2.90	±	0.34	3.02	±	0.11
Ni	0.34	±	0.09	0.39	±	0.022

	NMIJ CRM 7501–a (n = 43)					
mg/kg	Results	±	SD	literature (1)		
Cd	0.05	±	0.003	0.05	±	0.0024
Mn	6.78	±	0.37	6.75	±	0.26
Fe	3.53	±	0.46	4.04	±	0.24
Zn	19.89	±	1.01	20.1	±	0.7
Cu	2.47	±	0.18	2.49	±	0.09

NMIJ CRM 7502–a, Trace elements in White Rice Flour (Cd level II), 2015

NMIJ CRM 7501–a, Trace elements in White Rice Flour (Cd level I), 2015

2.3.3 Heavy metal concentration in white or polished rice

The concentration of eleven elements As, Pb, Cd, Mn, Fe, Zn, Cu, Ni, Mo, and Co were analyzed in 120 rice samples. Fig. (2.3 A–C) showed a box plot of each element concentration in rice samples from five townships. The maximum allowable limits of 8 elements iAs (inorganic As), Pb, Cd, Cr, Zn, Cu, and Ni) were from the FAO/WHO and USEPA guidelines described as a dotted line. The horizontal line in a shaded box enclosed 50% of the data, indicating the median value. The box's top and bottom edges (UQ and LQ) were marked 75 and 25 percentiles, and the difference between UQ and LQ was the interquartile distance (IQD). The lines extending from each box's edges indicated the maximum and minimum values between "UQ + 1.5 x IQD" and "LQ – 1.5 x IQD" values. Any outlier value outside the range was displaced as an individual point.

The mean and medium values of Pb, Cd, Cr, and Cu in 120 white rice grains were one order of magnitude lower than the corresponding MAC values. The upper quartile values of As and Ni contents in our samples were equivalent to the MAC values.

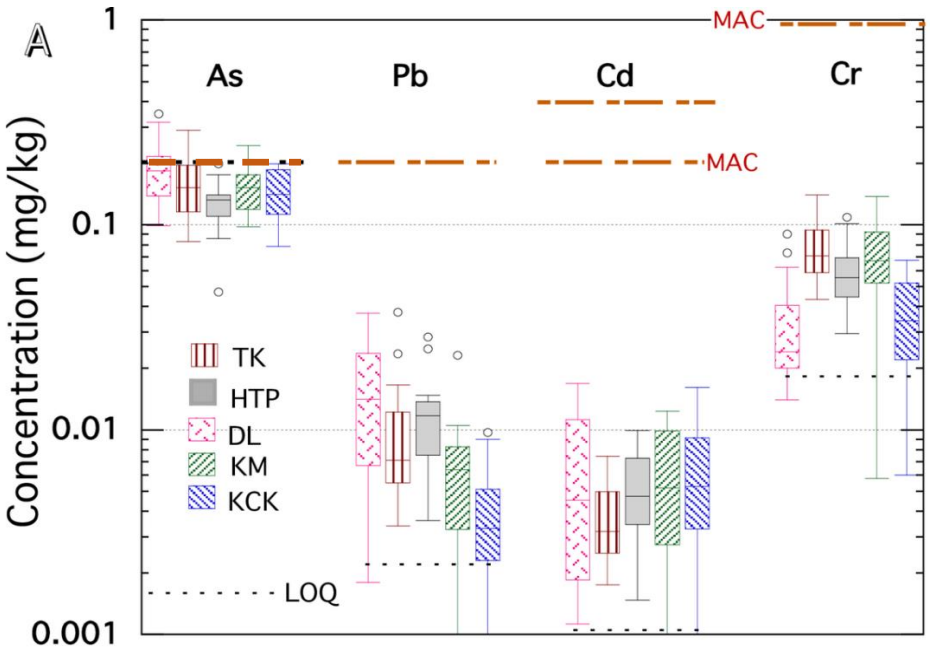
The highest median and the maximum value in As concentration of rice grains were observed in DL township (0.183, 0.348 mg/kg). The medians and highest As concentrations in TK and KM townships were 0.152, 0.290, and 0.152, 0.244 mg/kg. KCK and HTP were under the limit with medians of 0.141 and 0.133 mg/kg. The mean total-As content in 120 samples was 0.155 ± 0.052 µg/g, and the WHO proposed MAC of inorganic As (*i*As) at 0.2 µg/g for white rice (Fig. 2.3 (A)).

The inorganic As species (iAs) are considered to be fairly more toxic than methylated As species (Cullen and Reimer, 1989). iAs is a non-threshold Class 1 carcinogen (Smith et al., 2002). In this study, the proportion of inorganic As (*i*As%) in Myanmar white rice was not analyzed. Williams et al. (2005) and Nookabkaew et al. (2013) reported that *i*As% was about 80% and 63% for the white rice grains (n = 27 and 79) produced in Bangladesh & India and Thailand, respectively. Meharg et al. (2009) reported that 0.15 µg/g was the mean total-As content in white rice grains (n = 901) from global ten countries. Most of the As concentrations

in this study are below the MAC value and are close to the average rice concentration globally. Assuming that *i*As% was 100% for the white rice grains in this study, the mean As concentration was about two-thirds of the MAC value.

The range of the Ni in the rice was 0.168–0.935 mg/kg for DL, 0.12–1.06 mg/kg for KM, 0.18–0.89 mg/kg for TK, 0.174–1.66 mg/kg for HTP and 0.137–0.729 mg/kg for KCK. In contrast to the MAC values in white rice (1.0 mg/kg of Ni), this study's average of rice grains was 0.39 ± 0.19 mg/kg and lower than the MAC Fig. 2.3 (B). The graph shows that both Cu and Mo contents in TK rice grains are significantly lower than those from the other townships. TK township among the five, irrigation by farm pond water, was carried out for rice cultivation. The difference in irrigation source might contribute to the difference in Cu and Mo contents in Fig. 2.3 (B).

Fig. 2.3 (C) demonstrates that Fe levels in rice grains from the HTP and DL townships are significantly higher than those in the other three townships. The mean and range of Fe in HTP was 23.8 (3.38–130.9) mg/kg and in DL was 13.8 (4.91–35.8) mg/kg. It may translate that paddy soil in the HTP and DL townships is more enriched in Fe or acidic than the other three townships. Iron is primarily mobilized chiefly as dissolved Fe(II) under reducing conditions, and it is redistributed as particulate Fe(III) oxyhydroxides in oxygenated environments (Langmuir, 1997). The soil in the HTP and DL townships is richer in iron oxide components than the other township. Zn concentration in all areas was lower than MAC and the level of metals in the rice decreased in the following order: Zn > Fe > Mn > Cu > Ni > Mo > As > Cr > Co > Pb > Cd.


A
1
0.1
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Concentration (mg/kg)
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Pb
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MAC
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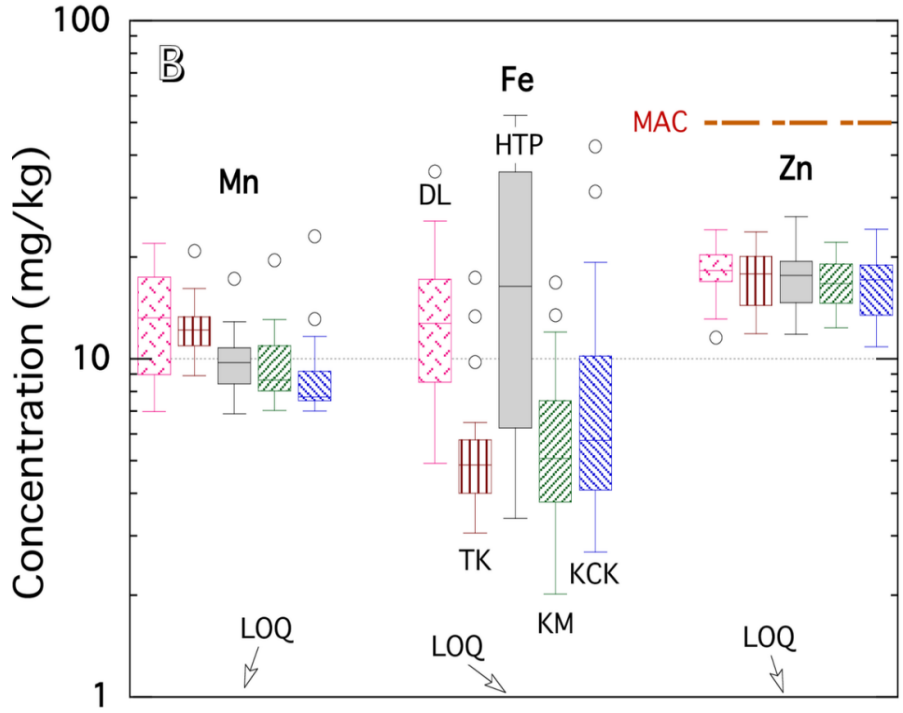
KM
KCK
LOQ


B
100
10
1
Concentration (mg/kg)
Mn
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MAC
HTP
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LOQ

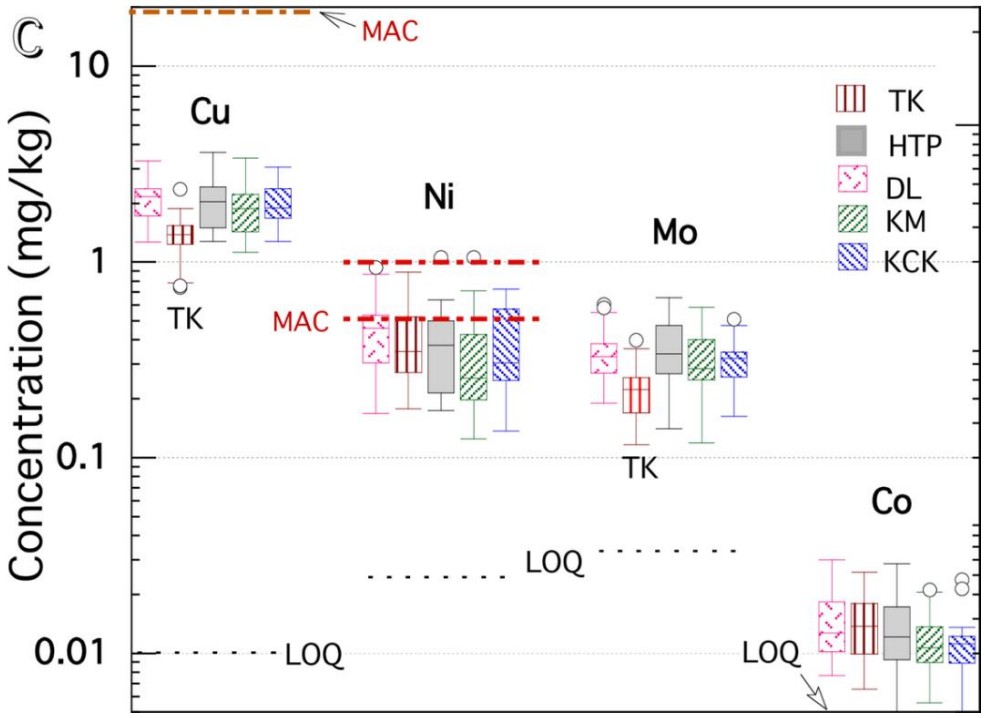


Fig. (2.3 A–C) Box plot of each element concentration in rice samples from five townships.

2.3.4 Elemental correlations with arsenic contents

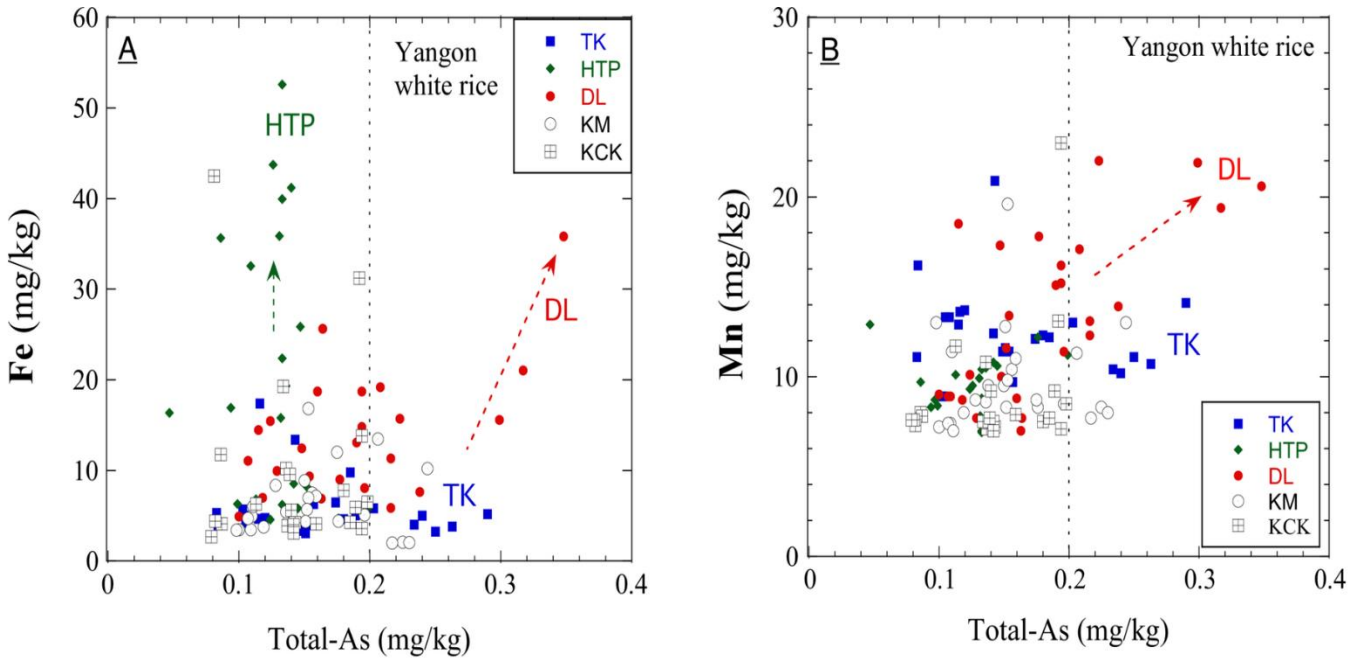
Figs 2.4 A–E show the correlation between As content and Fe, Mn, Cu, Zn, and Cd in this study's grains. Fig A suggests that the upstream HTP and downstream DL townships' paddy soils contain non-As-bearing and As-bearing iron oxide, respectively. Fig B implies that the paddy soils from DL have high Mn contents. It is well known that arsenic-polluted groundwater usually has a high Mn and Fe concentration as a worldwide phenomenon (e.g. (Buschmann et al., 2007)). For example, it was reported that a large amount of arsenic released from Holocene sediments, derived from the Himalaya ranges, to groundwater at a shallow valley was associated with a more significant elevation of manganese (Mn) levels in Cambodia (Nookabkaew et al., 2013). Among the five sampling sites, the altitudes of the upstream sides (TK, HTP) and downstream sides (DL, KM, and

KCK) are average about 12 m and 6 m. The northmost DL site among downstream sides may be where arsenic rich sedimentary layers eroded upstream could be carried and quickly deposited topographically during floods (Fig 2.1). Therefore, the migration of Himalaya-derived As bearing iron oxide particles may explain the high As content in some DL rice grain samples.

