

## MONITORING ARSENIC (As) SPECIES, HEAVY METALS CONTAMINANT (Fe, Mn, Zn) and NUTRIENT (SULFATE, NITRATE AND CHLORIDE) IN YONGSAN LAKE, MOKPO, SOUTH KOREA

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### Abstract

*Contaminant levels (As, Fe, Mn and Zn) as a function of depth in Yeongsan Lake, South Korea, was investigated. The measurement of arsenic speciation, iron, manganese, and zinc level was investigated to provide information about the amount of those heavy metals in varying depth during fall turn-over session. Several anion concentrations such as chloride, nitrate and sulfate were also measured as it is consider as the macro and micro nutrients for algae and other micro-plantation on freshwater. In order to understand the characteristics of lake water, several water parameters, such as Dissolved Oxygen, pH, temperature, Salinity, conductivity etc, were measured on site. Based on the result, the wind plays an important role in turbulence mixing the lake water and stratification generated during summer disappeared during the fall season. Thus the heavy metals and nutrients level observed was relatively constant along the depth in shallower lake (Sampling Site 1). However, this occurrence was not observed in deeper Lake (Sampling Site 2), the anoxic zone was still observed below the 15 meters from the surface, thus it is lead to the higher As(III) as comparison with As(V) species in the ambient water close the bottom area. Overall the arsenic species, heavy metals and other anions level were under the maximum contaminant level (MCL) permit for drinking water based on the the USEPA regulation. Hence, this lake is meeting the criteria of water quality Class I, which is comply the criteria of the potential source of drinking water supply.*

**Keywords:** arsenic speciation, heavy metal monitoring

### Abstrak

Level dari kontaminan (As, Fe, Mn and Zn) sebagai fungsi dari kedalaman telah diinvestigasi di Danau Yeongsan, Korea Selatan. Pengukuran dari kontaminan tersebut dilakukan untuk memberikan informasi kuantitatif mengenai tingkat kontaminasi dari logam berat tersebut sebagai fungsi dari kedalaman danau di saat pergantian musim dari musim panas ke musim gugur. Beberapa konsentrasi anion, seperti klorida, nitrat dan sulfat telah diukur pula pada studi minotoring ini, sebagaimana anion-anion tersebut merupakan mikro dan makro nutrient yang cukup penting bagi alga ataupun tumbuhan air lainnya. Untuk mengetahui karakteristik dari danau tersebut pada saat pengambilan sampel, beberapa parameter seperti Oksigen terlarut, salinitas, konduktifitas, juga diukur secara *in situ*. Berdasarkan hasil yang diperoleh, dapat diketahui bahwa, untuk danau yang relative dangkal. Angin pergantian musim berpengaruh sangat besar pada stratifikasi yang terjadi di musim panas. Pengaruh angin musim gugur menyebabkan pencampuran secara vertical pada air danau yang dangkal sehingga konsentrasi dari beberapa logam berat yang diamati relatif konstan. Namun, hal ini tidak berlaku pada danau yang lebih dalam, pada kedalaman tertentu, masih terlihat adanya zona anoxic, yaitu dibawah 15 meter dari permukaan danau. Secara umum, konsentrasi logam berat maupun nutrients di Danau Yeongsan ini tidak melebihi batas maksimum kontaminan berdasarkan peraturan USEPA untuk air minum, sehingga air pada danau ini dapat dimasukkan pada katogoru air kelas I yang berpotensi sebagai sumber atau bahan baku untuk air minum.

**Kata kunci :** spesiasi arsenic, monitoring logam berat

## Introduction

Surface water and groundwater are the main water resource for human being. However, the level of metal pollution in water was elevated by human activity [1]. Heavy metal contamination of natural water is a major problem in industrialized areas, and it can cause a detrimental effect on environment and finally on human health [2]. Arsenic is categorized as a metalloide which has been highlighted as one of world-wide problems in recent years because of its toxicity [3]. Arsenic is the twentieth most abundant element in the earth's crust [4]. It is release into the environment from natural processes such as weathering reactions, microbiological activity, and volcanic emissions as well as anthropogenic activities including gold and silver mining, metal and alloy industries, petroleum refining, pharmaceutical and chemical manufacturing; burning of fossil fuels, and waste incineration [5]. Arsenic is widely distributed in nature and principally exists in the form of inorganic or organic compounds. The toxicity of inorganic arsenic depends on its presence species. Arsenite is more toxic than arsenate [6].

