Groundwater contamination with nitrogenous compounds in Kumamoto Prefecture and Hanoi City -Present conditions and adopted countermeasures-

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Ⅰ. Introduction

Nitrate (NO^{3-}) groundwater pollution has become a serious problem in Kumamoto Prefecture, especially in areas of intensive agricultural activities including watermelon production in greenhouses and livestock farming. Numerous wells used for potable water production are in violation of the drinking water standard for NO_3-N of 10 mg /L. As a temporary measure, ion exchange is being used for NO^3 abatement in polluted well water of the northern part of Kumamoto Prefecture. Conversely, in Hanoi City, Vietnam, groundwater pollution with NH_4^+ has become a serious problem. Two potential sources for this ammonium (NH_4^+) contamination are considered: One is anthropogenic, such as the discharge of untreated sewage, and the other natural, such as buried manglobe forests in the alluvial soil.

Kumamoto University is a member of the Core University Program between Osaka University and the Vietnam National University sponsored by JSPS-NSCT. From the Department of Civil Engineering at Kumamoto University a research project, titled "Studies on the development of effective purification methods for groundwater polluted with nitrogenous compounds", was developed. This proposal was targeted for a joint research effort with the Center for Environmental Engineering of Towns and Industrial Areas (CEETIA) of Hanoi University of Civil Engineering (HUCE). As a part of this project, our research groups collected field samples from rivers and canals and from wells penetrating the Holocene and Pleitocene aquifers in greater Hanoi. Various chemical parameters were quantified in the field and samples were transported to Kumamoto University for further detailed analyses, the results of which are discussed with other data in this paper.

Ⅱ. Nitrate contamination in groundwater of Kumamoto Prefecture

Ⅱ-1 Nitrate contamination in tap water of Kumamoto City

The characteristics of well water used as potable water sources in Kumamoto City are shown in Table 1.

Table 1. Water quality of wells used for potable water in Kumamoto City[1)]

Well name (Number of wells)	Well Type	(NO_2+NO_3)-N (mg/L)	Total residue (mg/L)	p H	Alkalinity (mg/L)
Kenngunn(11)	DW(4) BW(7)	2.3	170	6.8	57
Numayazu(10)	DW	1.5	185	7.0	57
Akita(2)	DW	1.5	190	7.0	37
Nishikajio(3)	DW	4.0	175	6.8	50
Tobita(2)	DW	3.4	185	6.7	54
Mituzu(2)	DW	2.1	150	6.9	60
Kaiki(1)	DW	2.6	150	7.2	46
Turuhata(1)	DW	4.6	200	7.2	70
Kajio(1)	DW	4.7	224	7.1	70
Asota(9)	DW	2.5	150	7.2	60
Shoukuti(8)	DW	2.5	175	6.8	57
Takuma(5)	DW	4.0	170	6.8	60
Takuma No.5	DW	6.5	190	6.6	60
Funatsu(1)	DW	1.3	105	7.4	40
Shirahama(1)	DW	1.5	106	7.6	40
Shioya(1)	DW	6.9	230	6.9	45
Kawatoko(1)	S	1.1	95	7.3	35
Yoshino(1)	S	1.0	100	7.3	39
Oiwake(1)	DW	1.8	125	7.3	54
Yokoyamakuroishi(1)	DW	0.38	123	7.7	54
Hakkeimizutani(2)	SW	3.0	160	6.9	66
Hakkeimizutani(2)	SW	0.9	250	7.7	105
Ippongi(1)	SW	2.9	180	7.0	67
Ippongi(2)	DW	0.9	170	7.1	67
Ippongi(1)	DW	2.6	150	7.1	60
Yamamuro(2)	DW	0.5	230	7.6	85
Kamei(1)	SW	4.0	180	7.0	80
Kamei(3)	DW	2.0	170	7.3	70
Kawajiri(2)	DW	0.5	200	7.3	63
Shiroyama(4)	DW	<0.01	270	7.4	95
Ikegami(3)	DW	0.5	130	7.3	60
Akuta(2)	DW	<0.01	250	7.8	100
Tennmei(3)	DW	<0.01	300	7.8	100