Figure C shows that the low Cu contents in TK rice (Fig 2.3 C) might be associated with the high As contents likewise Fe contents in the rice. It was reported that the suppression effect of Cu on As accumulation in white rice (J. Liu et al., 2019a). Iron oxide plaque around rice roots is a strong adsorbent for As (V) and acts as a barrier to As uptake into the root cells (W. J. Liu et al., 2006). It is plausible that the top-layer pond water for irrigation is depleted in bio essential elements, Fe, Cu and Mo, relative to flood water containing suspended matter more abundantly because these elements easily incorporate sinking particles, including insoluble hydroxides and microorganisms in water. Therefore, we can infer that pond water irrigation contributes to the Cu and Fe-depleted paddy soils and high As contents in rice grains at TK township. However, it remains to be confirmed and clarified by the soil and water analysis in future.

Figure D shows a positive correlation ($r = 0.47$) between Zn and As in the white grains in this study. Previous studies have also shown, for example, positive correlations of Zn-As ($r = 0.24$) and of Cu-Zn-Cd in rice from an arsenic-unpolluted area (rice grain As 0.11 ± 0.04 mg/kg) in China were demonstrated (Tang et al., 2021). The heavy metal concentrations explain these positive correlations in soils, usually depending on Mn oxides because manganese oxides adsorb these heavy metals with high capacities (Tang et al., 2021) and iron oxides (W. J. Liu et al., 2006). The redox potential of paddy soil significantly affects the bioavailability of As and Cd. It has profound opposite effects on the concentration of As and Cd in rice grains (Zhao & Wang, 2020). A weak negative As-Cd correlation ($r = –0.27$) was found across all five regions (Fig. E). Also, significant negative correlations in township samples except DL were found (TK: –0.54; HTP: –0.60; KCK: –0.63; KM: –0.31; DL:

–0.14). The non-significant negative correlation for DL township may be related to the significant positive correlation between iron and arsenic in samples only from DL township (DL: 0.55; TK: –0.18; HTP: 0.00; KCK: –0.09; KM: –0.13; S3). The presence of arsenic-containing iron components in the soil only in DL areas may influence the behaviour of arsenic in the soil more strongly than pH or redox potential in paddy soil. Generally, there are significant differences in the bioaccumulation of toxic elements in rice grains by genotype (Spanu et al., 2020).



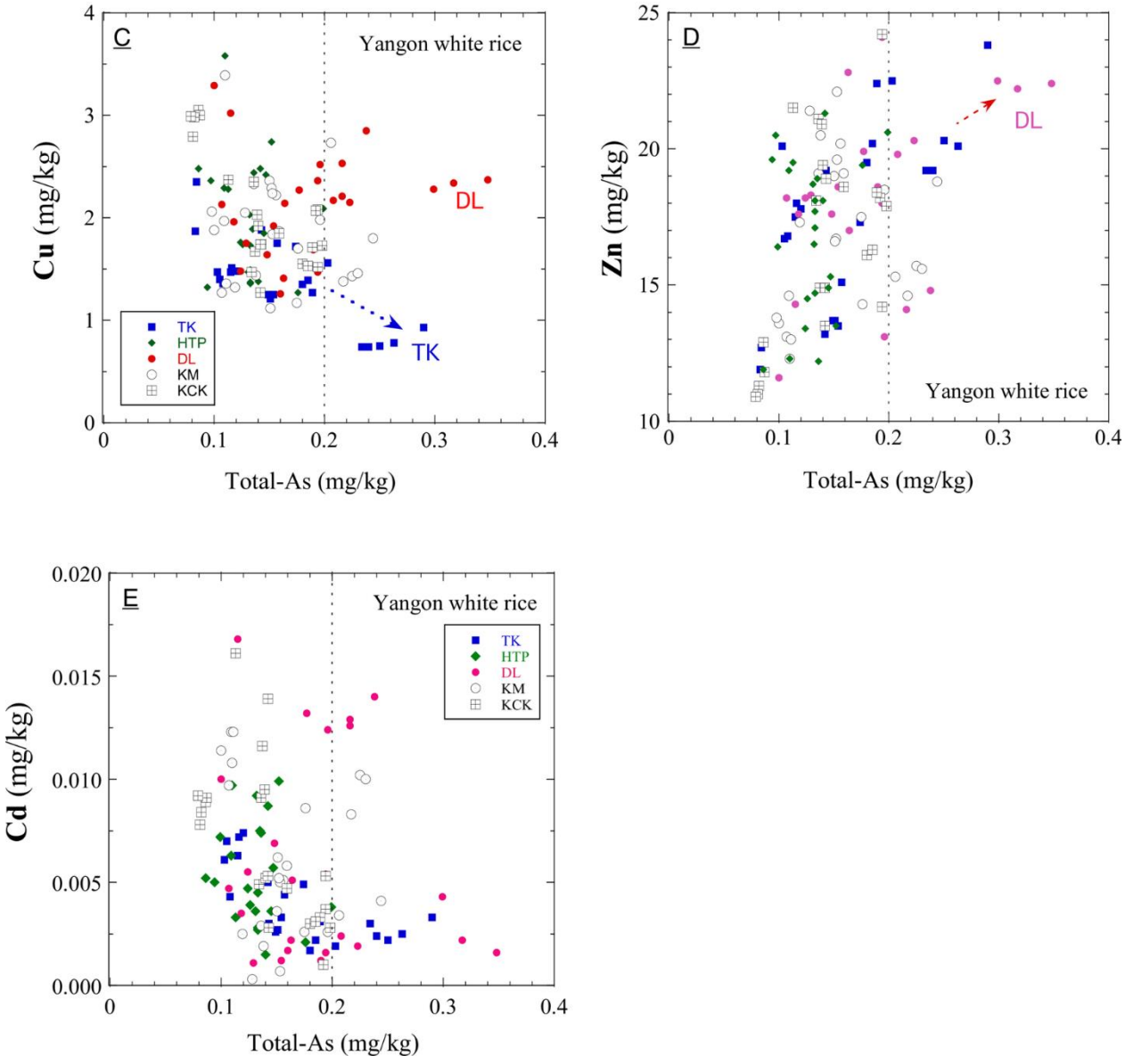


Fig 2.4 (A-E). Elemental correlation with total-As contents in white rice grain. A) Fe; B) Mn; C) Cu; D) Zn; E) Cd concentrations in the white rice grains.

2.3.5 Cadmium contents in rice grains from the world

Fig 2.5 is a scatter plot of Cd-Pb concentrations in this study compared to the reported mean concentrations in white rice grains from various 362 countries and regions (The data sources are described in the figure caption). In this study, the Cd concentrations in white rice grains were 5.6 ± 3.7 μg/kg (n = 115). It is comparable to the lowest concentration in the literature values (Fig 2.3.5). Previous papers reported that the white rice from Cambodia has the lowest mean Cd contents (6 μg/kg: n = 14) among the ones from 12 countries (Meharg et al., 2013); Egypt rice has 4 ± 1 μg/kg (n = 5), the lowest concentration among eight countries (Dittmar et al., 2010a); the mean Cd concentration in white rice from four areas in Thailand and rice from Saraburi provinces has 6 ± 4 μg/kg (n = 5) Cd concentration (Kukusamude et al., 2021); Malawian rice's mean Cd content is 9 μg/kg (n = 21) (Joy EJM et al., 2017). Therefore, Yangon rice is one of the rice grains with the lowest Cd content globally.

The Cd concentrations in rice grain vary by three orders of magnitude, depending on rice genotype and growing conditions (pH and redox condition) associated with the bioavailability of Cd in soil (Zhao & Wang, 2020). The median Cd transfer ratio from soil to grain increases by two and a half orders of magnitude when the soil pH value drops from 8 to 5 (Zhu et al., 2016). Volcanic acidic soil is usual in Japan, a highly volcanic country with 110 active volcanoes. It can be one of the reasons that the Cd average content in white rice collected in 1998–2000 in Japan is 50 μg/kg (n = 1198; 65 sites; (Shimbo et al., 2001)), an order of magnitude higher than the ones in samples of this study (Fig 2.3.5). In contrast, the Saraburi province (rice Cd contents: 6 ± 4 μg/kg) is a sedimentary carbonate area in Thailand. The paddy soils of Yangon rice may not be calcareous. However, they are certainly not acidic due to Myanmar's geology: no volcanic activity and several limestone mines are found in the middle stream of the Ayeyawady River (Friedrich Bender, 1983).

The redox potential of flooded paddy soil is usually held in reducing conditions (– 200 mV vs SCE; (Spanu et al., 2020)). Although the soil pH was slightly acid to neutral (6.4 ±|0.8) in the study area (Than AA, 2008), the seasonal changes in surface water pH during the rice-

growing season were in the range of 7.0–8.0 in the upstream area (Oo Az , et al., 2015). The Eh-pH diagram of Cd indicates that most Cd exists in the form of CdS, which has low solubility around neutral pH (log Ksp = –14.4; (Daskalokis KD, 1992). Since the study area in the Ayeyawady delta is annually exposed to flooding during the heavy monsoonal rainy season, the soil is under reducing conditions. The irrigation system should contribute to the rice grains' low Cd concentration.

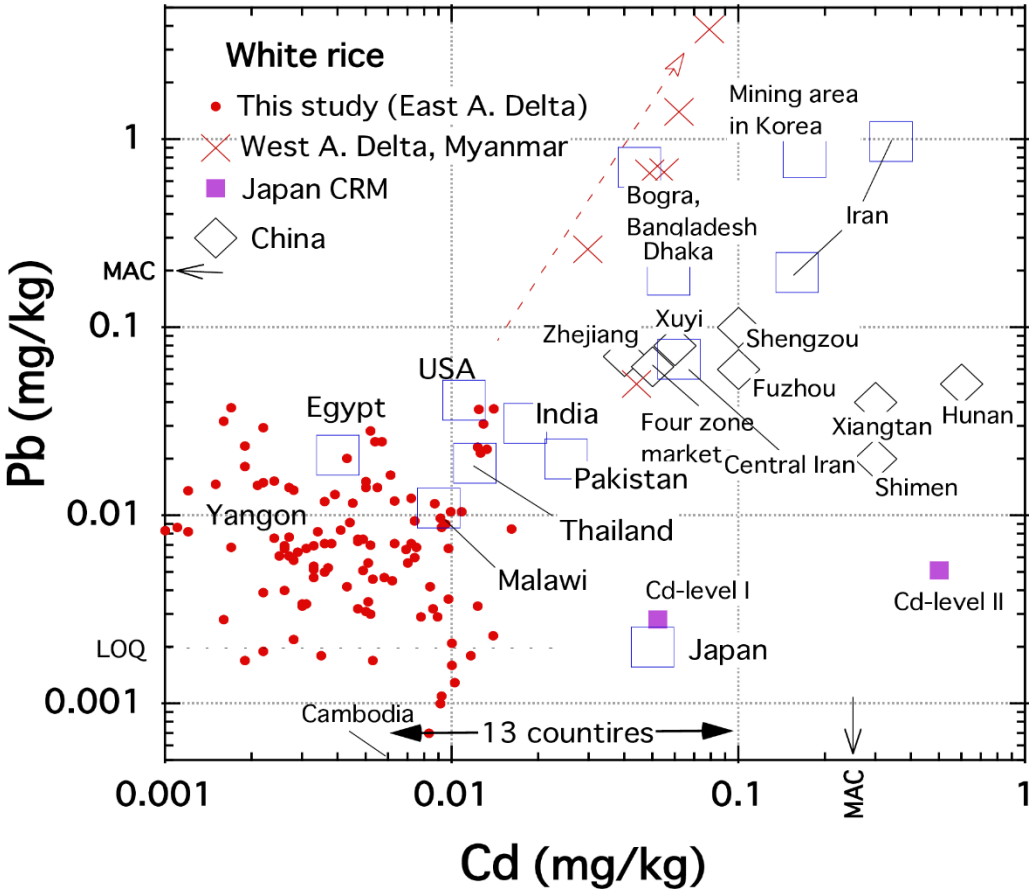


Fig 2.5. Correlation diagram between Pb and Cd concentrations in white rice from different countries and areas. Japanese certified reference materials (CRM) data (NISt, 2005) are from Table 2.2. The mean values of previous reports: West Ayeyawady delta in Myanmar (Khin Phyu Phyu et al., 2017) (Phyo wai Zin, 2018); Xuyi, China (Wang et al., 2020); Shengzou, Shimen, Fenghuang, and Xiangtan, in China (Zhao K et al., 2009) (Fu J et al., 2008),; Bangladesh (Islam et al., 2014) (Islam et al., 2015); Iran (Naseri et al., 2015)(Jafari et al.,

2018). The Cd range of 13 countries (Cambodia, France, Ghana, Italy, India, Japan, Nepal, Spain, SriLanka, Thailand, USA, Bangladesh, Jamaica) (Meharg et al., 2013)(Ahmad et al., 2018); China (A. Lu et al., 2018); Japan (Shimbo et al., 2001); Korea (Kwon et al., 2017); Malawi (Joy EJM et al., 2017); Central Iran (Shakerian A et al., 2012); Egypt, India, Pakistan, Thailand and USA (Dittmar et al., 2010b).

2.3.6 Lead contents in rice grains from the world

This study showed that the Pb mean concentration in white rice grains was 10 μg/kg (n = 115). It corresponds to the second-lowest concentration after Japan (1.8–3.0 μg/kg; n = 1198; 65 sites; (Shimbo et al., 2001)). It was reported that the mean Pb content of Malawian 20 rice was 11 μg/kg (n = 21) (Joy EJM et al., 2017); 17–20 μg/kg (n = 409) was the Pb median concentration in rice grains from Thailand, Pakistan, India and Egypt (Dittmar et al., 2010b). So, Yangon rice is one of the rice samples with the lowest Pb contents (Fig 2.5). The lead (Pb) has a lower solubility than cadmium in the paddy soil of the delta. The Eh-pH diagram of Pb (Brookins 1988) suggests that most Pb exist in the form of PbS or $PbCO_3$, which have low solubilities (log Ksp = –27.9, –13.1, respectively; (Charlot G. 2007) in the condition (pH = around 7; –200 mV vs SCE；(Spanu et al., 2020). The hydrous ferric oxide, formed at the soil surface under the floodwater, is known to have excellent adsorption properties against Pb ions and other toxic ions (e.g., (Waychunas et al., 2005)). Investment of phosphate-based fertiliser into paddy soils promotes the formation of stable lead phosphate mineral pyromorphite [$Pb_5(PO_4)_3$(OH Cl)] (Ruby et al., 1994), which has very low solubility (log Ksp = –79) (Flis et al., 2011). Myanmar's agricultural policy recommends fertilisation (commonly, P 30 kg/ha) (Oo Az et al., 2015). The associated high phosphate levels in paddy soils may be one of the reasons for the low Pb levels in Yangon rice. The extractable P level in nearby paddy fields was 0.3–195.7 mg/kg (Oo Az et al., 2015). One of the reasons for the low Pb content of Yangon rice might be the wind direction during the rice growth period. This study's two sampling sites are 2 km north-northwest of Yangon urban, and the other three are within 2 km just south of the area. South-West wind prevails during this area’s rainy

season (May–October). Hence, we assumed that rice plants in the sites grew without exposure to the Yangon urban area's aeolian dust, 5.2% of roadway emissions (Sricharoenvech et al., 2020). Many countries have reported high Pb contents in roadside soils (Walraven et al., 2014). Therefore, the weather may allow this study's grain samples to have little traffic-related lead pollution, although the sites are near the metropolitan.

