

Investigation of the groundwater contamination around landfill where slaughtered animals were buried

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Abstract

This study was designed to investigate if there were groundwater contamination in 17 landfill where slaughtered animals were buried during the crisis of 2002 foot-and-mouth-disease (FMD) outbreaks in Gyeonggi province. From March to August 2005 groundwater was collected once a month from 17 sites, and examined with potential for hydrogen (pH), colour, turbidity, lead (Pb), arsenic (As), mercury (Hg), cadmium (Cd), copper (Cu), zinc (Zn), iron (Fe), manganese (Mn), aluminium (Al), nitrate-nitrogen (NO₃-N), ammonia-nitrogen (NH₃-N), microbial pathogen and *Escherichia* spp.

In the examination of NH₃-N which of the mean concentration was from not-detected(ND) to 0.05mg/l. The range of NO₃-N level was 0.3 - 24.1 mg/ℓ. However, groundwater from four sites was to go beyond the drinking water quality standard (DWQS), i.e., the mean concentration of those were 15.5 mg/ℓ (site 1), 20.7 mg/ℓ (site 9), 24.1 mg/ℓ (site 13) and 10.6 mg/ℓ (site 17). In the investigation of pH, colour and turbidity, all of the pH were below of DWQS (pH 5.8 - 6.6), but one site in color test and four sites in turbidity test were over the standard level. Among 9 metal ions examined, Mn was in excess of DWQS, and its concentration was 2.4 mg/ℓ. Pb, Cd, Hg and As were not traced. The contents of Cu, Zn, Fe and Al were ND - 0.22 mg/ℓ, 0.01 - 0.05 mg/ℓ, ND - 0.05mg/ℓ and 0.03 - 0.16 mg/ℓ, respectively.

Escherichiae spp were not identified, but bacterial colonies were detected at 3 groundwater including 2 sites over the DWQS at the level of 491 CFU/ml (site 4) and 217 CFU/ml (site 15).

Key words : Groundwater, Landfill, NO₃-N, NH₃-N

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Introduction

Water is the origin of life and a basic and essential element for our healthy life. About 40% of global populations are now suffered from chronic stress of water, and it is forecast that even 2.5 billion people will face lack of water in coming 2025. Besides, Korea is already classified as a water-stressed country¹⁾. In order to relieve this lack of water, the development of groundwater is widely used, because it is advantageous in supplying water most easily and stably at affordable cost.

However, water quality becomes worse rapidly due to reckless development of undergroundwater and environmental pollutions. In further view of waste decomposition process around landfill, aerobic microorganisms multiply and consume O₂ rapidly in early step of waste reclamation to generate CO₂, H₂ and low grade hydrocarbon (HC) along with heats. As certain times go by, the filled-up layer turns anaerobic and gets decomposed by anaerobic organisms to generate fatty acid, alcohol, CO₂, NH₃, H₂ and more. Animal carcass is an easily decomposable constituent, so more and more gas gets decomposed from carcass in 2 years after reclamation. And it is known that the half-life period of wastes amounts to about 1 year, so 99% of wastes reclaimed become fully decomposed in 3 and half years²⁾.

On the other hand, the pollution of NO₃-N as final degradation product of organisms becomes more concentrated rapidly into shallow undergroundwater due

to the immixture of nitrogen fertilizer and other working pollutants include decayed animal/plant, domestic sewage, waste residues and more³⁾.

The toxicity of NO₃-N exists in form of (NH₄)₂CO₃ by a combination with CO₂, while organisms become decayed and decomposed. The organisms oxidized by bacteria become NH₃-N and re-oxidized for decomposition into NO₃-N as final degradation product of organisms⁴⁾. It was reported that NO₃-N is reduced to nitrite nitrogen (NO₂-N) by the microorganisms, and the reduced NO₂-N could react with hemoglobin (Hb). If It was absorbed into blood flow in vivo, it could cause the partial loss of O₂ transport system, particularly fatal Blue Baby disease to infants^{5,6)}. In addition, it is also reported that NO₃-N reacts with gastric acid to generate nitrosamines, which becomes very widespread harmful element. WHO already provides that NO₃-N should be contained below 10 ppm (tolerable level) in drinking water⁵⁾.