Inorganic arsenic exists as  $\text{As}^{5+}$ ,  $\text{As}^{3+}$ ,  $\text{As}^0$ , and  $\text{As}^{3-}$  in nature. It can form hydrides ( $\text{AsH}_3$ ), acids ( $\text{H}_3\text{AsO}_3$ ,  $\text{H}_2\text{AsO}_3^-$ ,  $\text{HAsO}_3^{2-}$ ,  $\text{H}_3\text{AsO}_4$ ,  $\text{H}_2\text{AsO}_4^-$ ,  $\text{HAsO}_4^{2-}$ ) and halides (gas  $\text{AsF}_4$ , solid  $\text{AsBr}_3$ , and  $\text{AsI}_3$ ). It can also form chlorine and methyl arsenic. Arsenic can also form bands with organic sulfur, nitrogen and carbon. As(III) and As(V) can also be formed by dissolving organic matter in natural environment. The most common arsenic compounds in groundwater and surface water are As(III) or As(V) [6]

Recent study for geochemical behavior of arsenic in surface water [7] showed that the behavior of arsenic diverged most strikingly upon the development of anoxic conditions. It was observed that arsenic concentrations increased more at sediments than in the shallower depths. It suggested that anoxic zone of the surface water depth would provide difference of arsenic species level. It is also observed that arsenic species present in varying depth affected by the presence of iron, manganese, phosphate, sulfides and other oxidizing metals that dissolved in organic matter which is naturally present in environment. Since surface water is the main source of water for daily activity, it is possible that arsenic enters to the human body easily. Hence, it is readily poisoning human body even at very low level. The arsenic level of several water depths must be quantified in order to know contamination degree for each depth, also to determine zoning area of the depth.

On the other hand, the Korean Government through their state of water environmental department has been successfully assigned about 87% of Korean freshwater quality into class II and III during the first period of monitoring since 1997 to 2007. However, Korea slightly revised its green vision and targeting about the similar percentage of their freshwater into class I and class II by conducting strict freshwater management and monitoring over decade, start from 2007 to 2017. Thus, the contaminant levels monitoring was conduct seasonally on the four major basin in South Korea (Han, Nakdong, Geum and Yongsan) and also include 114 watershed all over the nation.

Yongsan lake was located on Mokpo, Jeolla-province, South Korea. It is one of the major basin in South Korea, especially on the Southern region of the Korea Peninsula. This lake was a man-made lake that located on the Yeongsan River. The river was surrounding by three large cities on its middle stream region, thus it possibly to be the pathway to discharge the anthropogenic sewage. Additionally, on the lower stream there are several major industry such as the assembly of electronical device industries, which is believe to contribute the heavy metal levels on the Yongsan Lake.

Since Yeongsan Lake is one of the important freshwater resources in south-west part of South Korea. It is very important to monitor its water quality and heavy metal levels on this Lake. The main

purpose of this study was observing the different arsenic species level in various depths of Yeonsan Lake, since the electronic devices assembly industries may discharge arsenic in to the water body and sink to the estuarine, moreover the different arsenic species has differ toxicity level on human body. The iron, zinc, manganese, nitrate, chloride and sulfate was also reported on this study. These results will be able to indicate the arsenic species and several metals behavior on Yeongsan Lake during the turn-over season. In addition, the information gathered from this studied were used to complete the data set of monitoring the nutrients and heavy metal levels in Yongsan Lake, South Korea.

## Materials and Methods

### Surface Water Sample Collection

Surface water collected from Yeongsan Lake, Mokpo, South Korea on early fall season 2011. Water samples were taken from two different sites ( $34^{\circ}47'05$  N –  $126^{\circ}27'03$  E, and  $34^{\circ}47'25$  N –  $126^{\circ}31'10$  E) which can be seen on the sampling map on Fig. 1. Water samples collected using general water sampler from several different depths. The first sampling site was represent the Yeongsan river region, the samples were taken 0.5 meters (consider as surface freshwater), 3 meters and 6 meters from the surface. The second sampling site was represent the man-made Lake region, which is located on the river mouth area or estuarine. It has deeper depth thus the sample taken from 0.5 meters (surface), 13 meters, 14 meters and 19 meters below the surface.

Direct measurement of several parameters such as DO, pH, temperature, salinity, ORP and TDS was conducted in site using portable devices. The water sample was divided into raw water sample and soluble water sample. Soluble water sample was passed through  $0.45 \mu\text{m}$  membrane equipped with peristaltic pump. Arsenic speciation conducted immediately after taking samples. Arsenic speciation cartridge and arsenic speciation resin were used for arsenic speciation. These two different speciation method were used to crosscheck the result. Approximately, 50 ml samples were collected in tube at each sample point and were stored at  $5^{\circ}\text{C}$  before the measurement of concentrations.