2.3.7 Nickel and Chromium contents in rice grains from the world

Fig 2.6. shows the Ni-Cr plot of concentrations of previous reports and this study. It is plausible that the associated paddy soil concentrations primarily determine the Ni and Cr contents in rice grains. In contrast to the MAC values in white rice (1.0 mg/kg of Ni and 0.2 mg/kg of Cr), 2–3 mg/kg of Ni and 1–6 mg/kg of Cr contents are found in the rice grain grown on serpentine soils, the weathered product of peridotite host rock (ultramafic rock) in Philippines (a small patterned square in Fig 5) (Infante et al., 2021). The rice grown on the basaltic soil in areas of low human activity in Xuyi County, China, has 1.22–9.34 mg/kg of Ni and 0.24–0.91 mg/kg of Cr as the range of contents (a sizeable patterned rectangle with rounded corners in Fig 2.3.7) (Wang et al., 2020). The Ni, Cr and Co concentrations in Yangon rice grains were 0.39 ± 0.19 mg/kg, 0.056 ± 0.029 mg/kg and 0.013 ± 0.005 mg/kg (n = 118–119), which are as low as the literature values of white rice from Japan and Thailand, where the distribution area of mafic rock is few (https://ccop445 gsi.org/main). Felsic rocks distribute widely in Japan along the island arc volcanic zone. The low Ni and Cr content of Yangon rice is presumably due to the low concentration of Ni and Cr in the paddy soils, as there are no mafic rocks in the vicinity of the sampling points.

Figure 2.6. suggests that white rice samples from two Chinese locations, Iran, India,and Bangladesh (Jafari et al., 2018), project towards a Cr enrichment component, which is different from the trend of the dashed line (felsic–mafic rock area products: Cr/Ni = ca. 0.1) by the rock type difference connecting literature values for samples from areas with low human activity.These paddy fields (Xiangtan, Shengzhou, Fatehabad, Dhaka, and Bogra) are out of basaltic rock areas (https://ccop-gsi.org/main). The Cr enrichment in rice might be due

to Cr pollution by industrial activities such as metal plating, wood preservation, ink manufacture, dyes, pigments, glass and ceramics, tanning and textile industries, and corrosion inhibitors in cooling water (Dhal et al., 2013).

On the other hand, industrial Ni soil contamination seems insignificant compared to mining and refining nickel mines activities. However, our daily lives are full of nickel alloy parts, and nickel intake can cause various effects on human health (Genchi et al., 2020). The livelihoods of farmers who cultivate this paddy field area may be affected. The correlation tables between ten elemental abundances in sample grains by townships (S3 tables) reveal that Ni contents have high positive correlations with both Cu and Cd contents (Cu: r =0.70–0.88; Cd: r = 0.59–0.73, except KM township). Slightly higher correlation coefficients (Cu: r = 0.83, 0.84; Cd: r = 0.73, 0.69) were found in the groups with higher Fe contents at HTP and DL townships, indicating these three heavy metals may have an adsorbed form on iron hydroxide in the paddy soils. If human activity's influence contributed to these positive correlations, e.g., industrial waste disposal, rice grains' Pb and Cd concentrations should not be at the lowest level. Therefore, Ni contents in this study were close to the natural background level.

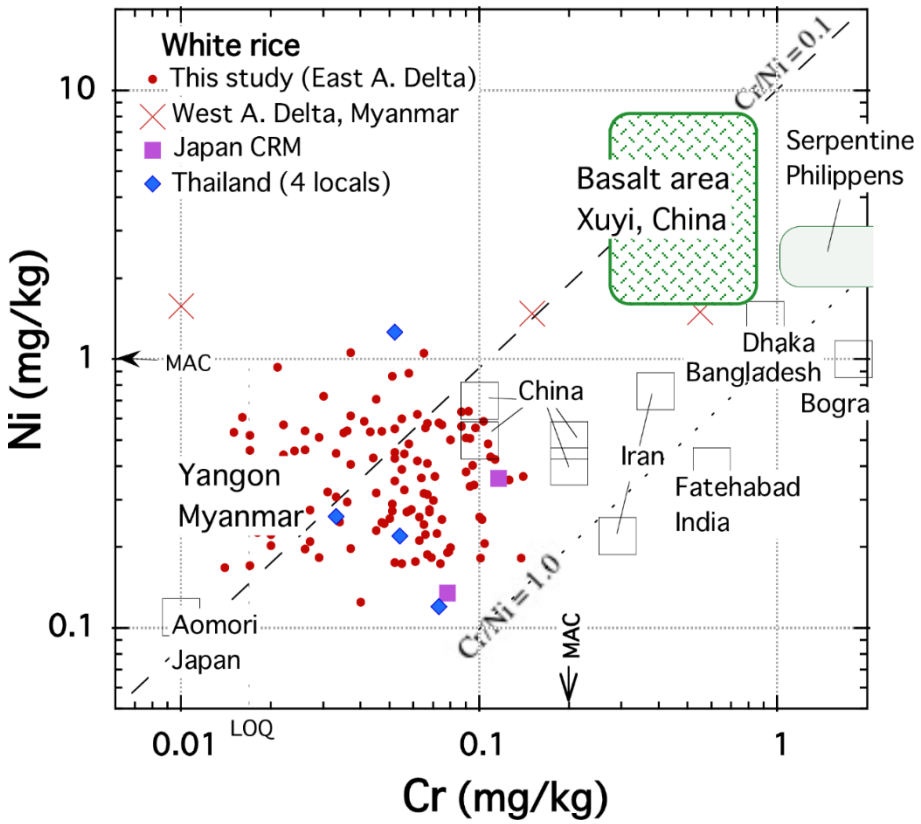


Fig 2.6. Correlation diagram between Ni and Cr concentrations in white rice from different countries and areas. Japan certified reference materials (CRM) data are from Table 2.2. The mean values of previous reports: West Ayeyawady delta in Myanmar (Khin Phyu Phyu 2017) (Phyo Wai Zin 2018); Japan certified reference materials (NMIJ CRM 7501-a and 7502-a) Xuyi, China (Wang et al., 2020); Shengzhou, Shimen, Fenghuang, and Xiangtan, in China (Zhao K et al., 2009) (Fu J et al., 2008); Bangladesh (Islam et al., 2014) (Islam et al., 2015); Iran (Naseri et al., 2015) (Jafari et al., 2018). Four local areas in Thailand (Kukusamude et al., 2021); Zambales, Philippines (Infante et al., 2021); Fatehabad, India (Poonam Yadav et al., 2016); Aomori, Japan (Tsukada H. et al., 2007). Incidentally, Aomori has one of the lowest population densities in Japan.

2.3.8 Daily intake of inorganic arsenic and exposure risk assessment from rice consumption

Table 2.3. Elemental concentration data in white rice samples, estimated average dietary intakes, and the potential health risks

Yangon white rice	**As**	**Pb**	**Cd**	**Cr**	**Ni**
Level (µg/kg)	155 ± 52	9.7 ± 8.2	5.6 ± 3.7	56 ± 29	393 ± 192
Range (µg/kg)	47–348	(0.7)–37.4	(0.3)–16.8	(6)–140	120–1060
MAC[c](mg/kg)	0.2 (iAs)	0.2	0.2/0.4*	1.0	0.5*/1.0
EDI[d](µg/kg–day)	1.34 ± 0.45	0.084 ± 0.070	0.048 ± 0.032	0.49 ± 0.25	3.39 ± 1.65
TDI[d] (RDA[d])	2.1 (iAs)	3.6	1	3.33	5.0
RfD[e/f](mg/kg–day)	0.0003 (iAs)	0.003	0.001	0.003	0.02/0.011
HQ [f]	4.46 ± 1.51	0.028 ± 0.023	0.048 ± 0.032	0.16 ± 0.08	0.31 ± 0.15
SF[c]/SFO[g](1/mg/kg–day)	1.5 (iAs)	0.0085	15.0	0.50	1.7/0.91
CR[f] (per 10,000 people)	7.5 ± 2.5	0.012 ± 0.010	2.7 ± 1.8	0.90 ± 0.47	11.4 ± 5.6

Yangon white rice	**Mn**	**Fe**	**Zn**	**Cu**	**Mo**	**Co**
Level (mg/kg)	10.9 ± 3.7	11.9 ± 14.9	17.3 ± 3.3	1.90 ± 0.60	0.32 ± 0.11	0.013 ± 0.005
Range (mg/kg)	6.9–23.0	2.0–52.6	10.9–26.3	0.74–3.64	0.12–0.66	0.004–0.030
MAC[c](mg/kg)			50	20		
EDI[d](µg/kg–day)	94.1 ± 31.5	103 ± 129	149 ± 29	16.3 ± 5.1	2.73 ± 0.99	0.112 ± 0.046
TDI[d] (RDA[d])			1000	71.4		
RfD[e/f](mg/kg–day)	0.14	0.7	0.3	0.04	0.0050	0.00030
HQ [f]	0.67 ± 0.23	0.15 ± 0.18	0.50 ± 0.10	0.41 ± 0.13	0.55 ± 0.20	0.37 ± 0.15

a: Results below LOD (limitation of detection) are shown as the LOD value in parentheses.

b: MAC: Maximum allowable concentration ((FAO, 2006) ; *(JECFA, 2005)); bw: body weight.

c: EDI: Estimated daily intake; TDI: Tolerable daily intake; RDA: Recommended daily allowance (Khanam et al., 2020; Kukusamude et al., 2021)

d: RfD (Oral Reference Dose); SF (Slope factor); HI: Hazard Index; TCR: Total CRs

e: RfD and SF values were compiled by (Adimalla, 2020; T. Li et al., 2018).

f: Non–carcinogenic and carcinogenic risks for As and Cr were calculated based on 100% inorganic As and Cr(VI) in rice samples.

g: Regional Screening level (RSL) Summary Table 2022 May (TR = 1E–06, THQ = 1.0) (*Regional Screening level (RSL) Summary Table 2020 November corrected (TR=1E-06, THQ=1.0)*, 2020)

Table 2.3. indicates that the total–As content in 120 polished rice samples was 0.155 ± 0.052 mg/kg, and the FAO/WHO proposed MAC of inorganic As (iAs) at 0.2 mg/kg for white rice (FAO, 2006). It indicates the tolerable daily intake (TDI) or recommended daily allowance (RDA) of inorganic As and some other elements (µg/kg body weight (bw)/day): TDI (= RAI) of inorganic As and Ni were 2.1 (µg/kg bw–day) and 5.0 (µg/kg bw–day (Khanam et al., 2020) On the contrary, the EDI of As and Ni in Yangon rice were 1.34 ± 0.45 and 3.39 ± 1.65 (µg/kg bw–day). For Pb, Cd, Cr, Zn, and Cu the EDI values by Yangon rice consumption were smaller than the TDI values by about an order of magnitude.

Although inorganic As (iAs%) in the Yangon white rice did not analyze, it is reported that Yangon deltaic rice has 0.04–0.05 mg/kg of inorganic arsenic (Nwe Ni Tun, 2020), and the iAs% value was assumed to be about 30% in this paper. Assuming iAs% to be 30%, the total value of the non–cancer hazard quotient index (HI) was 4.5 times the reference value of 1 (Fig 2.7. A). Expressing the value of HI in terms of the iAs (%) equation gives HI = 3.19 + 4.46 × iAs (%)/100. Arsenic (30%) is the main contributor to the HI value, although it changes with iAs%, followed by Mn, Zn, Cu, Mo, Co, and Ni (15–7%) and Pb, Cd, Cr, and Fe (below 4%). Assuming that iAs% was 30%, the total lifetime cancer risk (TCR) by these five elements was $1.73 \pm 0.59 \times 10^{-3}$, around 17 times higher than the USEPA upper limit of cancer risk for an environmental carcinogen of one case/ten thousand people (Table 2.3; Fig 2.7. B). TCR = $(1.50 + 0.75 \times \text{iAs}(\%)/100) \times 10^{-3}$. Nickel (66%) is the main contributor to the cancer risk value, followed by Cd (16%), As (13%), Cr (5.2%), and Pb (0.1%).

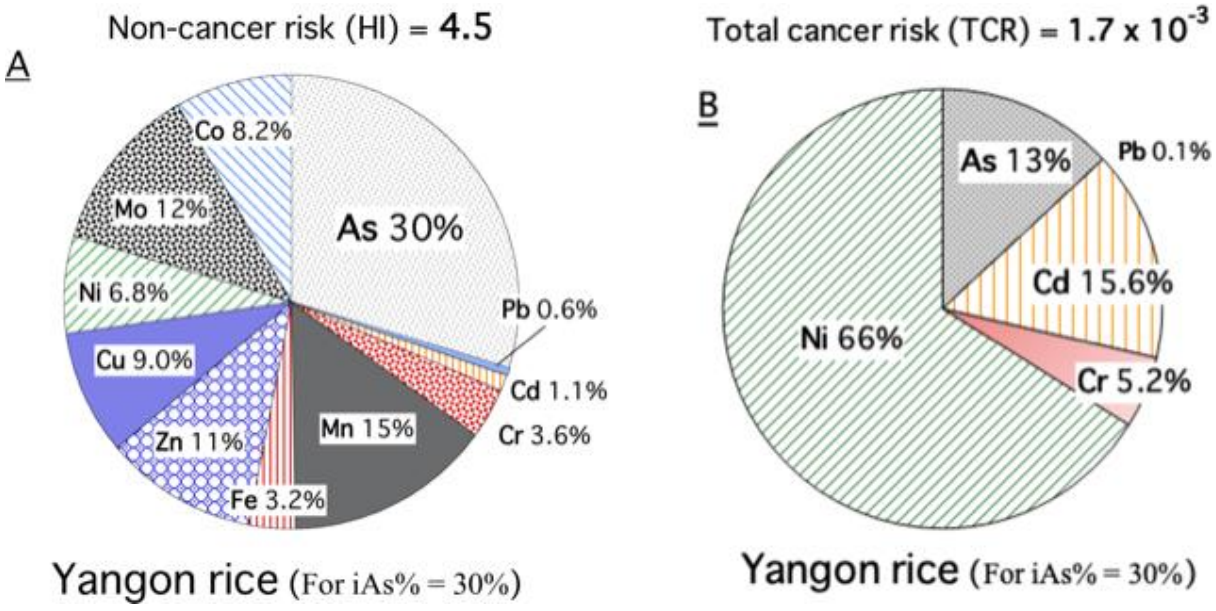


Fig. 2.7. (A,B) Potential health risks of non–cancerogenic and cancer risk.