Since Blue Baby disease was first reported from infant cases by Comly (1945) in the USA, the damage and pollution of groundwater due to NO₃-N has been steadily reported around the world including Czechoslovakia (from 1953 to 1960), Minnesota, USA and Germany (during 1960's) and more^{5,7)}, and such pollution occurs more often in rural areas using groundwater as potable one than urban areas. In Korea, the onset of Blue Baby disease was already reported from infant cases of feeding dry milk powder as mixed with polluted potable ground water.

Thus, focusing on farmhouses using ground water in the vicinity of the area buried with dead livestock due to onset of foot-and-mouth-disease (FMD) in 2002, this study was intended to provide the improved measures after investigating into the basic test items of drinking water maintenance guideline and the pollution extent of $\text{NO}_3\text{-N}$ and $\text{NH}_3\text{-N}$.

Materials and Methods

Sampling

In 2002, the FMD occurred largely around Anseong-city and Yongin-city, Gyeonggi province as shown in Fig 1. Any morbid animals and all the livestock in certain critical zone were destroyed and buried in accordance with the Livestock Plague Prevention Act. Target for sampling were a total of 10 farmhouses using groundwater at 200 meters or shorter distance from landfill to get drinking water for men or animals, and

water samples were collected once every month from March to August 2005 (during 6 months) for experiment, and the 10 farmhouses for water sampling were listed in Table 1. The water samples were collected using each sterile water sampling bottle (PE, 1 l). They were kept at cool temperature in icebox and carried into laboratory for experiment.

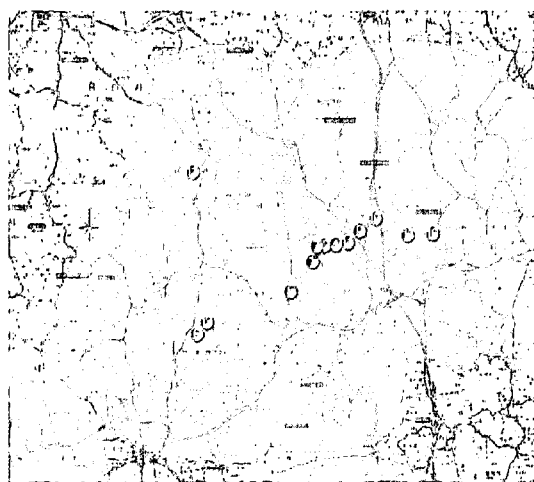


Fig 1. The map of Gyeonggi province where FMD occurred in 2002

Table 1. Details of the sites in which groundwater were collected

Number of site	City	Buried animals at landfill		Distance from landfill	Number of site	City	Buried animals at landfill		Distance from landfill
		Species	Number				Species	Number	
1	Yongin	swine	1,640	50 m	9.10	Ansung	bovine	78	70 m
2.3	Yongin	swine	11,028	200 m	11.12	Ansung	swine	1,047	50 m
4.5	Yongin	swine	1,416	50 m	13	Ansung	swine	1,206	50 m
6	Yongin	swine	913	50 m	14.15	Ansung	swine	600	50 m
7.8	Yongin	swine	1,300	50 m	16.17	Ansung	swine	700	50 m

Reagents and Instruments

Phenol, sodium hypochloride, sodium hydroxide used in this study were special grade reagents (Sigma, USA), while

standard solutions such as Pb, As, Hg, Cd, Cu, Zn, Fe, Mn and Al were purchased as products made at 1,000 ppm (Wako Chem, Japan). In addition, ion chromatography (IC, Dionex 300, USA),

pH meter (Sentron Titan, Japan), Turbidimeter (HACH, Japan), inductive couple plasma (ICP, GBC Xtra, USA) and UV/VIS spectrophotometer (GBC Xtra, USA) were used for data analysis.

Test items and methods

Out of regulations about drinking water quality criteria and test, the guideline for drinking water quality test⁸⁾ was applied to testing 16 items including NO₃-N, and analysis instruments used for each test item were listed in Table 2.