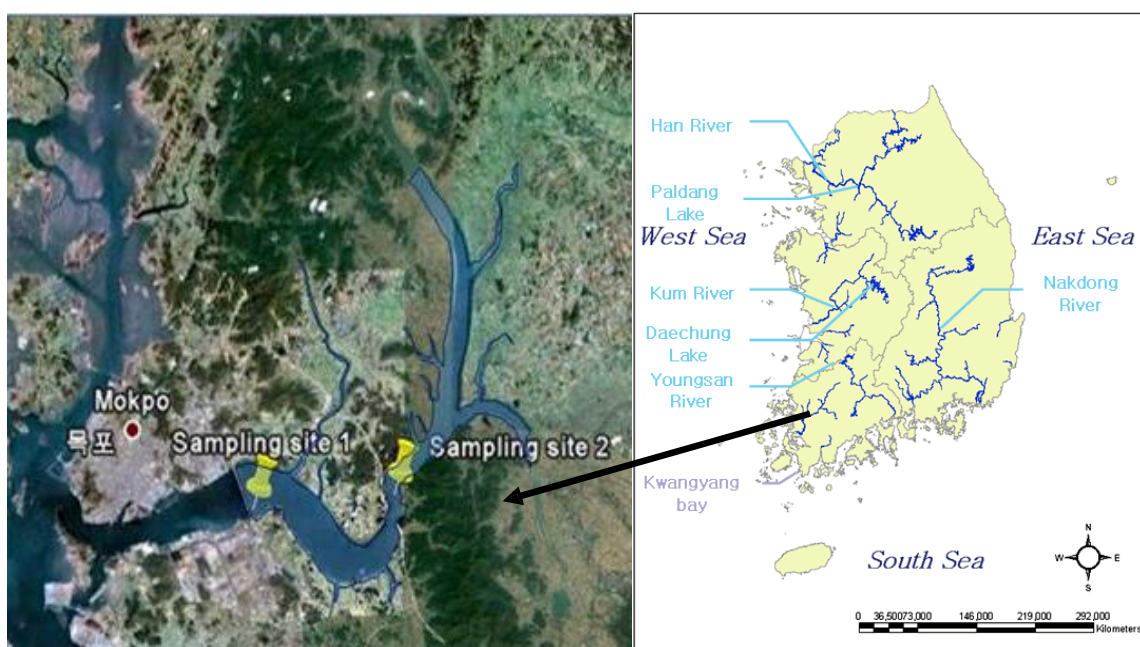


Figure 1. Sampling map: Yeongsan Lake, Mokpo, South Korea [8].

### Analytical Method

Arsenic and other heavy metals such as Fe, Mg, and Zn were detected using ICP-MS (Agilent 7200 series). All water samples were acidified using 2% HNO<sub>3</sub>. Standard solutions (1, 5, 10, 20, 50, and 100) were prepared by diluting 10 mg/L of ICP-standard solution with deionized-water contain 2% of HNO<sub>3</sub>. Anion analysis was conducted by ion chromatography (IC-Dionex 2000). Samples were prepared by making 10 times dilution of both raw samples and soluble samples, from each site. Standard solution were prepared by dilute 100 mg/L of stock solution (Dionex seven anion standard) into several concentration (0.1; 0.5; 1; 5; 10 mg/L).

### Result and Discussion

#### Surface Water Parameter Measurement

Table 1. Yeongsan Lake Water Parameter on Site 1

Depth	DO		pH	Temp (°C)	Conductivity (µS)	Salinity (ppt)	ORP (mV)	TDS (mg/L)
	(%)	(mg/L)						
Surface	79.8	6.56	7.28	18.9	547	0.3	166	300
3 m	51.7	4.90	6.84	18.8	522	0.3	132	266
6 m	62	5.68	7.24	19.7	562	0.3	129	295

Table 2. Yeongsan Lake Water Parameter on Site 2

Depth	DO		pH	Temp (°C)	Conductivity (µS)	Salinity (ppt)	ORP (mV)	TDS (mg/L)
	(%)	(mg/L)						
Surface	35.8	3.52	7.46	18.2	366.7	0.2	205	205
13 m	39.2	3.36	7.34	18.3	393.1	0.2	190	216
14 m	37.0	3.48	7.38	18.2	363.2	0.2	152	199.4
19 m	50.5	4.54	7.34	19.0	697	0.4	153	366