The Yangon rice grains have average contents of nickel and arsenic worldwide from an unpolluted paddy field, but the lower contents of lead, cadmium, and chromium by about one magnitude than the worldwide contents, explaining the high–percentage carcinogenic risks by As and Ni of this study. The CR for our result was 2.2 (As), 11.4 (Ni), 2.7 (Cd), and 0.012 (Pb) with a TCR of 17.3 per 10^4 people, and these CR values were calculated by considering an exposure duration (ED) of 26 years in Yangon, Myanmar.

In the case of rural households in West Bengal, India, it was reported that the mean CR for these elements was 5.45 (As), 2.24 (Ni), 6.22 (Cd), and 0.16 (Pb) with a TCR of 14.1 per 10^4 people with ED of 10 years (Halder et al., 2020). As an extension of the ED will increase the value of CR, the comparison suggests that the carcinogenic risk of non–essential trace elements in Yangon rice is around an ordinary level.

2.4 Conclusion

This study revealed that “Yangon rice” samples from the east end area of the Ayeyawady delta have concentrations comparable to the lowest levels of lead, cadmium, and chromium contents of the literature values and a worldwide average level of As and Ni for the first time. We considered that these contents were natural background levels of the region. The data make it possible to detect and diagnose anthropogenic pollution in other delta parts and future pollution in this area. However, the chemical analysis of phosphorus and these elements in paddy soils and irrigation water is needed to confirm that the results of this study are at the natural background level. If the genotype of rice is responsible for the lowest levels of Pb, Cd and Cr in the world, it will be necessary to preserve the genotype in this region to reduce heavy metal intake. It is also necessary to find out the situation in most other parts of the Ayeyarwady Delta. Although arsenic and nickel, and not cadmium, are the main problems due to low cadmium concentrations in Yangon rice grains, cadmium contamination is a problem in other parts of this delta (Win-Thu et al., 2021).

The total non-cancer hazard quotient index (HI) caused by the Yangon rice's heavy metals intake was calculated to be 4.5 times the reference value. The As intake accounted for 30%, assuming that iAs% was 30%. The average daily intake of total arsenic in Myanmar is calculated to be 78 ± 26 (μg/person/day) in Table 2. Cardiovascular disease (CVD) in association with iAs exposure are well documented; for example, it is reported that CVD risks increased with iAs exposure from rice at exposures above 0.3 μg/person/day (Xu et al., 2020). The potential health hazards of As intake via drinking water in Myanmar have been reported (Mochizuki et al., 2019) (Mar Wai et al., 2019). Therefore, inorganic arsenic intake from rice (Nwe Ni Tun, 2019) should also be noted as a potential issue. Various countermeasures (e.g.,(Atsuda, 2009)(Wang et al., 2020)), such as rice cooking improvement (e.g., (Menon et al., 2021)) and copper oxide nanoparticles input (J. Liu et al., 2019b), have been proposed to reduce the As intake amount by rice consumption. The cancer risk index (TCR) from heavy metal intake in Yangon rice was calculated to be 17 times higher than the USEPA limit, with Ni intake accounting for two-thirds of the index. The high cancer risk of

Ni intake has been reported in environmentally contaminated or mafic/ultramafic areas: nickel concentrations in rice in the literature (Wang et al., 2020) (H. B. Li et al., 2020) were 0.84 ± 0.40 mg/kg, 2.66 ± 1.46 mg/kg, and 1.22-9.34 mg/kg, which were two times to one order of magnitude higher than the Ni level (0.39 ± 0.19 mg/kg) of this study. Even though the Ni level in Yangon rice grains is around the global average range in uncontaminated and non-mafic areas, the high carcinogenic risk of Ni intake from Yangon rice consumption was higher than that of arsenic or cadmium. Although the risk of nickel intake has received less attention, hidden by the magnitude of the risk of Cd and As so far, this study indicates that Ni intake from rice should be highlighted as an alternative potential risk.

Ultramafic rock areas are located in northern Myanmar, and nickel content and estimated daily intake (EDI) in other foods, such as beans (Shaheen et al., 2016), remain to be investigated. Unlike arsenic, improving cooking may not reduce nickel intake because Ni and Cd even distribute throughout the endosperm of white rice, while As localised in the outer surface of a white rice grain (Lombi et al., 2009). The geochemical behaviour of nickel in paddy soils (Dhal et al., 2013) allows envisaging the improvement to reduce the bioaccumulation of nickel in rice grains remains challenged. The rice genotype might be a helpful tool for mitigating Ni toxicity (Spanu et al., 2020).

Chapter 3- Heavy metal concentrations in paddy soils and brown rice in Yangon Division

3.1 Introduction

Arsenic is a harmful element and it can be found in the earths crust naturally. It can be used in many way of industries and agriculture. Natural weathering and anthropogenic activities can pose arsenic poisoning or contamination which can enter in the environment such as air, water, soil and food. Arsenic can accumulate in paddy soil throughout natural weathering and alluvial deposits. Usage of As accumulated ground water for irrigation in the paddy fields cause As concentration high (Jia et al., 2015). Other sources of As in paddy soils came from agriculture poducts such as phosphate based fertilizers, cattle manures, and use of arsenical based pesticides. (Meharg and Zhao, 2012). In paddy soil, As is mainly found in its inorganic form as trivalent arsenite (As(III)) or pentavalent arsenate (As(V)), with the most common organic As species being monomethylarsonic acid (MMA(V)) and dimethylarsinic acid (DMA(V)), which are generally exist as lower concentrations (Campbell & Nordstrom, 2014). The main source of As bioavailabily for human is food and drinking water. From As concentration in the paddy soil, the rice plant take up As and come into human life via dietary intake.

Rice is usually grown in Yangon division, Ayeyarwady Delta under anaerobic conditions in which As (III) was main form in the flooded water and can be easily take up by the rice plant. It is well known that rice grown under flooded conditions cause much more arsenic to accumulate than that grown under aerobic conditions. Arsenic accumulation in rice grain was observed to be 10- to 15-fold higher in flooded than in aerobically grown rice (Gharachorloo et al., 2019). It has been reported that rice plants can accumulate arsenic from the soil approximately 13- and 20-fold greater amounts as compared to wheat and barley, respectively (Gharachorloo et al., 2019). The study area grows rice mainly the whole year and cover the half of thr rice production of the whole country.

3.1.1 Problem Statement

Rainfed paddy field of the study area is under flooded conditions . This area is not in the residential area and it is far from mining and industrial zone. As and Ni concentration of rice grain in this study is due to the soil concentration as geogenic source or anthropogenic activities.

3.1.2 Objective

- To determine the concentration of heavy metal in the rainfed paddy soil

3.2 Material and Method

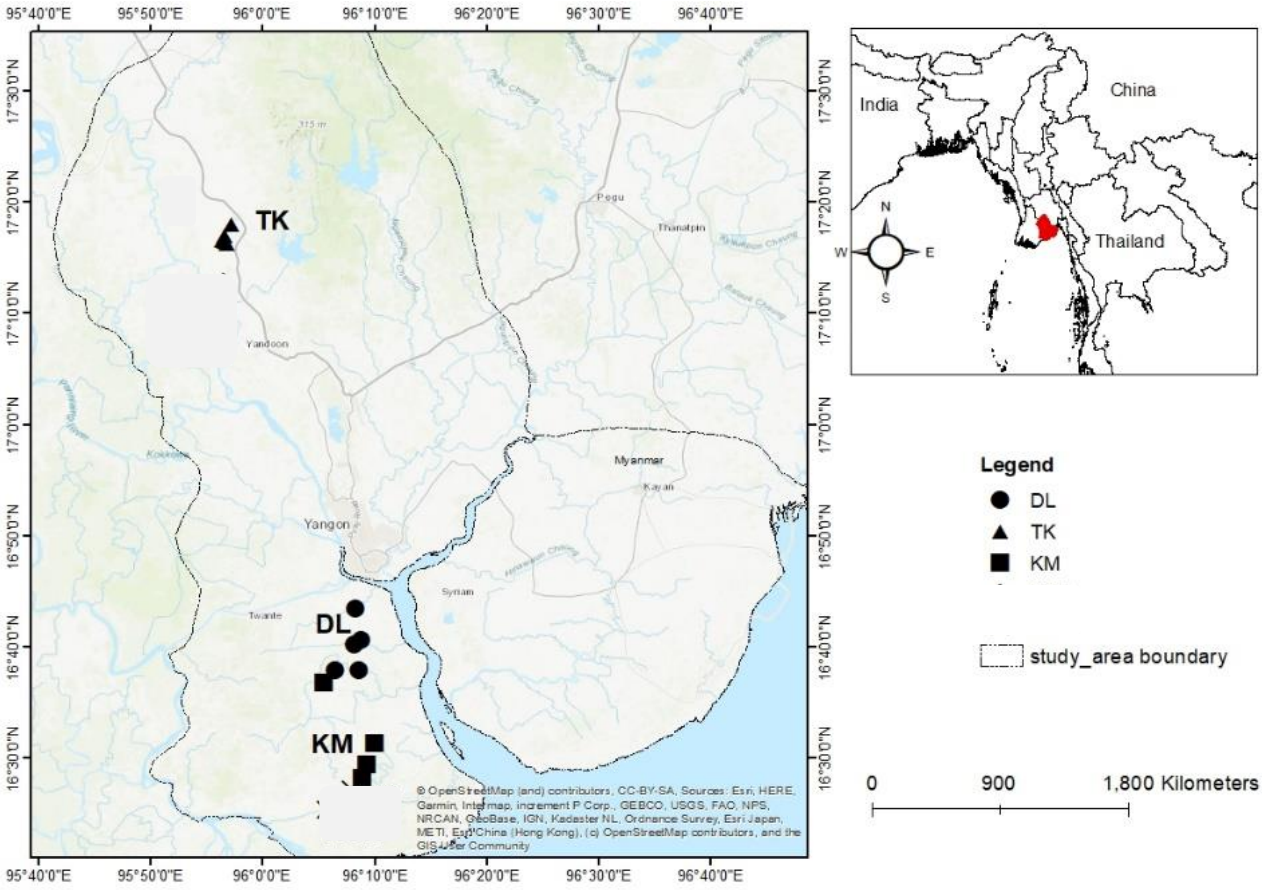


Fig. 3.1 Map of the study area and sampling points.

3.2.1 Sample collection

This second time sampling was done in December 2019 and the paddy, soil, and water samples were collected from three townships, TK, DL, and KM. A total of 82 soil samples TK (n = 28), DL (n = 30), KM (n = 24), and 200 rice paddy samples TK (n = 70), DL (n = 73),

KM (n = 57) were collected from the three sites in each field and included four corners and the middle of the paddy.

The soil was obtained from ca. 0−15 cm depth using a soil probe that was pushed down into the soil and was collected from three townships (DL, KM, and TK). The latitude, longitude, and altitude data of the sampling point were obtained with the GPS tool. A total of 50 polished rice samples, which were freshly removed from the husks at the factories before transporting them to the markets were collected from the Ayeyarwady Delta. Several 46 rice samples were obtained at Yangon Rice Market where the farmers were trading their rice.

All the rice samples were sent for checking pests to Yangon, Myanmar Agriculture and Irrigation Minister under Plant Pest Quarantine Regulation to import to Japan for research purposes. Additionally, all the samples in this research were imported under the permit of the Minister of Agriculture, Forestry and Fisheries, Japan under the Plant Protection Law.

3.2.2 Sample preparation

The second time samples December 2019 were field paddies and polished rice. The husks of the paddies were crushed and removed by rolling with the log. The samples were treated on a clean bench and digested with certified reference materials (CRMs) and blank samples in runs. About 10 g of collected white (polished) rice grains were finely powdered by agate mortar, homogenized, and dried in a hot air oven (55°C for 36 h) until the weight became constant.

3.2.3 Soil

The soil samples are air–dried at room temperature at Toyoda laboratory of Hokkaido University. The dried samples were pulverized using a motor and pestle and then passed over a 1.5 mm sieve.

Fig. 3.2 Soil sample air–dried at Toyoda laboratory.

3.2.4 Analytical Procedure for brown rice

The powdered rice sample (30 mg) was weighed and placed in a 7 ml PFA vial (Savillex) and then mixed with 1mL of ultrapure–grade 69% (v/v) nitric acid and 0.1 mL of 30% (v/v) H_2O_2 (Kanto Chemical). After the digestion in the closed PFA vial in a hot air oven at 80°C for 12 hours, each sample solution was diluted into 30 mL with ultrapure water (milli–Q, Simplicity UV, and Gradient with Elix10, Millipore) from the cleanroom facility of the school.

Eleven elements in all sample solutions stocked in clean polypropylene bottles were determined with an Agilent 8800 triple quadrupole ICP–MS (ICP–QQQ) at the Open Facility

of Hokkaido University. The result of raw data was ppb and the calculation was carried out based on the weight and solution of the sample in terms of mg/kg.

3.2.5 Analytical procedure for soil

For the extraction process, 1 g of soil sample was digested with 4 ml of hydrochloric acid and 1 ml of conc. nitric acid at 70°C for 3 hours. The digested solution was diluted with deionized water and then centrifuged at 5800 rpm for 30 minutes. The samples were filtered using 0.22 μm cellulose acetate membrane syringe filters and then diluted the sample before the analysis. Analysis of heavy metals was done using Inductively Coupled Plasma Atomic Emission Spectrometry (ICP–AES).

Fig. 3.3. Soil sample extraction at Toyoda laboratory.

3.2.6 Method of validation

Data precision and accuracy were checked by the analysis of certified reference materials: NMIJ CRM 7501–a, 7502–a, from the National Metrology Institute of Japan for the rice and JSd–2 and JSd–3 standard samples from the Geological Survey of Japan for the soil samples. The reference samples were put in each batch of digestion rice and soil. After analysis, the recovery rates were checked and the results were compared with the certified values for the

respective standard sample. The blank samples were set for checking the contamination of the digestion.

3.3 Results and Discussions

3.3.1 Accuracy Analysis

The quality of analytical procedures checked by the analysis of certified reference materials were shown in Table 3.1. The results of the standard reference material for sediment and the results were consistent.