Table 2. Methods and main instruments used for the test of each item

Items	Methods	Main instruments
NO ₃ -N	IC method	IC*
NH ₃ -N	Spectrophotometer method	UV spectrophotometer
Pb, As, Hg, Cd Cu, Zn, Fe, Mn, Al	ICP method	ICP
pH	Electrode method	pH meter
Color	Visual comparison method	Colorimeter
Turbidity	Visual comparison method	Turbidimeter
Total colony counts	Pour plate method	Incubator
Total coliforms	MPN method	Incubator

* : IC = ion chromatography, ICP = inductive couple plasma, UV = ultraviolet and visible, MPN = most probable number

Results and Discussions

Nitrate-nitrogen (NO₃-N) and ammonia-nitrogen (NH₃-N)

As shown in Table 3, contents of NH₃-N was little or not detected. All groundwater samples remained below reference drinking water quality level equivalent to 0.5mg/ℓ.

However, there were 4 sites (23.5%) of ground water in which NO₃-N exceeded 10 mg/ℓ of drinking water standard level, and the highest concentration of NO₃-N reached up to 24.1 mg/ℓ, while its lowest concentration was 0.3 mg/ℓ (mean = 8.46 mg/ℓ).

After a 6-month examination from March to August 2005, the concentration

of NO₃-N showed that there were significant monthly differences (25.7, 22.1, 14.9, 8.5, 11.2 and 10.8mg/ℓ) in concentration at one site (site 1) as shown in Fig 2, while there was not any significant monthly difference in concentration of other groundwater.

As shown in Table 1 and 3, there was no difference in the NO₃-N concentration of groundwater sample between site 2 and site 17. As a result, it was found that the concentration of NO₃-N in groundwater was in no association with the number of animal carcasses buried and the distance from landfill. NH₃-N was generated while organisms such as animal carcass in natural world were decayed and decomposed, and also could exist in form of (NH₄)₂CO₃. It is used as

an index of relatively recent pollutions including human or animal excretions, domestic sewage and industrial sewage, while $\text{NO}_3\text{-N}$ is used as an index of conventional pollutions induced by organisms because it is the final degradation product of organisms⁹⁾. As like in Table 3, it was found that the concentration of $\text{NO}_3\text{-N}$ in 4 ground-water samples exceeded the reference water quality level ($10\text{mg}/\ell$), which was enough to estimate

pollutions induced by organisms in past. But it would be necessary to conduct additional confirmatory tests and studies to determine whether or not the pollution was caused by animal carcasses buried in landfill.

On the other hand, the extent of pollution induced by $\text{NO}_3\text{-N}$ (4 sites, 23.5%) was comparable to that as reported in other studies. For instance, Kang's study¹⁰⁾ on groundwater pollution induced by

Table 3. Mean concentration(mg/ℓ) of $\text{NH}_3\text{-N}$ and $\text{NO}_3\text{-N}$ in groundwater

Site No	$\text{NH}_3\text{-N}$	$\text{NO}_3\text{-N}$	Site No	$\text{NH}_3\text{-N}$	$\text{NO}_3\text{-N}$
1	0.05	15.5	10	-	8.5
2	0.01	9.4	11	-	2.1
3	*	10.0	12	0.01	5.6
4	-	9.0	13	-	24.1
5	-	3.3	14	-	6.0
6	-	8.9	15	-	2.4
7	-	0.3	16	-	5.4
8	-	0.4	17	-	10.6
9	0.01	20.7			

* : Not detected

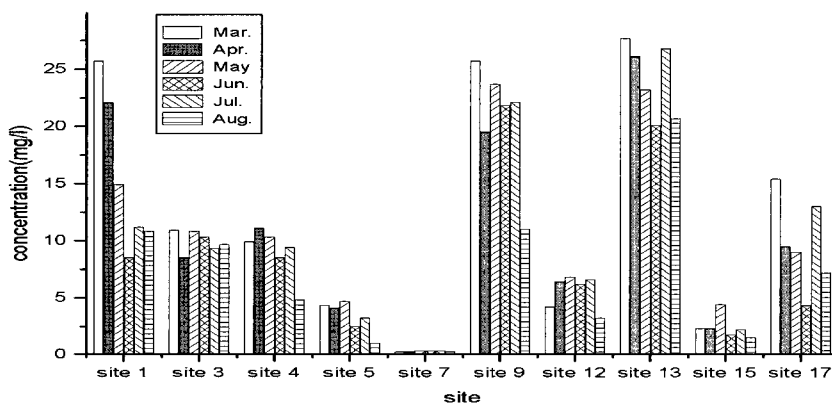


Fig. 2. Monthly distribution of concentration(mg/ℓ) of $\text{NH}_3\text{-N}$ from March to August in 2005