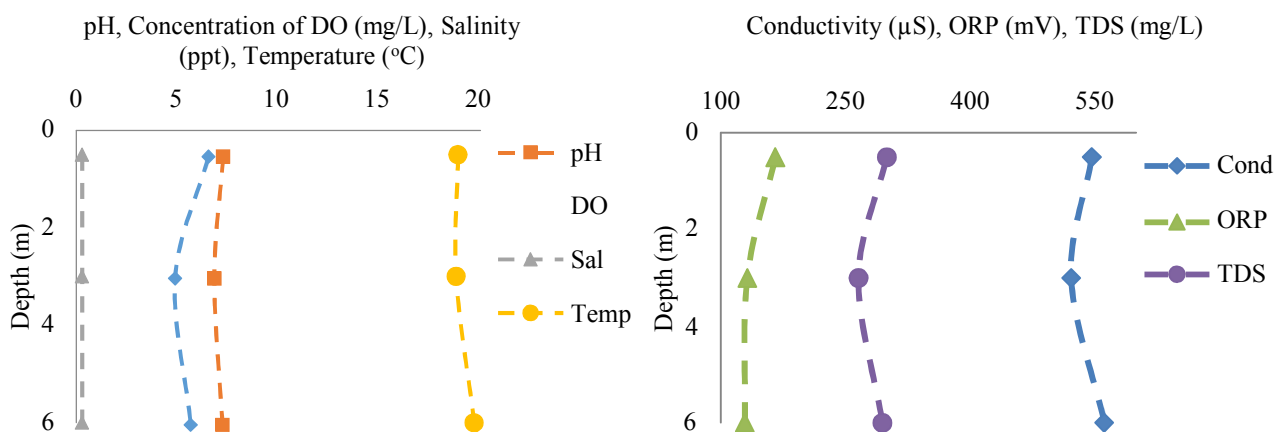


Figure 2. Profile of pH, DO, salinity, temperature, Conductivity, ORP and TDS on Site 1.

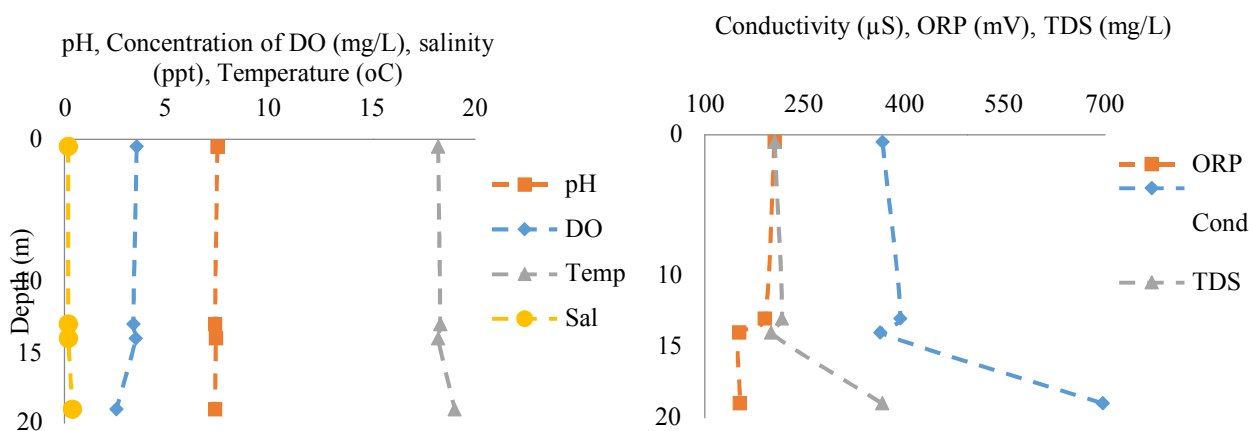


Figure 3. Profile of pH, DO, salinity, temperature, Conductivity, ORP and TDS on Site 2

### The general water quality assessment

The general water quality measured for Site 1 and Site 2, respectively, was shown in Table 2 and Table 3, and its vertical profile shows in Fig. 2 and Fig. 3. The Dissolved oxygen (DO) level can be an indication of how polluted the water is and how well the water can support aquatic plant and animal life. In General, a higher dissolved oxygen level indicates better water quality and it is affected by water temperature, pressure and salinity. Theoretically, the DO level may change dramatically with depth of the lake. Changes in lake water levels, volumes of inflows and outflows, and presence of ice cover can be caused by natural variation in DO levels. Oxygen production occurs around the surface of the lake, where sunlight used for photosynthesis. Oxygen consumption is greater near the bottom of the lake, where organic matter accumulates and decomposes. Hence, at the deeper lake, the DO levels decreased. Seasonal changes were also affected by DO levels in the lake. For the Site 1, the lake is shallow and sampling conducted on early autumn, which is the wind plays an important role in turbulent mixing of the water lake. Therefore, the DO concentration may be fairly consistent throughout the depth and it's also the case for the Site 2. Both sampling sites showing similar level of DO, which is around 3.5 – 6.5 mg/L. This DO level is categorized as poor level for supported living organism. The relatively poor DO level is related to the seasonal impacts. On early autumn, most of the freshwater micro-plantation are died and their decomposition is required large amount of oxygen consumption, and it is eventually lead to the low level of DO measurement on the water. Unfortunately, the biological oxygen demand (BOD) was not able to measure in situ. Thus, the quantification of oxygen utilization on this region was not able to evaluate quantitatively.