DW : Deep well, SW : Shallow well, BW : Blow up well, S : Spring

Most of the wells used for Kumamoto's water supply extend to deep aquifer zones. The compositions of these groundwaters vary considerably with ranges of total solids from 130 to 300 mg/L, alkalinity from 40 to 110 mg/L, NO_2+NO_3-N from 0.01 to 6.9 mg/L and pH from 6.4 to 7.8. A significant characteristic of these well waters is a high concentration of NO_2+NO_3-N. The highest NO_2+NO_3-N level of 6.9 mg/L was recorded at the Shioya well. The reason for the high concentration at this well is believed to be due to application of chemical fertilizers for orange cultivation. High NO_2+NO_3-N levels were also

recorded at the Takuma wells and the reason is also thought to be due to use of nitrogenous fertilizers in the surrounding agricultural areas. Another likely contributing factor is a decrease in groundwater volume. Large factories of the IT industry located in the upper region of the greater Kumamoto groundwater zone have extracted large amounts of water from the aquifer. To compensate for the high nitrogen levels, the Water Supply Bureau blends various well waters to maintain NO_3-N levels at 2 to 4 mg/L in the delivery system. Furthermore, Kumamoto City is now developing and action plan intended to increase groundwater volumes by improved resource management in the upper aquifer zone.

Ⅱ-2 Groundwater contamination in northern Kumamoto Prefecture

From the results of a recent field survey, groundwater in the northern part of Kumamoto Prefecture was found to be highly polluted with NO_3^-. Hirohata et al. [2] classified the groundwater of Kumamoto into four groups as shown in Figure 1 based on relationships between NO_3-N concentration and $\delta^{15}N$ values.

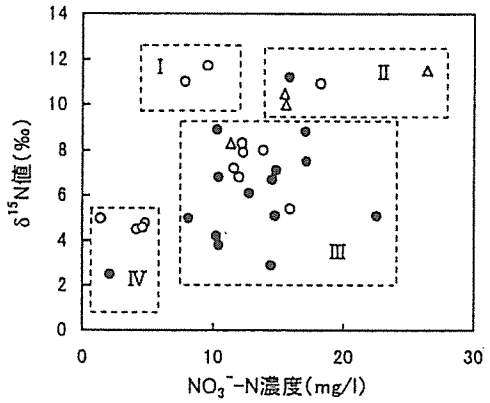

Figure 2. Relationship between NO_3-N concentration and $\delta^{15}N$ value.

(○: Feb. 1995, △: No.4. 1995, ●: Feb. 1996)

The results of their investigation revealed 19 sites as being in the group III category. The soils in the areas categorized as group III were from greenhouses and vegetable farms. Testing for fecal coliforms yielded negative results in 18 of the group III sites. These results indicated the absence of contamination from gray water and wastewater from livestock operations in those areas. NO^{3-} pollution in the groundwater at group III locations was thus best explained as originating from chemical fertilizers.

Ⅱ-3 Nitrate abatement in groundwater of Kumamoto Prefecture

Kumamoto Prefecture has initiated an action plan for the development of fertile soil with a concurrent reduction in the use of agricultural chemicals. The plan, which started in 1990, calls for a 30% weight reduction in application of nitrogenous fertilizers. The target values for the amounts of agricultural

chemicals and fertilizers to be used are shown in Table 2.

The amount of chemical fertilizer used in 1999 (98,022t/y) was reduced to 58% of the amount used in the reference year (170,000t/y, 1986-1988). Therefore, the target was attained before the target year of 2000.

Table 2. Target values for the amounts of chemical fertilizers and agricultural chemicals used in Kumamoto Prefecture

	Usage in reference year	Target values	Value for 2000
Chemical fertilizers	170,000(100)	153,000(90)	119,000(70)
Agricultural chemicals	26,000(100)	23,000(90)	18,000(70)

Unit: ton(%, with respect to reference year)

Trends in the amounts of chemicals and fertilizers used annually in Kumamoto Prefecture are shown in Table 3.