NO₃-N reported that out of all 117 sites with water supply facilities for civil defense, there were 17 sites (14.5%) in which pollutions exceeded the reference drinking water quality level, and those pollutions were not associated with the depth of groundwater. In a study on water quality test applied to 83 ground water sites for multi-household tenement, Jung et al¹¹⁾ reported that the concentration of NO₃-N in some samples from Pocheon (30%), Euijeongbu (33.3 %) and Goyang (40%) failed to meet reference water quality level. Oh et al¹²⁾ also reported that 51 of 330 groundwater samples from sites without waterworks exceeded reference drinking water quality level. In this context, even though there were more or less differences depending on regions, the findings of this study also showed comparable percentages of NO₃-N at concentration exceeding reference level across different sites. However, in view of oxidation and reduction of organisms, in order

to prevent pollutants from leak into adjacent sites when burying carcasses, it will be required to install the safe water containment facilities to prevent leached wastewater from polluting groundwater, and fill up the sites for easy discharge of ammoniac gas, so that organisms can be fully decomposed in shorter periods.

Potential of hydrogen (pH), color and turbidity

Potential of hydrogen (pH) depends upon the concentration of free carbon and carbonate dissolved in water, and is also used as an index of artificial pollution level. CO₂ in land soil and the water with much CO₂ dissolved by degradation product from organisms indicate subacid level. As shown in Table 4, this study showed pH ranging from 5.8 to 6.6, and there was no groundwater exceeding reference drinking water quality level (i.e, 5.8 to 8.5).

Table 4. Mean value of pH, color and turbidity (NTU) in the groundwater of each site

Items	Sites																
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
pH	5.8	6.5	6.5	6.4	5.9	5.9	6.6	6.6	6.0	5.9	6.6	6.3	5.9	5.9	5.8	5.8	5.9
Color	1.5	2.3	1.5	1.5	1.5	1.7	1.5	1.8	3.5	2.0	2.2	2.3	2.7	8.5	2.0	4.7	2.0
Turbidity	0.1	0.7	0.9	0.1	1.6	0.3	0.2	0.1	0.4	0.2	0.1	0.3	0.5	2.6	2.9	2.2	0.1

Color may be increased under the influence of microorganisms or plankton. Reference water quality level is set up below Grade 5, and turbidity is expressed in NTU. Reference level of turbidity is below 1 NTU (for drinking water quality). The sources causing in-

creased turbidity in water include minerals, floating organisms, microorganisms and floats. In rainy days, for example, the turbidity of groundwater and surface water get higher largely due to floats. Certain water purification methods such as filtering and coagulation settling may

be used to decrease turbidity¹³). As like Table 4, sample from one site (site 14) exceeded the reference color level, while sample from 4 sites (site 5, 14, 15, 16) exceeded reference turbidity level. Although it was difficult to exclude that this finding resulted from worse quality of ground water, it was assumed that main reasons of increased turbidity came from the influence of lichen or other substances attached upon water tank and piping exposed to sun-light because stockbreeding farmhouses store groundwater in water tank and water pipe made mainly of transparent plastics without using any light-shielding material. As shown in Table 1 and 4, it was found that pH, color and turbidity of groundwater in vicinity of landfill were not associated with the number of livestock buried and the distance from landfill.

Metal ion

The pollution of metal ion into groundwater is closely associated with soil pollution, and related with the quality of rock comprising water catchment zone. Any pollutant of soils is typically permeated into soil and comes to groundwater as like raindrop does. Factors causing high concentration of heavy metals in groundwater include the contact with strata containing interstitial water and high concentrate metals, long or short-term mixture due water-leaking-factors, and artificial behaviors¹⁴).