The pH is a measure of the acidity or alkalinity of a substance. It is important parameter to measure in water quality because it determines the solubility, biological availability of chemical constituents, and also heavy metals form in water. Similar to the DO concentrations, pH may change with depth in a lake due to changes in photosynthesis and other chemical reactions. However, lake water is complex and full of chemical natural-absorbent that prevents major changes in pH. The pH measurement in both sampling sites shows that there was only small change in pH. Due to its complexity, the water lake has buffering capacity which is able to resist change in pH. Change in pH is quickly modified by various chemical reactions, so little change in pH was measured in both sites.



Temperature is also important parameter because of its influence on water chemistry. The rate of chemical reactions generally increases at high temperature, which in turn affects to biological activity. Warm water holds less oxygen, so it may be saturated with oxygen but it still not contains enough oxygen

for survival of aquatic life. Some compounds are also more toxic to aquatic life at high temperatures. The most obvious reason for temperature change in lakes is the change in seasonal air temperature. Thermal stratification of lake water is developed during summer, and the surface water temperature will be differ from the deeper water temperature. The surface water is warmed by the sun, but the bottom of the lake remains cold. During fall session, the stratification is lost and the wind can turbulently mix the water. Therefore, there was little or even no changes in temperature measurement for each depth, either Site 1 or Site 2.

The conductivity is defined as the estimation amount of the total amount of dissolved ions in the water. The water conductivity strongly depends on temperature. At the Site 1, there was no significant change in conductivity for each depth. It means that the conductivity for each depth is almost the same due to the shallow part of the lake and the strong influence of the in fall turn-over session to mix up the lake water. However, for the Site 2, the significant change of conductivity was measured at 19 m depth from the surface. It reached up to 697  $\mu\text{S}$  while the surface region only 366.9  $\mu\text{S}$  (Table 2). This result indicated that the dissolved material (ion) is increased at the bottom of the lake, which can be caused by the diffusion of the bottom lake material to the ambient lake water. The similar case seen on the level of The Total Dissolved Solids (TDS).

The Salinity is related to the salt content in water. It is measured as total of all the salts dissolved in the water. The majority of the salt in natural water is sodium chloride, but there are other salts as well. Salinity was influenced by several factors such as evaporation or precipitation, temperature, organism, and also the large amount of river estuary. The salinity measured from lake water samples was almost similar in both sampling sites. However, in the deeper lake shows higher salinity, especially in Site 2 (Table 2). Higher salinity of near the bottom area on Site 2 can be caused by its sampling location, which is the lake is near the estuarine area, where the salinity instruction from the seaside may occur.

The Oxidation-reduction potential (ORP) indicates the relative capacity of a solution to oxidize or reduce. It is measured by the activity or strength of oxidizers and reducers in relation to their concentration. ORP shows the activity of oxidizing agent that is present in the water, which has indirect correlation with the presence of several metals and some kind of bacteria in water. Based on the measurement, the ORP tends to decrease along with increasing the depth. This is in agreement with the theoretical studies which stated that the deeper water depth the more anoxic condition it will be, and thus the redox potential decreased.

### ***The Arsenic and Heavy Metal level***

The arsenic speciation in this study was conducted using two speciation methods. The first method was using cation resin. It was applied to separate As(III) from total arsenic. Thus, by flowing the water samples through the cation resin, it can be assumed that the effluent water samples contained As(V). By Measuring the total As concentration on the unfiltered water samples, it can be calculate the amount of As(III) present on the sample that retained on the cation resin. The second method was using the arsenic speciation cartridge, which is able to absorb As(V) on the materials cartridge, and the effluent will be As(III). Since As(III) concentration in this sampling point was low, thus it is very challenging to detect such low concentration during the analysis. It is lead to the un stable detection result (the data not shown in this study). Based on those considetation, for further discussion, the arsenic data obtained from the cation resin speciation will be used.