Table 3. Annual amounts of chemical fertilizers and agricultural chemicals used in Kumamoto Prefecture

	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Chemical fertilizers	165.0 (97)	145.5 (86)	143.3 (84)	136.8 (80)	129.1 (76)	127.9 (75)	123.4 (73)	116.8 (69)	109.4 (64)	105.4 (62)	98.0 (58)
Agricultural chemicals	22.5 (87)	22.1 (85)	21.5 (83)	18.5 (71)	19.2 (74)	18.3 (70)	17.4 (67)	16.1 (62)	14.6 (56)	13.9 (54)	13.4 (52)

Unit: x 1000 ton(%, with respect to reference year)

Ⅱ-4 Future targets

Kumamoto Prefecture is now conducting the action plan, whose target is the 20% weight reduction of nitrogenous chemical fertilizers based on the "Action plan for the preparation of healthy soil and reduction of agricultural chemicals" which started in 2000. Table 4 shows the target values for the amounts of chemical fertilizers and agricultural chemicals addressed in this action plan.

Table 4. Targets for the agricultural plan of Kumamoto Prefecture

Name of index	Reference yaer (2000)	Target yaer (2010)
・ Total amounts of chemical fertilizers and agricultural chemicals used (%)	100	80
・ Total amounts of compost circulationg(t)	11,000	30,000
・ Number of eco-farmers	60	1,000
・ Certificate amounts of "Yusaku-kun"(t)	2,500	7,500

Ⅱ-5 Action plans

Kumamoto Prefecture is now tackling existing groundwater pollution problems with the goal of early achievement of the stated targets. Action plans now in progress include the following activities:

1) Determination of appropriate dosage requirements for chemical fertilizers based on soil diagnoses.

2) Field surveys of soil and groundwater and testing for improved fertilizing practices to reduce the environmental impacts resulting from agricultural activities.

3) Development of programs for demonstrating to the public the methods and purposes of existing and proposed action plans.

4) Proper disposal of livestock manure and promotion of its use as a beneficial resource.

Ⅲ. Ammonium contamination in groundwater of Hanoi City

Ⅲ-1 Surface water contamination in Hanoi City

The locations of surface water and groundwater sampling points in the Hanoi area for this survey are shown in Figure 2.

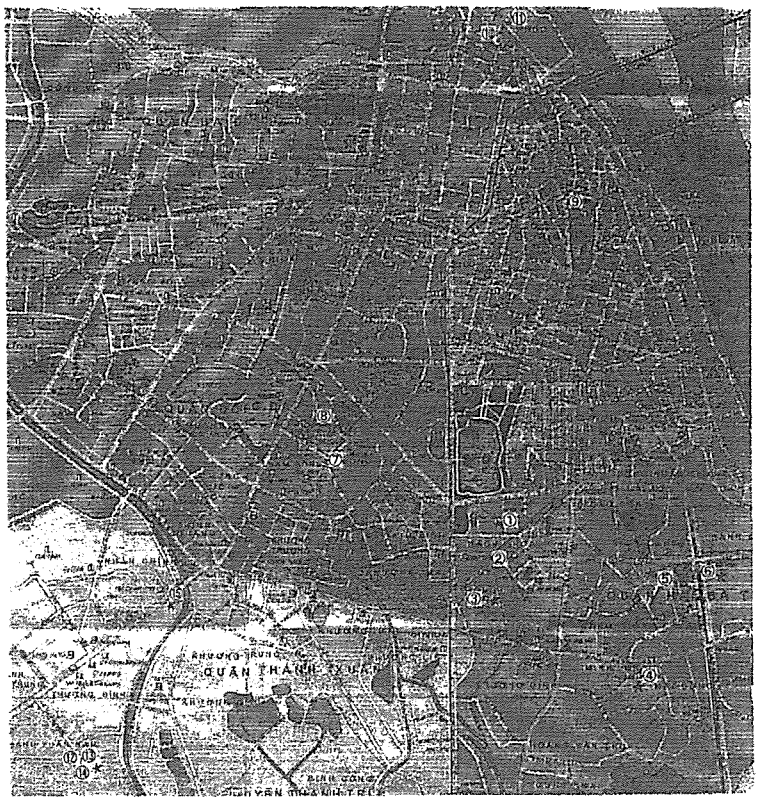

Figure 2. Surface water and groundwater sampling points in Hanoi area.