Most harmful metals were not dissolved in chemical compound or stabilized in water, but remained a mixture enough

to pollute water and soil. If they were carried *in vivo* along with a variety of foods along food chain, they became accumulated more depending on bioconcentration, causing dreadful diseases like neuroplegia, language disorder, paraplegia by harmful metal poisoning¹⁴). In particular, the harmful metals, if once accumulated *in vivo*, are hardly discharged from body. This heavy metal accumulation *in vivo* is so terrible and difficult to cure of. That is why the reference level of metal contained in drinking water are legally provided and applied to practice as follows: Pb, As 0.05 $\mu\text{g}/\text{ml}$, Hg 0.001 $\mu\text{g}/\text{ml}$, Cd 0.01 $\mu\text{g}/\text{ml}$, Fe, Mn 0.3 $\mu\text{g}/\text{ml}$, Cu, Zn 1.0 $\mu\text{g}/\text{ml}$ and Al 0.2 $\mu\text{g}/\text{ml}$ or lower⁸).

In this study, harmful metals against human body such as Pb, Cd, As and Hg were not detected as shown in Table 5. And the metals indispensable for human body were detected within reference level as follows: Cu = ND - 0.22 mg/ℓ , Zn = 0.01 - 0.05 mg/ℓ , Fe = ND - 0.05 mg/ℓ , and Al = 0.03 - 0.16 mg/ℓ .

On the other hand, Zn was detected at ND - 2.49 mg/ℓ . Here, only one site showed very high Zn concentration at 2.49 mg/ℓ , which was 83 times as high as reference level at 0.3 mg/ℓ .

There was no domestic case of exceeding the reference water quality level in the investigation of Pb, Cd, As and Hg. However, as shown in Table 5, Mn was detected from one site (site 10) in great excess of reference water quality level, but was detected within the reference level (0.11 mg/ℓ) in vicinity of same farm. This difference was possibly attributed to influence of rock quality around

Table 5. Mean concentration(mg/ ℓ) of metal ions in the groundwater of each site

Metals	Sites																
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Pb	-*	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
As	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Hg	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Cd	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Cu	-	-	-	0.22	0.01	-	-	-	0.01	0.01	-	0.01	0.01	0.01	0.01	0.01	-
Zn	0.04	0.01	0.02	0.02	0.04	0.06	0.01	0.02	0.05	0.06	0.01	0.19	0.13	0.29	0.04	0.03	0.05
Fe	0.05	-	-	-	-	-	-	-	-	-	-	-	-	0.04	0.05	0.02	-
Mn	0.19	-	-	0.02	0.02	0.01	-	-	0.11	2.49	0.01	-	0.01	0.01	0.03	0.03	0.01
Al	0.16	0.08	0.09	0.06	0.14	0.10	0.04	0.04	0.10	0.18	0.03	0.06	0.15	0.13	0.13	0.11	0.11

*: - : Not done

Table 6. The contamination of total colony counts and *Escherichiae* spp in the groundwater of each site (CFU/ml)

Bacteria	Site																
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Total colonies	-	-	-	49	-	-	-	-	-	50	-	-	-	-	21	-	-
<i>Escherichiae</i> spp	-	-	-	1	-	-	-	-	-	-	-	-	-	-	7	-	-

water catchment zone.

Bacterial Pollution

Generally, common bacteria and coliforms are critical index of groundwater pollution induced by microorganisms, so reference level for them is set up and applied in practice. As shown in Table 6, common bacteria were detected in 2 sites (site 4, 15) at level of 491 CFU/ml and 217 CFU/ml respectively, which were excess of reference level equivalent to 100 CFU/ml. It was detected in 1 site (site 10) at 50 CFU/ml but not detected in other sites. Coliform was not detected

in any site. In a study on actual profile of water quality around multi-household tenement using groundwater, Jung et al¹¹⁾ reported that the percentage of bacteriologically inadequate samples amounted to 15.7%. In another study on actual profile of water quality around sites without waterworks, Oh et al¹²⁾ reported that the percentage of common bacteriologically inadequate samples amounted to 11.5%, while coliforms and fecal coliforms were detected in 17 out of all 330 cases.

However, there was the less number of samples in excess of reference level, i.e. the less number of microorganisms detected than other comparable studies,

possibly because UV lamp was used in water tank to reduce microorganisms in ground water widely used around stock-breeding farmhouses.

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