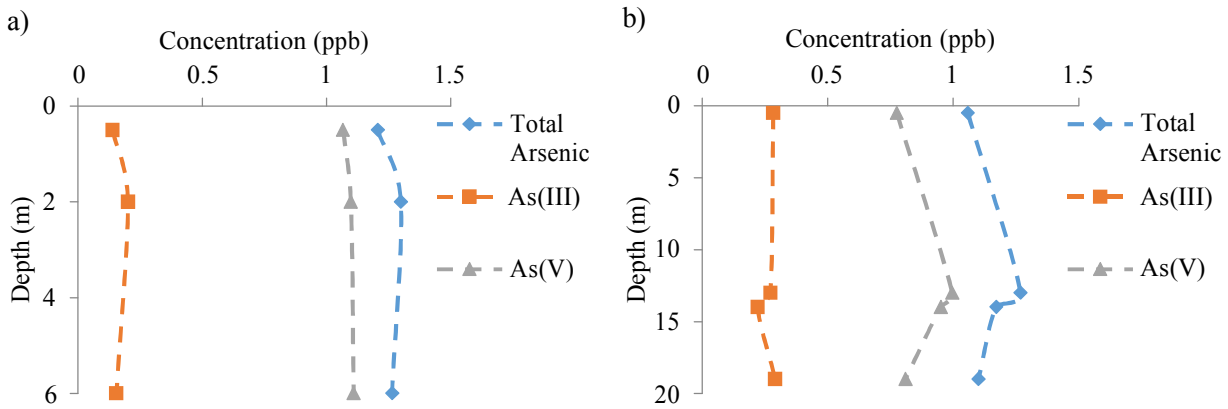


Figure 4. Profile of arsenic species concentration on Yeonsang Lake a) site 1 and b) site 2.

From the Fig. 4 either Site I and Site 2, it can be seen that most of the arsenic species exist as As(V) form as it is the more stable arsenic compound compare to As(III). Interestingly, on the Site 2 the increment of the As(III) concentration was observed in the deeper depth close to the bottom area (about 19 meters below the surface). This interesting occurrence can be explained by the lake stratification during the summer. Although, it is believed that the lake stratification would disappear along with the season changes, there is still a slight possibility that the stratification is still ongoing in the deeper lake region since the turbulence mixing from the surface may not reach the bottom area. Over 15 meters from the surface, it is likely nearby the anoxic zoning area where the decrement of dissolved oxygen and the Oxidation-reduction Potential were observed as shown in Figure 3. In the anoxic zoning area, most heavy metals were present in reductive form, hence the As(III) species tend to be higher in this point, in consequence, the As(V) concentration will be decreased.

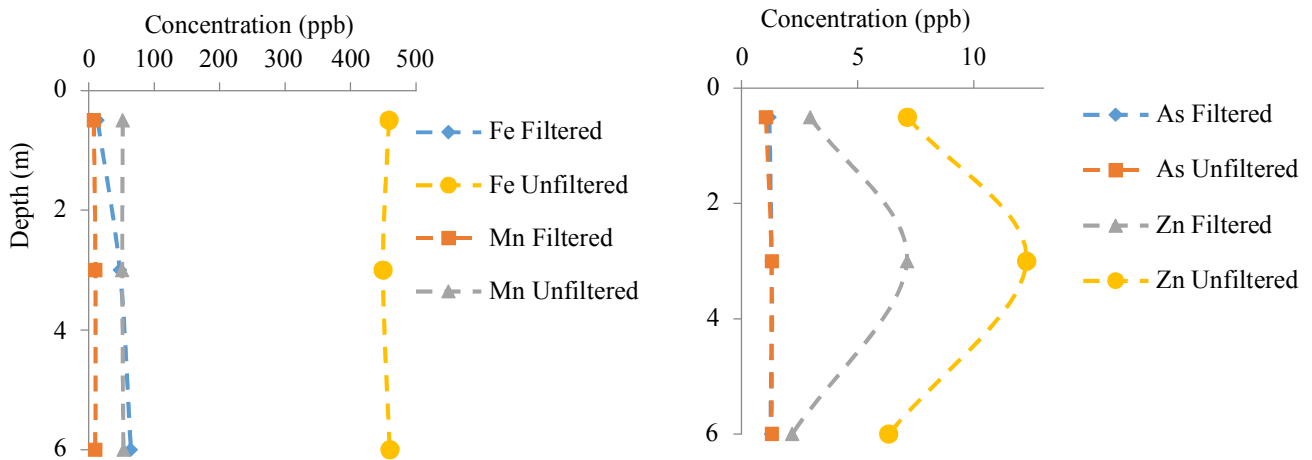


Figure 5. The vertical profile of Fe, Mn, As and Zn in Site 1

The vertical profile of Fe, Mn and As level in Fig. 5 shows that the concentrations of these metals in

both filtered or unfiltered samples on each depth were relatively constant. It is already known that the sampling Site 1 has shallower depth than sampling Site 2, and it is common that in turn-over season

the vertical profile of certain heavy metals relatively constant along the depth, as reported by Kamau *et al* [9]. However, the Zn level slightly increased in 3 meters below the surface, this occurrence was also observed by Kamau *et al* [9] that zinc in labile-fraction form mobilized in the water, not accumulated in the sediment. Therefore, the increasing level of zinc in certain depth can be explained by the suggestion that zinc mobilized mostly on the observed depth.