Table 3.1: Analytical result of heavy metals in two certified reference materials (mg/kg)

	Extracted JSd–3 (n=6)			
mg/kg	Results	±	SD	Bulk of literature (2)
As	84.3	±	9.71	252
Pb	60.8	±	6.35	82.1
Cd	1.46	±	0.16	1.05
Zn	78.5	±	9.35	136
Cu	372	±	34.04	426
Ni	12.5	±	1.15	19.6
Co	8.37	±	0.91	12.7
Cr	11.0	±	1.20	35.3
Fe (%)	1.14	±	0.27	3.06
Mn (%)	0.04	±	0.01	0.11
Ca (%)	0.101	±	0.01	0.56
Mg (%)	0.116	±	0.09	1.17
Na (%)	0.020	±	0.00	0.41
K (%)	0.199	±	0.02	1.97

Jsd–3,sediment reference material, Geological Survey of Japan, 1996

3.3.2 Heavy metal concentration in soils

DL, TK, and KM were chosen to collect the soil samples due to the results of the arsenic concentration of first–time polished rice. The concentration nine elements Co, As, Cd, Cr, Fe, Mn, Cu, Zn and Ni were analyzed in 82 soil samples. Fig. 3.4. A-C shows the heavy metal concentrations in the soil from DL, TK, and KM. For As and Cd, a total of 5 samples from each area represent a whole.

The mean and range of soil As in TK, DL and KM were 1.14 (0.67−1.52), 4.90 (3.41−8.50), and 5.26 (3.48–7.26) mg/kg. The total As the concentration of TK area had lower soil As than DL and KM. The range of As concentration in soil (0.67−8.50) of this study fell into Cambodian rainfed soil range (0.8–18 mg/kg) and Bangladesh rainfed range (0.8−21 mg/kg) (Seyfferth et al., 2014) (Ahmad, 2003).

The typical Cd concentrations in uncontaminated soils occur in the range of 0.06–1.1mg/kg (Gałuszka et al., 2015). In this study, the mean and range of Cd concentrations in the soil of DL, KM, and TK were 0.22 (0.02−0.03), 0.02 (0.017−0.03), and 0.02 (0.02−0.04) mg/kg respectively. The concentrations were below the typical range and it can be assumed no manmade activities were there (Fig. 3.4. A).

Of the other 8 elements of Co, As, Cr, Fe, Mn, Cu, Zn and Ni, TK has the lowest concentrations among the three townships. Compared with downstream floodplain DL and KM (4 −7 m), the high land of TK area (16–18 m) has low elemental composition due to the fewer deposits. The DL and KM site on the downstream sides may be arsenic–rich sedimentary layers eroded from the upstream that could be carried and quickly deposited topographically during floods.

The low concentration of As in TK might be related to the low iron and manganese oxides of this area (Fig. 3.4. A, B). High As concentrations in KM and DL are associated with high Fe

and Mn in these areas. It can be the paddy soil of KM and DL is enriched with As bearing iron and manganese hydroxides. It is well known that As derived from the Holocene sediments of Himalaya ranges was associated with high concentration and Fe and Manganese. The migration of Himalaya derived As bearing iron oxides show high As concentration in DL and KM. (Fig. 3.4. A-B).

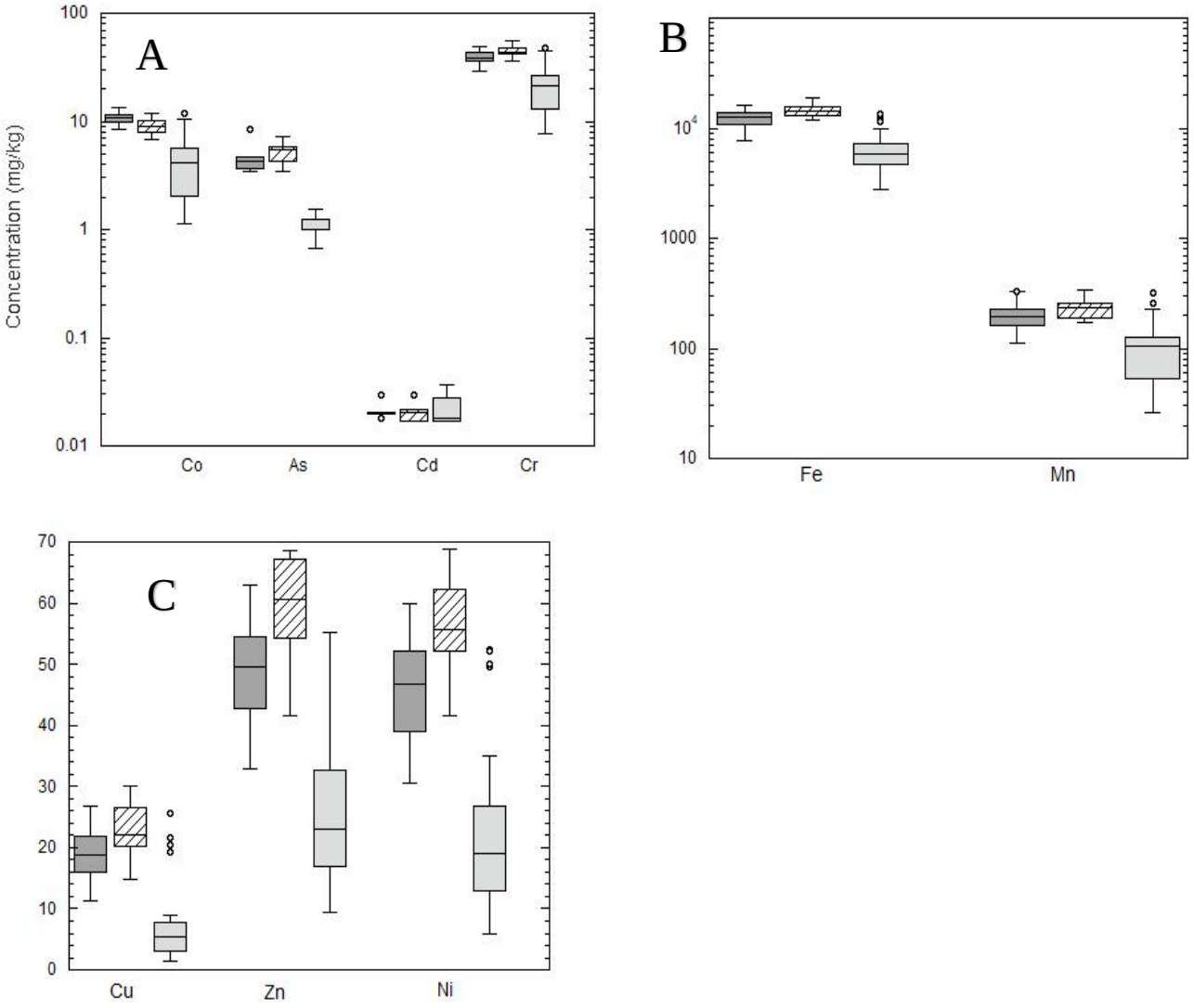


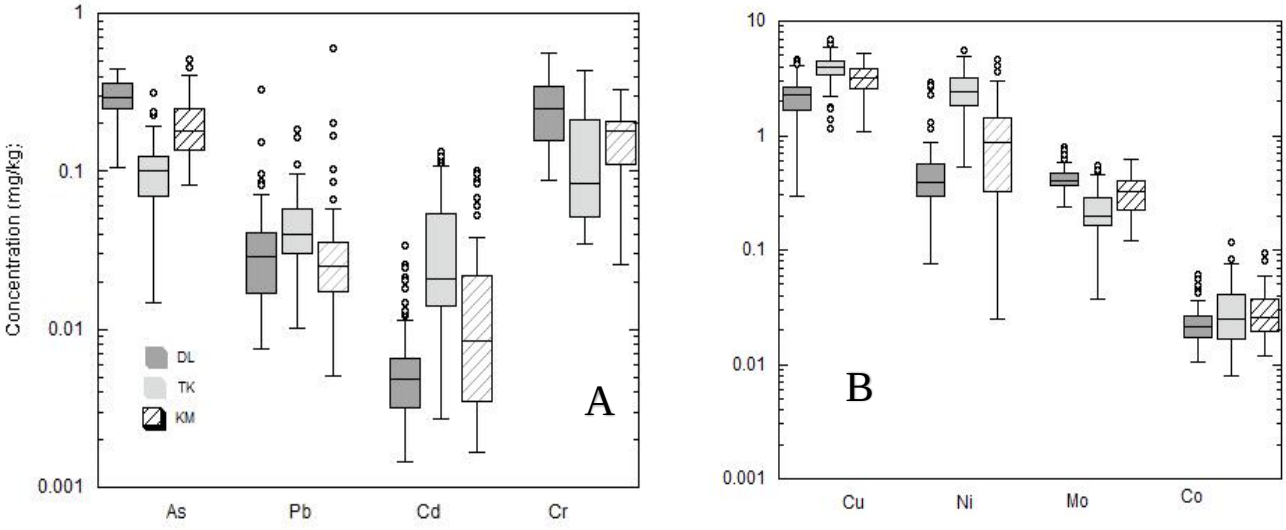
Fig. 3.4. (A-C) Co, As, Cd, Cr, Fe, Mn, Cu, Zn and Ni content in soil.

3.3.3 Heavy metal concentration in unpolished rice

The concentration of nine elements As, Pb, Cd, Cr, Mn, Fe, Zn, Cu, Ni, Mo, and Co were analyzed in 200 rice samples from DL, KM, and TK. Fig. (3.5. A–C) showed a box plot of each element concentration in unpolished rice samples from three townships.

Total As concentrations in unpolished rice grains in the present study ranged from 0.015–0.513 mg/kg. The range of Cambodia rainfed unpolished rice was (0.10 – 0.37 μg g^{-1}) and this study result is similar to this range. The average As is 0.20 mg/kg in this study and it is consistent with Cambodia unpolished rice (0.20 μg g^{-1}) which was affected by As owing to the deposition of Himalayan-derived Mekong River sediments (Seyfferth et al., 2014). The high As concentration was found in KM (0.513 mg/kg) and DL (0.441 mg/kg) whereas the high As concentration in soil was found.

The low As concentration found in TK might due to the high Cu and Ni concentration because high Cu and Ni concentration suppress the transfer of As from root to shoot (Yao et al., 2020).



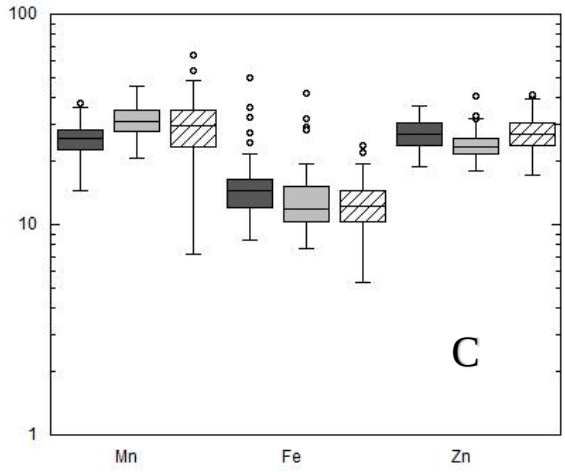


Fig. 3.5. (A-C) As, Pb, Cd, Cr, Cu, Ni, Mo, Co, Fe, Mn, Fe and Zn content in unpolished rice.

3.3.4 Regression analysis between unpolish rice and soil (As and Ni)

The unpolished rice was collected to check the migration of As and Ni from soil to unpolished rice because they were collected at the same spot of soil and paddy. Regression analyses were performed on the potential relationships between unpolish rice and soil. It was performed to find the transfer of element from soil to rice grains. Soil As in our study had significantly straightforward relationship with grain (Fig. 3.6 (A)). It is consistent with the study of Bengal delta rainfed paddy rice and soil (Y. Lu et al., 2009). It might be suggested that the arsenic concentration in unpolished rice is related to the arsenic concentration in the soil. Therefore, the amount of grain As in flooded rice paddies is more heavily influenced by soil chemical conditions.

In this study, the soil nickel concentration and grain nickel was found moderate negative correlation (Fig. 3.6 (B)). It is reported that phosphatic fertilizers are manufactured by using the phosphate ore produces Ni as an impurities. That can be one of the reasons for Ni in grain. Another one is excessive usage of pesticide and the sewage sludge usage as fertilizer. The

study area used the cattels for ploughing the soil and the cattle dung might be a reason for nickel concentration in the grain (Tariq & Rashid, 2013).

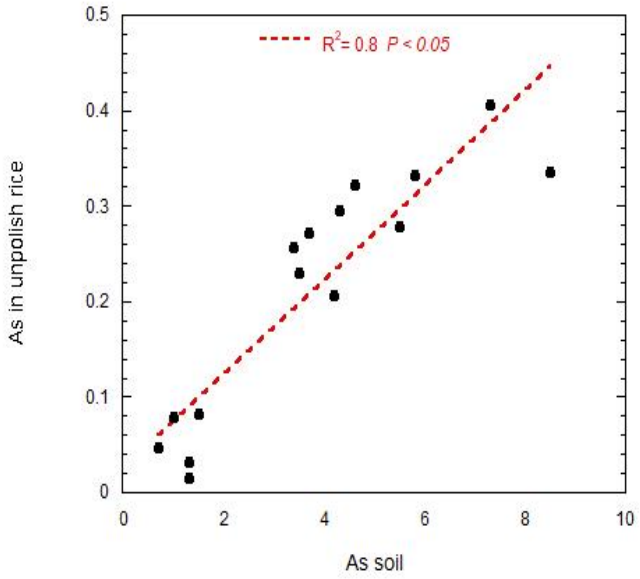


Fig. 3.6. (A) Regression plot of soil As with unpolished rice grain As.

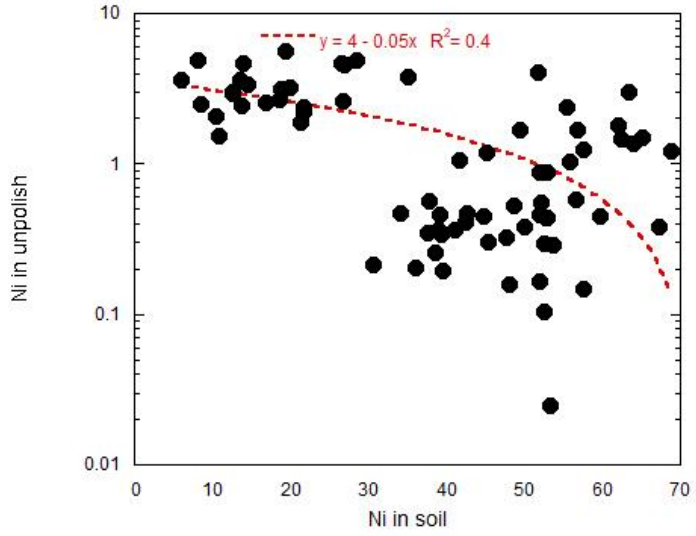


Fig. 3.6. (B) Regression plot of soil Ni with unpolished rice grain Ni.