Table 5(1) and 5(2) shows the analytical results of surface water and groundwater in Hanoi area.

Table 5(1). Analytical results of water quality in Hanoi area

No	Name of well	Temp oC	pH	NO_3-N mg/L	NH_4-N mg/L	SS mg/L	SO_4^{2-} mg/L	SiO_2 mg/L	S^{2-} mg/L	IC mg/L	TOC mg/L	TC mg/L
1	Bach khoa	26.1	6.84	4.3	12.4		1.6	52.8	0.5	24.8	1.4	26.2
2	Dong Tam **	25.3	6.60	5.0	25.0		0.8	23.7	0.5	26.0	2.8	28.8
3	HUCE	25.3	6.69	7.8	10.0		5.2	53.8	0.0	26.6	1.6	28.2
4	Minh Khai **	34.3	6.70	9.7	7.6		14.4	42.6	0.9	25.5	1.7	27.2
5	Quynh Mai	25.3	6.80	0.9	3.6		4.2	35.0	1.3	13.6	0.5	14.1
6	KimNguu canal *	23.4	7.00	12.0	34.0	80	18.2	41.7	1.6	20.9	13.8	34.7
7	Lu River-out *	21.3	8.11	6.5	10.1	35	23.4	23.0	0.5	14.3	3.2	17.5
8	Lu River-in *	22.3	7.72	8.5	37	50	5.0	34.9	1.3	20.2	5.9	26.1
9	HoanKiem Lake *	19.9	10.30	4.3	0.3	175	1.6	21.8	1.4	1.9	8.3	10.2
10	Yen Phu 12	25.0	7.38	1.7	1.1		2.1	22.1	1.4	4.6	0.5	5.1
11	Yen Phu 20	25.2	7.10	3.6	4.6		2.6	45.1	2.2	6.3	0.6	6.9
12	Ha Dinh 5	26.0	6.82	3.3	12.3		2.2	41.3	0.1	23.5	2.5	26.0
13	Ha Dinh 6	26.3	6.88	3.2	11.6		2.6	37.9	0.9	22.3	1.3	23.6
14	Ha Dinh 8	26.0	6.86	3.7	13.1		2.1	44.6	0.1	17.8	1.7	19.5
15	Cau Moi *	22.0	7.43	7.2	31.0	25	11.7	90.9	0.8	18.1	5.4	23.5
16	Van Dien *	21.3	7.80	10.2	13.3	50	29.2	29.3	0.5	12.6	4.6	17.2
17	Phap Van No.1	27.0	6.90	3.2	28.0		2.8	30.9	0.0	20.7	3.2	23.9
18	Phap Van No.2	26.3	6.80	2.8	19.0		2.4	44.0	0.0	21.1	3.0	24.1
19	Phap Van No.3	26.3	6.80	2.9	15.4		2.1	41.4	1.2	18.7	2.8	21.5
20	Univ. tap	20.3	8.06	6.5	0.1		5.3	15.8	0.4	7.2	0.4	7.6