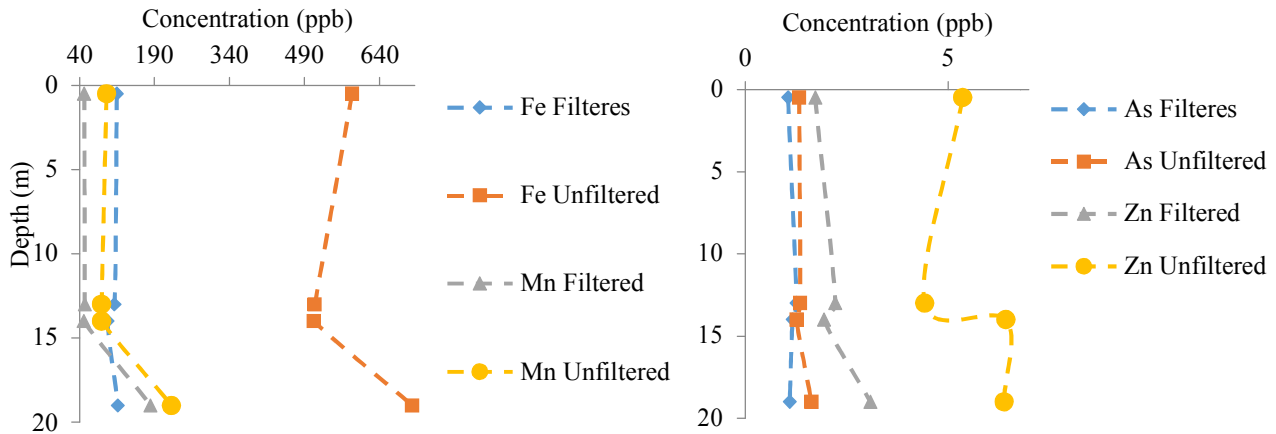


Figure 6. The Vertical profile of Fe, Mn, As and Zn in Site 2

The graph in the Figure 6 depicts the heavy metal level on several depths in sampling Site 2. Based on this vertical profile, There no significant changes in observed metal levels up to 13 meters below the surface. However, the deeper depth shows increasing level of most of observed metals. This occurrence can be explained by the sediment interaction with its ambient water. The last sampling point in Site 2 was 19 meters below the surface, which is close to the lake bottom area or lake sediment. It is commonly known that most of the heavy metals were deposited and accumulated in the sediment. The heavy metal and nutrients sinking on the sediment were also observed on previous study [10], [11]. In addition, the sinking event usually followed by the diffusion of certain material to the ambient water, thus the elevation of certain heavy metals or nutrients in certain depth close to the bottom area of the lake is commonly observed. This suggestion strongly support by the TDS data on this point (Site 2). The TDS was increased near the bottom area (Fig. 3), which is means there are higher amount on dissolved materials at this point.

Additionally, the maximum contaminant levels (MCL) of iron, manganese, arsenic and zinc by the USEPA for drinking water, were 0.3, 0.05, 0.01, and 5 mg/L, respectively. Since this Yeongsan Lake suggest to be the surface water that proposed as drinking water supply, it should be comply those USEPA criteria for drinking water, in particular, for its heavy metals content. Based on the data taken during this early fall season, It can be concluded that these heavy metals levels in Yeongsan Lake were under maximum level. In case of these heavy metals contain, the Yeonsan Lake is still comply the criteria of freshwater Class I for drinking water supply, despite surround by three big cities and several industries. For the past decades, this water reservoir successfully upraise from the water quality Class II into Class I. Based on these observation result in this study, the Yeongsan Lake is able to maintained

its water quality, thus it is still meet the criteria of drinking water supply.