3.3.5 Elemental Concentration in Factories rice and Market rice

The average concentration of As, Cd, Co, Mo, Mn, Fe, Zn, and Cu in factories were 0.13, 0.08, 0.014, 0.42, 12.26, 4.81, 20.8, and 2.88 mg/kg respectively. For market rice, the concentrations were 0.11, 0.02, 0.01, 0.33, 10.54, 5.12, 17.22, and 2.12 mg/kg (Fig.3.7. (A)(B)). Factories rice is collected from the rice factories in the rainfed delta area. Markets rice is collected at the rice markets where we can get the rice grown all over Myanmar. Although the factory rice has some high content, people don't buy the rice at the factory. Storing time and transporting duration to the market might cause the concentration to reduce in market rice.

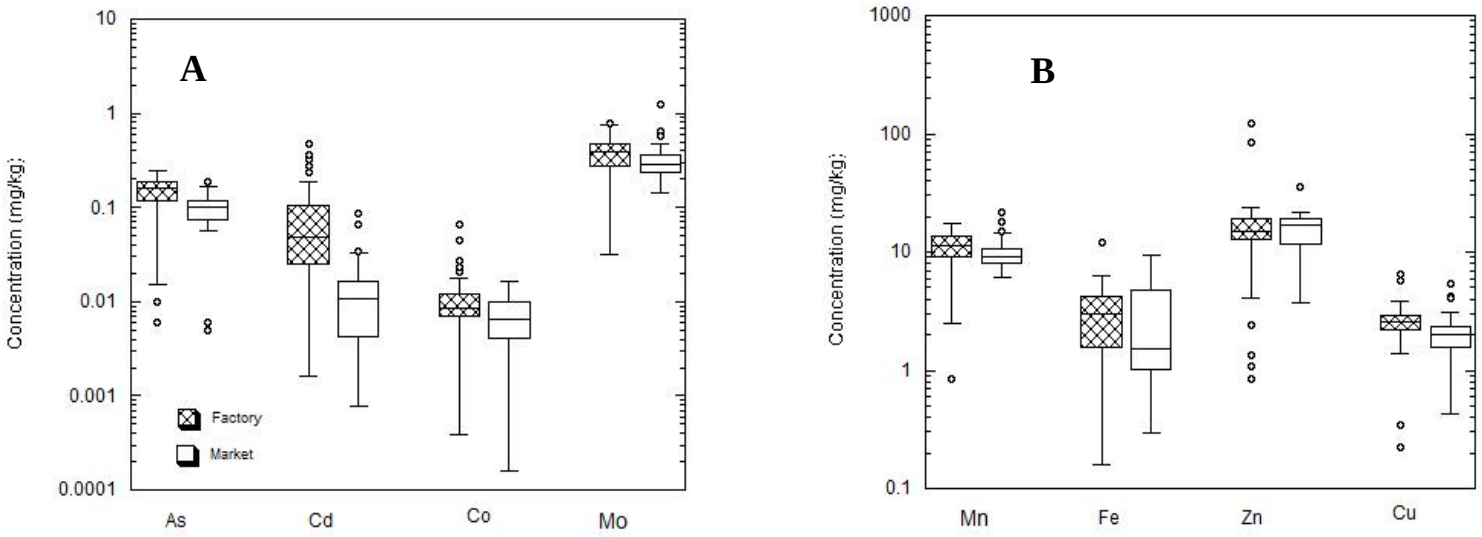


Fig. 3.7. (A-B) As, Cd, Co, Mo, Mn, Fe, Zn, and Cu contents in factories and market rice.

3.4 Conclusion

In this study, the concentration of As in unpolished rice was controlled by soil As. Anthropogenic activities like usage of fertilizers and pesticides can cause high As concentration in rice grain. But it has been found that the soil of this study has the enrich Fe, Mn and As as a geologic source. The strong and positive regression analysis reveals the rice grain As was under the influences of soil As. The range of As concentration in soil of this study fell into rainfed As soil range which is far from the human activities.

The Cd concentrations of soil were below the typical range and it can be said no contaminated by manmade activities. The regression analysis of nickel showed grain nickel was not correlated with soil nickel. There might be some factors of fertilizer, pesticides and cattle waste which applied in rice growing. It can be seen As contents in unpolished rice was reduced by milling or polishing processes at factories. The As contents of factories rice was again reduced by storing time to the market rice.

Chapter 4 General Discussion and Conclusion

4.1 The concentration of heavy metal in white rice

In Asia, As contamination in rice was well known due to As contaminated irrigation water. But in this study we want to check As concentration in rice where rain water is used for growing rice. But only one area, TK of this study uses pond water for irrigation. This study aim to check the safety of Yangon rainfed rice. The study area, five townships in Yangon division is important because it is a part of Ayeyarwady Delta which bears half of country rice production. The elemental analysis was done in white rice by ICPMS and compared with maximum allowable concentration set by FAO and WHO.

The highest median and the maximum value in As concentration of rice grains were observed in DL township (0.183, 0.348 mg/kg). The medians and highest As concentrations in TK and KM townships were 0.152, 0.290, and 0.152, 0.244 mg/kg. The mean total-As content in 120 samples was 0.155 ± 0.052 µg/g, and the WHO proposed MAC of inorganic As (*i*As) at 0.2 µg/g for white rice. Meharg et al. (2009) reported that 0.15 µg/g was the mean total-As content in white rice grains (n = 901) from global ten countries. Most of the As concentrations in this study are below the MAC value and are close to the average rice concentration globally.

The range of the Ni in the rice was 0.168–0.935 mg/kg for DL, 0.12–1.06 mg/kg for KM, 0.18–0.89 mg/kg for TK, 0.174–1.66 mg/kg for HTP and 0.137–0.729 mg/kg for KCK. In contrast to the MAC values in white rice (1.0 mg/kg of Ni), Ni average of rice grains was 0.39 ± 0.19 mg/kg and was lower than the MAC. The mean and range of Fe in DL was 13.8 (4.91–35.8) mg/kg. It may translate that paddy soil in DL townships is more enriched in Fe than the other townships. Iron is primarily mobilized chiefly as dissolved Fe(II) under reducing conditions, and it is redistributed as particulate Fe (III) oxyhydroxides in oxygenated environments (Langmuir, 1997). The soil in the DL townships is richer in iron oxide components than the other township.

The low Cu contents in TK rice might be associated with the high As contents likewise Fe contents in the rice. It was reported that the suppression effect of Cu on As accumulation in white rice (J. Liu et al., 2019a). The pond water irrigation contributes to the Cu and Fe-depleted paddy soils and high As contents in rice grains at TK township.

In this study, the Cd concentrations in white rice grains were 5.6 ± 3.7 μg/kg. It is comparable to the lowest concentration in the literature values. Most Cd exists in the form of CdS, which has low solubility (Daskalokis KD, 1992). Since the study area in the Ayeyawady delta is annually exposed to flooding during the heavy monsoonal rainy season, the soil is under reducing conditions. The irrigation system should contribute to the rice grains' low Cd concentration.

This study showed that the Pb mean concentration in white rice grains was 10 μg/kg. It corresponds to the second-lowest concentration after Japan (1.8–3.0 μg/kg; (Shimbo et al., 2001)). The lead (Pb) has a lower solubility than cadmium in the paddy soil of the delta. Brookins 1988 suggests that most Pb exist in the form of PbS or $PbCO_3$, which have low solubilities. The hydrous ferric oxide, formed at the soil surface under the floodwater has excellent adsorption properties against Pb ions and other toxic ions (Waychunas et al., 2005)). The phosphate-based fertilizer into paddy soils promotes the formation of stable lead phosphate mineral pyromorphite (Ruby et al., 1994), which has very low solubility (Flis et al., 2011).

As a summary this study revealed that “Yangon rice” samples from the east end area of the Ayeyawady delta have concentrations comparable to the lowest levels of lead, cadmium, and chromium contents of the literature values and a worldwide average level of As and Ni for the first time.

4.2 Health risk assessment

In this study, the estimated daily intake of As and Ni in Yangon rice were 1.34 ± 0.45 and 3.39 ± 1.65 (μg/kg bw–day). For Pb, Cd, Cr, Zn, and Cu, the EDI values by Yangon rice consumption were smaller than the TDI values by about an order of magnitude. The total value of the non–cancer hazard quotient index (HI) was 4.5 times the reference value of 1. Arsenic (30%) is the main contributor to the HI value, although it changes with iAs%, followed by Mn, Zn, Cu, Mo, Co, and Ni (15–7%) and Pb, Cd, Cr, and Fe (below 4%). Assuming that iAs% was 30%, the total lifetime cancer risk (TCR) by these five elements was $1.73 \pm 0.59 \times 10^{-3}$, around 17 times higher than the USEPA upper limit of cancer risk for an environmental carcinogen of one case/ten thousand people. Nickel (66%) is the main contributor to the cancer risk value.

4.3 Heavy metal concentration in paddy soil

Rainfed paddy field of this study area is under flooded conditions except TK area which uses pond water. This study aim to confirm As and Ni concentration of rice grain is due to the soil concentration as geogenic source or anthropogenic activities. It was found that high As concentrations in KM and DL are associated with high Fe and Mn in these areas. It can be the paddy soil of KM and DL is enriched with As bearing iron and manganese hydroxides. It is well known that As derived from the Holocene sediments of Himalaya ranges was associated with high concentration and Fe and Mn. The migration of Himalaya derived As bearing iron oxides show high As concentration in DL and KM. The low concentration of As in TK might be related to the low iron and manganese oxides of this area as a geologic source.

Soil As in our study had significantly straightforward relationship with grain. It is consistent with the study of Bengal delta rainfed paddy rice and soil (Y. Lu et al., 2009). It might be

suggested that the arsenic concentration in unpolished rice is related to the arsenic concentration in the soil. Therefore, the amount of grain As in flooded rice paddies is more heavily influenced by soil chemical conditions.

The soil nickel concentration and grain nickel was found moderate negative correlation. It is reported that phosphatic fertilizers, excessive usage of pesticide and the sewage sludge produces Ni as an impurities. The study area use the cattel for ploughing the farm cattel dung used for ploughing the farm and the cattle dung might be a reason for nickel concentration in the grain (Tariq & Rashid, 2013). Therefore this study shows As concentration of the rice grain was due to geologic source and Ni concentration might be anthropogenic activities.

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Appendices

Table. Concentrations (mg/kg) of 11 elements in 120 white rice samples from five townships (TK, HTP, DL, KM, KCK) in Myanmar.

Area name		As	Pb	Cd	Cr	Mn	Fe	Zn	Cu	Ni	Mo	Co
	No.	120	115	115	118	120	119	117	119	119	120	119
TK	**1**	0.240	0.0076	0.0024	0.051	10.2	5.0	19.2	0.74	0.273	0.251	0.0194
	2	0.120	0.0060	0.0074	0.075	13.7	4.7	17.8	1.48	0.570	0.221	0.0153
	3	0.083	0.0093	———	0.087	11.1	5.3	11.9	1.87	0.563	0.361	0.0066
	4	0.180	0.0374	0.0017	0.065	12.3	4.6	19.5	1.35	0.317	0.135	0.0156
	5	0.174	0.0075	0.0049	0.107	12.1	6.5	17.3	1.72	0.484	0.228	0.0149
	6	0.234	0.0034	0.0030	0.102	10.4	4.0	19.2	0.74	0.253	0.247	0.0187
	7	0.143	———	0.0030	0.080	20.9	13.4	19.2	1.88	0.502	0.177	0.0260
	8	0.154	0.0054	0.0033	0.059	11.4	4.0	13.5	1.25	0.277	0.116	0.0101
	9	0.185	0.0150	0.0022	0.096	12.2	9.8	20.2	1.39	0.341	0.146	0.0174
	10	0.250	0.0039	0.0022	0.068	11.1	3.2	20.3	0.75	0.270	0.247	0.0178
	11	0.105	0.0056	0.0070	0.066	13.3	4.2	16.7	1.40	0.555	0.211	0.0126
	12	0.149	0.0040	0.0026	0.058	11.4	3.3	13.7	1.25	0.273	0.121	0.0091
	13	0.142	0.0152	0.0050	0.070	12.4	4.3	13.2	1.26	0.299	0.235	0.0098
	14	0.103	0.0165	0.0061	0.140	8.9	5.7	20.1	1.47	0.367	0.307	0.0148
	15	0.290	0.0052	0.0033	0.126	14.1	5.2	23.8	0.93	0.356	0.309	0.0215
	16	0.151	0.0061	0.0027	0.057	11.6	3.0	13.7	1.21	0.270	0.130	0.0090
	17	0.108	0.0042	0.0043	0.093	13.3	5.0	16.8	1.36	0.508	0.208	0.0126
	18	0.203	0.0235	0.0019	0.071	13.0	5.8	22.5	1.56	0.366	0.161	0.0182
	19	0.116	0.0071	0.0072	0.052	13.6	17.4	18.0	1.51	0.552	0.226	0.0120
	20	0.157	0.0092	0.0044	0.080	9.7	6.3	15.1	1.75	0.199	0.398	0.0091
	21	0.115	0.0071	0.0063	0.043	12.9	4.7	17.5	1.47	0.537	0.219	0.0105
	22	0.263	0.0061	0.0025	0.100	10.7	3.8	20.1	0.78	0.259	0.262	0.0180
	23	0.084	0.0161	———	0.058	16.2	4.0	12.7	2.35	0.887	0.197	0.0077
	24	0.189	0.0067	0.0031	0.061	9.2	5.0	22.4	1.27	0.177	0.340	0.0124

HTP	**1**	0.133	0.0137	0.0028	0.101	6.9	22.3	18.1	1.49	0.182	0.236	0.0172
	2	0.097	0.0037	———	0.051	8.7	3.4	20.5	2.36	0.289	0.658	0.0038
	3	0.124	0.0075	0.0047	0.055	9.3	4.6	13.4	1.76	0.174	0.338	0.0083
	4	0.133	0.0117	0.0045	0.068	10.4	6.2	14.7	1.73	0.277	0.311	0.0083
	5	0.131	0.0071	0.0036	0.095	9.9	35.8	18.7	1.47	0.402	0.442	0.0225
	6	0.110	0.0036	0.0097	0.065	11.6	6.3	12.3	3.58	1.055	0.222	0.0122
	7	0.199	0.0071	0.0038	0.069	11.2	5.8	20.6	2.09	0.183	0.492	0.0118
	8	0.133	0.0141	0.0027	0.052	7.0	39.9	17.7	1.36	0.175	0.203	0.0180
	9	0.113	0.0069	0.0033	0.064	10.1	6.8	19.5	2.28	0.362	0.322	0.0090
	10	0.086	0.0283	0.0052	0.087	9.7	35.6	11.9	2.48	0.638	0.256	0.0277
	11	0.099	0.0124	0.0072	0.033	8.4	6.3	16.4	2.05	0.447	0.333	0.0079
	12	0.145	0.0119	0.0036	0.047	10.6	5.8	14.9	1.85	0.247	0.357	0.0099
	13	0.132	0.0087	0.0092	0.056	7.8	15.8	16.5	2.03	0.444	0.482	0.0093
	14	0.176	0.0145	0.0021	0.033	12.2	4.7	19.4	1.27	0.307	0.141	0.0150
	15	0.142	0.0116	0.0087	0.036	10.8	8.5	21.3	2.48	0.541	0.493	0.0129
	16	0.126	0.0130	0.0039	0.045	9.5	43.7	14.5	1.74	0.231	0.314	0.0099
	17	0.133	0.0077	0.0027	0.055	8.8	52.6	17.1	1.37	0.390	0.458	0.0193
	18	0.140	0.0147	0.0015	0.079	7.0	41.2	18.1	1.38	0.193	0.207	0.0193
	19	0.136	0.0094	0.0074	0.029	10.5	19.3	12.2	2.44	0.513	0.403	0.0121
	20	0.047	0.0123	———	0.082	12.9	16.4	———	———	———	0.501	0.0286
	21	0.094	0.0141	0.0050	0.037	8.3	16.9	19.6	1.32	0.198	0.268	0.0098
	22	0.109	0.0120	0.0063	0.047	8.9	32.5	19.2	2.29	0.540	0.398	0.0084
	23	0.135	0.0068	0.0075	0.109	7.2	———	18.9	1.89	0.434	0.490	0.0123
	24	0.152	0.0105	0.0099	0.041	11.6	8.2	13.5	2.74	0.588	0.474	0.0142
	25	0.147	0.0248	0.0057	0.058	17.3	25.9	15.3	2.42	0.485	0.311	0.0117

DL	**1**	0.107	0.0073	0.0047	0.056	8.9	11.1	18.2	2.13	0.326	0.298	0.0096
	2	0.118	(0.0018)	0.0035	0.044	8.7	7.0	17.6	1.96	0.430	0.237	———
	3	0.194	0.0028	0.0016	0.037	16.2	14.8	24.1	1.47	0.406	0.310	0.0300
	4	0.238	0.0371	0.0140	0.062	13.9	7.6	14.8	2.85	0.625	0.344	0.0121
	5	0.100	(0.0021)	0.0100	0.021	9.0	4.9	11.6	3.29	0.935	0.190	0.0108
	6	0.148	0.0066	0.0069	0.031	10.0	12.4	17.6	1.64	0.321	0.345	0.0098
	7	0.164	0.0035	0.0051	0.024	7.7	25.6	17.0	2.14	0.289	0.332	0.0096
	8	0.163	(0.0019)	0.0022	0.021	7.0	6.9	22.8	1.41	0.260	0.421	0.0108
	9	0.216	0.0307	0.0129	0.026	12.3	5.9	———	2.53	0.543	0.331	0.0242
								–				
	10	0.129	0.0087	0.0011	(0.014)	7.7	9.9	18.3	1.75	0.168	0.270	0.0077
	11	0.208	0.0153	0.0024	0.022	17.1	19.2	19.8	2.17	0.441	0.365	0.0146
	12	0.177	0.0225	0.0132	(0.015)	17.8	9.0	19.9	2.27	0.534	0.246	0.0127
	13	0.223	0.0182	0.0019	0.026	22.0	15.7	20.3	2.15	0.461	0.485	0.0105
	14	0.317	0.0294	0.0022	0.073	19.4	21.0	22.2	2.34	0.581	0.554	0.0242
	15	0.160	0.0068	0.0017	0.020	8.8	18.7	———	1.26	0.203	0.271	0.0243
								–				
	16	0.196	0.0368	0.0124	0.090	11.4	8.1	13.1	2.52	0.510	0.300	0.0088
	17	0.115	———	0.0168	0.051	18.5	14.5	14.3	3.02	0.867	0.395	0.0098
	18	0.190	0.0082	0.0012	0.020	15.1	13.1	18.6	1.69	0.223	0.373	0.0154
	19	0.194	0.0247	0.0054	0.035	15.2	18.7	18.0	2.36	0.532	0.295	0.0181
	20	0.299	0.0202	0.0043	0.024	21.9	15.6	22.5	2.28	0.456	0.607	0.0152
	21	0.154	0.0136	0.0012	0.018	13.4	9.4	18.6	1.92	0.227	0.262	0.0108
	22	0.348	0.0317	0.0016	(0.017)	20.6	35.8	22.4	2.37	0.459	0.584	0.0185
	23	0.216	0.0215	0.0126	(0.017)	13.1	11.3	14.1	2.21	0.522	0.327	0.0155
	24	0.124	0.0141	0.0055	———	10.1	15.4	18.2	1.48	0.467	0.208	0.0190

KM	**1**	0.206	0.0082	0.0034	0.045	11.3	13.5	15.3	2.73	0.710	0.274	0.0205
	2	0.225	(0.0013)	0.0102	0.078	8.3	2.1	15.7	1.43	0.191	0.389	0.0059
	3	0.156	0.0056	0.0051	0.092	10.4	7.5	20.2	2.22	0.642	0.209	0.0116
	4	0.159	0.0047	0.0058	0.090	11.0	7.2	19.1	1.87	0.381	0.285	0.0096
	5	0.244	0.0084	0.0041	0.075	13.0	10.2	18.8	1.80	0.254	0.498	0.0178
	6	0.230	(0.0016)	0.0100	0.104	8.0	2.0	15.6	1.46	0.206	0.414	0.0076
	7	0.100	———	0.0114	0.062	7.2	3.5	13.6	1.88	0.418	0.275	0.0114
	8	0.138	(0.0017)	0.0019	0.067	9.5	4.8	20.5	1.44	0.188	0.249	0.0093
	9	0.150	0.0050	0.0036	0.113	9.5	8.9	19.0	2.36	0.425	0.394	0.0145
	10	0.098	0.0093	———	0.103	13.0	3.4	13.8	2.06	0.589	0.401	0.0073
	11	0.176	0.0032	0.0086	0.063	8.3	4.4	14.3	1.70	0.212	0.332	0.0056
	12	0.136	0.0064	0.0029	0.072	8.6	5.5	19.1	2.33	0.225	0.254	0.0091
	13	0.109	0.0231	0.0123	0.067	7.4	3.4	14.6	1.97	0.410	0.320	0.0117
	14	0.119	———	0.0025	0.074	8.0	3.8	17.3	1.32	0.174	0.241	0.0079
	15	0.217	< 0.0007	0.0083	0.138	7.7	2.0	14.6	1.38	0.182	0.388	0.0067
	16	0.153	0.0031	0.0050	0.097	9.8	7.0	19.6	1.84	0.556	0.143	0.0111
	17	0.110	0.0105	0.0108	0.037	11.4	5.0	12.3	3.39	1.057	0.207	0.0136
	18	0.196	0.0067	0.0026	0.040	8.5	5.1	18.5	1.98	0.125	0.452	0.0107
	19	0.152	0.0070	0.0052	0.052	8.3	5.7	16.7	2.29	0.428	0.272	0.0090
	20	0.151	0.0045	0.0062	0.093	12.8	4.4	16.6	1.12	0.337	0.233	0.0157
	21	0.128	0.0083	< 0.0003	0.045	8.7	8.4	21.4	2.05	0.318	0.265	0.0093
	22	0.107	0.0067	0.0097	0.026	7.4	4.7	13.1	1.27	0.197	0.507	0.0112
	23	0.175	0.0069	0.0026	(0.006)	8.7	12.0	17.5	1.17	0.157	0.424	0.0101
	24	0.111	0.0033	0.0123	0.066	7.0	6.0	13.0	1.36	0.223	0.587	0.0146
	25	0.153	0.0104	(0.0007)	0.065	19.6	16.8	22.1	2.24	0.243	0.119	0.0210

KCK	**1**	0.194	0.0046	0.0053	———	23.0	13.8	24.2	2.08	0.434	0.278	0.0237
	2	0.143	(0.0022)	0.0028	0.027	7.2	4.2	18.9	1.74	0.210	0.319	0.0100
	3	0.137	(0.0018)	0.0116	0.034	7.1	3.9	14.9	1.67	0.248	0.162	0.0035
	4	0.086	0.0029	0.0089	(0.016)	8.0	11.8	12.9	3.05	0.609	0.271	0.0119
	5	0.134	0.0051	0.0049	(0.017)	7.5	19.3	18.1	1.47	0.171	0.233	0.0096
	6	0.087	(0.0010)	0.0091	0.022	7.8	4.1	11.8	3.00	0.571	0.262	0.0134
	7	0.142	0.0023	0.0139	0.063	7.5	4.0	14.9	1.74	0.259	0.169	0.0049
	8	0.180	0.0033	0.0030	0.036	7.5	7.8	16.1	1.55	0.295	0.338	0.0097
	9	0.192	———	(0.0010)	0.050	13.1	31.2	18.2	2.07	0.256	0.296	0.0213
	10	0.136	0.0097	0.0091	< 0.006	10.8	10.2	21.1	2.35	0.577	0.465	0.0119
	11	0.140	0.0030	0.0052	< 0.006	9.2	5.6	19.4	1.92	0.266	0.408	0.0135
	12	0.082	0.0042	0.0084	0.055	7.3	4.4	11.3	2.98	0.600	0.258	0.0122
	13	0.194	0.0053	0.0037	(0.008)	7.1	3.6	14.2	1.52	0.137	0.473	0.0050
	14	0.189	0.0047	0.0033	0.052	9.2	5.9	18.4	1.71	0.352	0.346	0.0108
	15	0.081	0.0029	0.0078	0.067	7.6	42.5	11.0	2.79	0.578	0.237	0.0120
	16	0.142	(0.0017)	0.0053	0.029	7.0	3.1	13.5	1.27	0.183	0.219	0.0061
	17	0.159	0.0032	0.0047	0.048	7.9	4.1	18.6	1.85	0.245	0.332	0.0075
	18	0.198	0.0058	0.0028	0.067	8.5	6.5	17.9	1.73	0.314	0.339	0.0117
	19	0.079	(0.0011)	0.0092	0.037	7.6	2.7	10.9	2.99	0.617	0.329	0.0108
	20	0.139	0.0090	0.0095	0.052	7.7	9.6	20.9	2.03	0.451	0.511	0.0132
	21	0.113	0.0085	0.0161	0.030	11.7	6.3	21.5	2.37	0.729	0.356	0.0115
	22	0.185	0.0034	0.0031	0.027	7.7	4.3	16.3	1.53	0.275	0.323	0.0089
LOQ*		0.0016	0.0022	0.00103	0.018	0.013	0.625	0.072	0.0095	0.025	0.033	0.0025

*LOQ: limitations of quantitation (= standard deviation (SD) x 10) of blank solutions were converted into rice sample abundance (mg/kg).

Values below LOQ and above LOD (limitation of detection) are shown in parentheses, and the inequality sign shows values below LOD.

Table. Concentrations (mg/kg) of 18 elements in 82 Sediments samples from five townships (DL, KM, TK) in Myanmar.

City	Location	UTM	Elevation(m)	Al	As	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	Pb	Sc	Sr	V	Zn
DL	47Q 0194988	1845398	2	2159	3.7	265	0.02	7.77	36.6	16.5	11172	971	3453	148	117	42.3	8.95	4854	12.1	26.4	45.7
				2127	3.4	181	0.02	7.25	36.2	16.9	12671	926	3251	126	105	39.0	8.28	28542	11.9	27.4	42.8
	47Q 0194976	1845239	0	2085	4.3	187	0.03	7.14	35.7	17.0	10622	810	2998	113	91	38.9	9.06	28083	11.2	27.3	42.9
				2065	8.5	189	0.02	7.06	36.5	17.1	11189	817	2969	115	95	38.4	9.01	29274	11.5	28.2	42.3
	47Q 0194109	1845134	4	2156	4.6	229	0.02	9.03	37.5	17.9	12088	1100	3405	174	120	45.4	9.76	30744	11.5	27.4	49.0
				2180		232		8.71	37.7	18.1	12578	1103	3374	196	119	44.8	9.25	30796	12.2	28.1	48.2
	47Q 0194116	1845145	3	2325		245		9.56	42.9	21.9	16290	1165	3504	199	131	48.6	10.16	37339	15.3	33.5	51.7
				2264		206		8.03	40.3	19.4	12673	1035	3069	152	112	42.7	9.39	33097	12.2	32.0	46.1
	47Q 0193696	1845036	-4	2371		305		11.00	43.2	20.0	13662	1176	4378	258	115	56.7	9.33	34005	14.2	29.5	54.4
				2338		275		10.31	39.0	19.6	12429	1001	3901	256	113	50.1	11.04	30045	12.5	27.8	50.1
	47Q 0194373	1845199	6	2169		437		8.26	32.2	14.6	9615	645	3010	196	56	39.5	6.75	22204	10.8	21.2	43.7
				2123		319		7.82	29.6	12.5	9174	646	2733	184	55	37.5	5.36	3702	9.7	19.4	39.7
	47Q 0195774	1843992	0	2090		210		6.72	29.4	11.3	7720	498	2579	128	42	34.0	5.93	3351	9.4	18.4	36.1
				2261		287		8.26	33.4	14.5	8949	620	3074	167	72	41.0	7.43	3955	10.9	21.7	44.9
	47Q 0195150	1846896	2	2295		316		10.36	43.5	21.8	13374	960	4521	239	134	52.0	13.12	32146	15.1	31.5	55.7
				2341		366		10.46	44.8	24.6	12737	877	4190	227	113	52.0	13.81	32057	15.0	32.8	54.2
	47Q 0195006	1846921	-1	2417		327		10.15	43.6	21.9	13658	955	4437	218	116	52.5	12.11	33717	15.9	32.3	54.9
				2383		239		10.19	42.0	20.6	12553	910	4161	192	89	52.2	11.81	32951	14.2	30.1	54.0
	47Q 0200594	1843254	5	2235		178		7.93	37.0	15.9	11420	712	2628	147	92	37.8	9.60	4943	10.7	27.7	42.5
				2334		221		8.57	35.3	15.1	10514	733	2797	201	108	39.2	10.04	27941	10.9	26.5	42.7
	47Q 0200559	1843194	4	2210		192		8.33	35.9	15.0	10901	603	2346	203	88	35.9	9.41	4582	10.1	26.7	36.6
				2103		225		7.10	32.8	12.5	9558	529	2179	160	104	30.6	7.74	4064	9.4	24.0	32.8
	47Q 0200578	1843140	3	2410		308		9.91	42.9	19.9	12916	1139	4055	223	111	52.8	11.76	34409	14.1	29.5	55.9
				2381		251		8.92	44.5	21.6	13331	1066	3555	177	90	48.1	12.47	35912	14.3	34.0	51.5
	47Q0190961	1849218	-1	2331		415		10.19	40.0	19.7	13711	1136	3870	326	119	50.0	11.35	33226	17.0	29.7	53.0
				2319		367		9.59	37.1	16.6	13299	1094	3548	332	125	48.3	8.38	31023	15.0	26.3	50.7
	47Q 0194741	1851529	0	2485		381		10.52	47.9	25.5	14798	1379	4873	213	193	58.0	14.16	38369	16.5	36.8	60.2
				2464		468		11.90	48.7	26.8	15784	1520	5214	307	228	60.0	14.83	38242	18.5	37.9	61.3
	47Q 0194713	1851541	-3	2489		349		11.18	48.3	25.5	14537	1367	5052	230	131	58.7	13.87	37794	15.7	35.5	62.4
				2482		341		10.93	46.9	24.7	14427	1266	5109	225	139	57.8	13.13	36508	15.5	34.5	63.0

Table. Concentrations (mg/kg) of 18 elements in 82 Sediments samples from five townships (DL, KM, TK) in Myanmar.