### The Anion Analysis for nutrients assessment

Sulfate and nitrate consider as the source of mineral S and N for micro-plantation on fresh water. Thus, it's also essential to access these element level in order to know the availability of these nutrient during early fall season. Fig. 7 shows the vertical profile of sulfate, nitrate and chloride. For the Site

1, the vertical profile suggested that these anion levels tended to be constant along the depth. As discussed above, as the summer season has end, the lake stratification disappear and the wind has strong effect on the turbulent mixing of the lake water vertically. Thus, these anion level on the Site 1, which is known as shallow area will not differ from each depth. However, this is not likely the case for the Site 2. Since the sampling point relatively deep, which is up to 20 meters from the lake water surface. The significant increment was observed on the near bottom area (19 meters), especially in chloride and sulfate level. The chloride level was significantly increased, this is in agreement with the salinity data that shows significant increment on the bottom area of Site 2, as the salinity value is actually shows the amount of chloride.

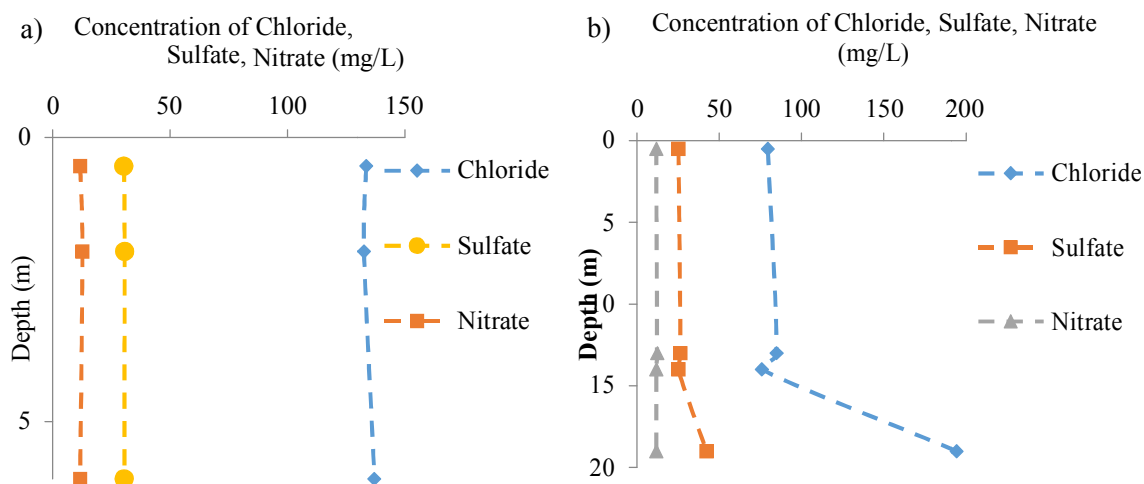


Figure 7. Chloride, sulfate and nitrate measurement in Yeongsan Lake  
 a) sampling site 1 and b) sampling site 2.

Sulfur is present in reduction form which is precipitated during its interaction with other elements. Oxygen was reduced with the depth, and some others heavy metals and nutrients were increased while the lake water was turbulently mixed with the wind. In this case, the oxidation-reduction process might be occurring. In the present of oxygen, sulfuric-precipitated compound might be oxidizing into sulfate form and leached out from the sediment by wind. This suggestion was supported by DO and also ORP level on site measurement. The DO levels of each depth were relatively constant and the ORP levels were not much different for each depth [12]. The Yeongsan Lake is one of the major water reservoir included in Class I, which play the key role as the drinking water supply in south-west Korea Peninsula, therefore from time to time it should be meet the criteria for drinking water resources. Refers to the USEPA regulation for drinking water, the maximum contaminant levels (MCL) of chloride, sulfate and nitrate respectively 250 mg/L, 250 mg/L, and 10 mg/L. Based the data above, these anion level on

Yeongsan Lake were under maximum level. Thus, it is comply the criteria of Class I freshwater reservoir.



## Conclusions

The water quality of the Yeonsang Lake as one of the major water resources in South Korea has been observed. Based on this study, the arsenic as the toxic contaminant is still below the maximum level that permitted for potential drinking water supply. Its levels along the depth relatively constant in the shallow lake, and its dominant species was As(V). Similarly, for the other heavy metals observed, Fe, Mn and Zn, it mostly shows constant level along the depth, especially for the Site I, the shallower sites. This occurrence strongly affected by the turn-over seasonal wind, which is the wind played an important role in turbulence mixing of the lake water and the stratification generated during summer disappeared during fall turn-over season. Interestingly, this behavior slightly differ for the deeper lake in Site 2, as the bottom lake still showing the present of summer stratification based on the DO and ORP data. Overall the observed arsenic, heavy metals and other anions level were under the maximum contaminant level (MCL) permit for drinking water by the USEPA. Thus, in general the Yeongsan lake is comply the water quality Class I that potential for drinking water supply.

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