City	Sampling	Location	UTM	Elevation(m)	Wt(g)	Volume(ml)	Al	As	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	Pb	Sc	Sr	V	Zn
KM	16A	47Q 0197580	1828791	-1	1.01	52.5	2346	3.5	343	0.02	9.83	38.1	17.1	11854	1351	4700	214	108	51.8	8.74	4770	13.6	26.1	54.3
	16B				1.07	52.1	2496	4.2	440	0.03	10.69	41.8	20.8	14236	1480	5070	233	152	55.9	11.28	31559	16.1	30.6	58.1
	17A	47Q 0197823	1828789	0	1.01	54.0	2315	5.8	468	0.02	9.99	39.8	20.5	12348	1395	4946	215	167	52.8	9.98	29639	15.5	29.2	54.4
	17B				1.00	53.6	2434	5.5	483	0.02	11.25	44.2	21.5	13920	1631	5519	230	204	59.8	11.58	33716	16.5	31.1	62.3
	18A	47Q 0196055	1825592	2	1.01	50.7	2413	7.3	375	0.02	9.44	42.4	19.3	13674	1157	3618	200	231	47.7	10.98	33943	15.5	31.7	48.3
	18B				1.10	53.5	2270		268		8.52	36.3	14.9	12402	1020	3056	188	182	41.5	6.90	4988	12.9	26.5	41.7
	19A	47Q 0196111	1825532	1	1.03	52.4	2273		279		9.15	39.8	17.3	12635	1017	3386	191	221	45.2	9.72	5321	13.9	29.3	44.4
	19B				1.11	52.1	2404		325		10.04	42.5	19.8	13112	1166	3838	204	285	49.5	11.20	33850	15.2	31.3	49.8
	20A	47Q 0195071	1823082	3	1.01	50.8	2402		363		9.57	42.6	21.6	14100	1307	4786	186	268	52.8	11.91	33782	16.4	31.9	52.8
	20B				1.01	51.9	2414		431		11.29	47.1	27.3	16765	1404	5150	256	254	57.5	15.88	37507	19.0	36.5	56.6
	21A	47Q 0195083	1822326	3	1.01	52.6	2368		551		9.82	42.7	25.6	14358	1783	5406	243	343	53.3	16.57	31773	15.3	31.9	68.6
	21B				1.03	53.1	2361		473		10.19	41.8	22.6	13323	1495	5212	305	155	52.5	13.75	30646	14.2	29.0	59.3
	22A	47Q 0199138	1827337	2	1.02	50.2	2596		647		13.07	56.2	28.8	19026	1810	6406	335	411	68.8	16.02	41477	21.4	38.3	67.1
	22B				1.01	51.2	2576		504		13.36	50.0	24.8	15391	1537	6041	310	240	67.2	14.58	37999	17.6	34.7	67.3
	23A	47Q 0199038	1827267	5	1.01	50.8	2547		815		12.99	47.3	23.8	15118	1526	6338		416	65.2	12.06	34645	19.8	33.2	65.7
	23B				1.03	50.5	2576		852		12.72	47.3	24.1	15409	1670	6522			63.9	11.73	35220	20.7	33.4	67.7
	24A	47Q 0189764	1838913	-1	1.02	51.3	2444		430		11.74	43.5	20.6	14439	1150	4743	314	154	57.6	10.22	32481	22.1	30.6	56.7
	24B				1.02	49.1	2515		380		10.80	45.2	21.7	16027	1199	3771	289	242	53.7	10.52	36403	26.9	33.1	54.7
	25A	47Q 0189874	1838756	0	1.01	52.9	2494		579		11.48	49.0	30.1	16797	1411	4330	184	568	62.4	14.72	36343	38.5	38.4	67.9
	25B				1.00	52.0	2483		609		11.46	48.6	27.5	16921	1522	4522	191	412	63.3	14.06	39672	46.7	37.0	68.5
	26A	47Q 0189740	1839919	3	1.03	52.3	2483		519		9.90	50.2	26.8	15273	1435	4364	249	298	55.4	13.58	39081	33.5	38.2	61.7
	26B				1.01	51.1	2562		539		11.54	53.0	28.5	16353	1566	4816	258	367	62.1	15.03	40943	36.1	39.1	67.9
	27A	47Q 0189743	1839977	-3	1.01	53.1	2473		536		11.24	47.2	26.4	14987	1413	4413	247	343	56.9	13.72	35519	32.9	35.4	65.7
	27B				1.03	52.9	2481		426		8.46	43.8	19.7	12163	1491	4362	172	299	51.7	10.88	5864	30.5	27.2	62.4

Table. Concentrations (mg/kg) of 18 elements in 82 Sediments samples from five townships (DL, KM, TK) in Myanmar.

City	Sampling	Location	UTM	Elevation(m)	Wt(g)	Volume(ml)	Al	As	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	Pb	Sc	Sr	V	Zn
TK	28A	46Q 0812994	1911771	9	1.08	51.9	2048	1.25	198	0.03	5.41	25.0	7.6	5829	281	1339	113	41	26.4	8.57	2947	5.9	19.4	33.2
	28B				1.02	53.0	1995	1.01	308	0.04	5.73	24.5	7.6	6072	272	1336	168	38	26.7	7.84	2929	7.2	19.4	32.3
	29A	46Q 0812860	1911738	8	1.02	53.5	2050	0.67	306	0.02	5.07	26.6	8.5	5388	309	1479	115	40	28.5	9.89	2844	6.5	21.2	36.8
	29B				1.02	53.8	1970	1.25	205	0.02	5.68	25.7	7.8	6468	269	1352	148	43	26.9	8.44	2978	6.3	21.1	30.4
	30A	46Q 0812765	192295	9	1.03	50.9	3780	1.52	113	0.02	2.99	15.2	3.4	5962	256	714	81	21	13.8	4.79	1323	3.3	11.5	19.3
	30B				1.10	51.9	5140		138		3.96	17.9	4.6	7875	190	912	96	22	18.7	5.05	1924	4.4	14.3	23.1
	31A	46Q 0812759	1912220	5	1.02	52.5	1824		178		3.86	18.4	4.7	8031	199	913	118	28	18.4	5.01	1906	4.9	14.8	22.1
	31B				1.04	53.0	2252		310		9.30	29.9	9.0	9722	419	1837		45	35.0	8.41	3611	9.3	24.5	40.8
	32A	46Q 0813146	1913081	10	1.03	51.2	1881		159		3.57	21.5	5.7	4958	222	744	90	45	19.2	6.29	2323	5.1	20.2	22.2
	32B				1.08	51.2	2240		69		1.48	12.1	2.3	3915	85	311	28	22	7.9	3.25	946	2.2	11.1	9.4
	33A	46Q 0812937	1913059	11	1.02	52.0	1889		109		5.28	22.3	5.3	6198	206	729	119	29	19.9	6.62	2459	5.5	19.6	23.1
	33B				1.03	53.0	1919		119		4.92	24.0	6.0	5988	230	830	121	31	21.6	7.30	2779	5.6	21.2	24.4
	34A	46Q 0814579	1915473	13	1.09	52.7	3790		141		1.88	13.4	3.0	4567	172		37	38	14.3	4.34	1105	3.2	11.2	16.6
	34B				1.02	53.6	3208		97		1.99	12.4	2.7	4592	134	731	50	23	13.4	3.95	1016	2.9	10.5	15.2
	35A	46Q 0814589	1915426	10	1.14	51.9	3899		127		2.10	12.7	2.8	4857	145	539	65	28	13.7	4.04	1046	3.5	9.8	16.3
	35B				1.02	52.9	3076		101		1.91	12.1	2.5	4314	119	711	50	25	12.5	3.93	938	3.0	9.4	14.7
	36A	46Q 0812943	1910642	6	1.09	52.4	3935		231		2.65	14.6	3.7	6465	186	959	96	27	12.4	5.86	1180	4.1	11.8	20.9
	36B				1.05	52.7	1734		69		1.15	7.7	1.4	2757	84	682	26	14	5.9	3.28	437	1.7	6.0	9.7
	37A	46Q 0812957	1910624	7	1.02	52.7	2293		137		1.54	10.1	2.4	3239	103	317	45	17	8.3	3.62	821	2.6	7.4	14.4
	37B				1.04	52.8	2971		165		1.76	12.6	3.1	3571	185	437	45	22	10.2	4.15	1047	3.2	8.4	18.3
	38A	46Q 0812245	1910817	3	1.05	52.7	1771		240		4.43	21.6	5.3	4835	188	558	104	28	16.8	6.27	2271	4.8	16.8	27.5
	38B				1.08	51.9	3075		100		2.85	16.5	3.2	6155	115	725	55	20	10.7	4.14	1464	3.1	13.2	17.1
	39A	46Q 0812130	1910790	1	1.07	51.8	1885		170		5.75	26.3	6.3	5715	223	483	125	48	21.1	7.07	2763	5.2	21.4	29.2
	39B				1.06	52.1	1919		165		5.69	26.7	6.2	5789	233	931	127	45	21.6	6.93	2912	5.3	21.9	28.4
	40A	46Q 0810092	1910041	2	1.02	52.1	2342		550		10.02	44.7	19.2	11506	402	979	256	90	49.6	9.71	6053	12.6	35.9	51.5
	40B				1.02	52.8	2340		674		9.80	47.5	21.7	12395	409	2376	230	90	50.0	11.23	6560	14.6	41.5	51.3
	41A	46Q 0810119	1910107	1	1.03	52.9	2379		613		11.79	47.7	25.6	13393	399	2462	324	117	52.5	15.75	38501	14.6	45.0	55.2
	41B				1.02	53.4	2371		454		10.64	44.9	20.5	11479	400	2502	229	78	52.2	10.86	6577	12.4	37.3	54.8

Table. Concentrations (mg/kg) of 12 elements in 23 water samples from three townships (DL, KM, TK) in Myanmar

	Location	UTM	Elevation(m)	Sample na	Al(ppm)	Cr	Mn	Fe	Co	Ni	Cu	Zn	Se	As	Mo	Cd	Pb
DL	47Q 0195031	1845326	3	A	0.005	0.002	0.0004	0.0073	0.00009		0.0005	0.0007	0.001	0.0011	0.0012	0.00003	0.00007
	47Q 0194125	1845136	1	B-river	2.74	0.009	0.0056	1.6116	0.00047	0.003	0.0042	0.0038	0.006	0.0012	0.0009	0.00000	0.00063
	47Q 0194125	1845136	1	C-pond		0.002	0.0008	0.0094	0.00006		0.0003	0.0014	0.004	0.0012	0.0005	0.00002	0.00008
	47Q 0193692	1845006	-4	D	0.279	0.002	0.1227	0.1507	0.00023	0.007	0.0034	0.0083	0.007	0.0016	0.0005	0.00006	0.00016
	47Q 0193692	1845006	1	E	0.093	0.003	0.0007	0.0725	0.00006		0.0014	0.0023	0.004	0.0008	0.0003	0.00002	0.00008
	47Q 0195718	1843908		F	0.040	0.001	0.0010	0.0528	0.00008		0.0010	0.0011		0.0012	0.0003	0.00005	0.00007
				G-pond	0.303	0.002	0.0029	0.2391	0.00013		0.0012	0.0012		0.0010	0.0004	0.00002	0.00011
				H	0.989	0.003	0.0032	0.7018	0.00021		0.0017	0.0026		0.0014	0.0003	0.00001	0.00041
				I	0.002	0.003	0.0020	0.0122	0.00006		0.0015	0.0012	0.002	0.0032	0.0013	0.00004	0.00006
KM	47Q 0194682	1851613	0	J	0.043	0.001	0.0005	0.0473	0.00006		0.0011	0.0004	0.006	0.0019	0.0004	0.00002	0.00004
	47Q 0185736	1835014	-3	K		0.003	0.0002	0.0033	0.00009		0.0004	0.0004	0.001	0.0009	0.0002	0.00003	0.00003
	47Q 0195091	1822305	4	L-1-river	0.019	0.006	0.0002	0.0112	0.00007		0.0020	0.0016	0.008	0.0017	0.0005	0.00000	0.00004
	47Q 0189811	1838922	4	L-2-using	0.003	0.004	0.0002	0.0069	0.00004		0.0003	0.0012	0.001	0.0014	0.0002	0.00001	0.00004
				M	0.007	0.002	0.0004	0.0048	0.00002		0.0009	0.0006		0.0031	0.0003	0.00000	0.00021
				N	0.005	0.004	0.0001	0.0027	0.00001			0.0006	0.002	0.0041	0.0008	0.000000	0.00002
				O	0.002	0.002	0.2945	0.0066	0.00002		0.0002	0.0006	0.002	0.0003	0.0002	0.00001	0.00003
TK	46Q 0813018	1911703	13	P	0.003	0.000	0.0017	0.1159	0.00002		0.0001		0.002	0.0004	0.0002	0.00003	0.00002
	46Q 0813000	1912384	13	Q		0.000	0.0003	0.0050	0.00001		0.00001	0.0003	0.004	0.0003	0.0004	0.00001	0.00002
	46Q 0813189	1913082	14	R		0.001	0.00003	0.0139	0.00001		0.00003	0.0001	0.009	0.0004	0.0001	0.00000	0.00002
	46Q 0814315	1915548	9	S		0.003	0.0462	0.0021	0.00198			0.0011	0.002	0.0001	0.00003	0.00003	0.00004
	46Q 0812987	1910610	5	T	0.015	0.003	0.0009	0.9729	0.00008		0.0005	0.0027	0.004	0.0016	0.0002	0.00003	0.00375
	46Q 0812204	1910827	3	U		0.001	0.0002	0.0046	0.00001		0.0001	0.0004	0.001	0.0016	0.0001	0.00001	0.00003
	46Q 0810025	1910004	3	V		0.002	0.0035	0.0042	0.00001			0.0001	0.001	0.0021	0.0001	0.000000	0.00002