* Surface water ** Personal well

Table 5(2). Analytical results of water quality in Hanoi area

No	Name of well	Fe mg/L	Ca mg/L	Mg mg/L	Na mg/L	K mg/L	Mn mg/L	As mg/L
1	Bach khoa	11.5	25.1	12.4	29.0	3.0	0	0.054
2	Dong Tam **	20.5	51.8	15.9	26.2	2.5	0	0.088
3	HUCE	31.6	42.8	18	44.0	1.9	0	0.020
4	Minh Khai **	24.0	45.9	20.8	72.4	2.3	0	0.017
5	Quynh Mai	4.3	15.2	10.9	9.1	1.7	0	0.050
6	Kim Nguu canal*	3.3	37.2	14.3	44.6	20.2	0	0.020
7	Lu River-out *	0.7	30.8	10.6	31.2	14.0	0	0.006
8	Lu River-in *	1.2	36.1	12.9	40.2	17.5	0	0.013
9	HoanKiem Lake *	1.0	13.7	1.4	8.3	6.1	0	0.005
10	Yen Phu 12	2.5	32.8	8.6	7.2	1.9	0	0.053
11	Yen Phu 20	9.1	40.5	13.8	18.2	6.7	0	0.080
12	Ha Dinh 5	15.6	25.5	14.8	27.4	3.1	0	0.120
13	Ha Dinh 6	12.1	23.8	12.7	30.8	3.6	0	0.120
14	Ha Dinh 8	15.0	23.4	13.5	28.7	3.8	0	0.078
15	Cau Moi (*)	1.0	33.1	16.6	44.4	14.8	0	0.008
16	Van Dien *	1.5	33.5	14	47.9	21.1	0	0.014
17	Phap Van No.1	9.5	25.3	13.2	32.6	5.7	0	0.070
18	Phap Van No.2	9.7	21.2	12	34.2	5.3	0	0.051
19	Phap Van No.3	9.5	20.9	11.8	36.2	5.4	0	0.039
20	Univ. tap	0.1	26.9	6.3	3.0	1.7	0	0.020

* Surface water ** Personal well

Table 5(1) and 5(2) shows the analytical results of surface water and groundwater in Hanoi area. The Kim Nguu River is one of the four primary tributaries in Hanoi which transverses through the east and south-east parts of the city. This river serves as a canal for the transport of domestic and industrial wastewater and is heavily polluted, especially in the dry season. The ranges of various water quality parameters during the period from 1995 to 1999 in the Kim Nguu River at the Minh Khai Bridge and the Yen So Watergate are as follows: temperature, 26.3 to 27.5 ℃; pH, 7.2 to 7.0; suspended solids (SS), 96 to 110 mg/L; turbidity, 48 to 90 NTU; conductivity (EC), 750 to 609 μs/cm; total dissolved solids (TDS), 376 to 304 mg/L; dissolved oxygen (DO), 0.69 to 2.1mg/L; BOD_5, 153.6 to 89.6 mg/L; COD, 292 to 164 mg/L; NH_4-N, 12.5 to 6.6 mg/L; NO_2-N, 0.03 to 0.39 mg/L; PO_4^{3-}, 1.2 to 0.9 mg/L; Cl^-, 43 to 23.6 mg/L; total iron, 0.70 to 0.97 mg/L; and total coliforms, 500 to 345 MPN/100 ml.

The results of chemical analyses from the November, 2000, survey revealed an NH_4-N level of 34 mg/L at a location 150 m downstream from the Lo Duc discharge point. In addition, at the junction of the Kim Nguu, Lu and Set Rivers near the Van Dien Bridge on Road No. 1, 10 km south from the center of Hanoi City, the NH_4-N level was 13.3 mg/L.

The sewerage system in the Hanoi urban area is mostly a combined system. During the dry season, all of the city's wastewater is discharged into the Nhue River through the Thanhliet Gate. The ranges of various water quality parameters of the Tolich River, one of the largest of Hanoi's four inner-city rivers, from the Cau Moi Bridge to the Thanhliet Gate are as follows: temperature, 26.3 to 27.5 ℃; pH, 6.8 to 7.0; SS, 65 to 19 mg/L; turbidity, 52 to 34 NTU; EC, 635 to 632 μs/cm; TDS, 329 to 314 mg/L; DO, 0.53 to 2.1; BOD_5, 132.1 to 66.2 mg/L; COD, 343 to 171 mg/L; NH_4-N 11.4 to 7.9 mg/L; NO_2-N, below detection to 0.12 mg/L; PO_4^{3-}, 1.4 to 0.6 mg/L; Cl^-, 52.4 to 45.6 mg/L; total iron, 0.79 to 0.36 mg/L; and total coliforms, 130,000 to 35,000 MPN/100 ml.

From the sampling event of November, 2000, the NH_4-N level at the Cau Moi Bridge was 31 mg/L. In addition, at the inlet from the Lu River into Trungtu Lake, the NH_4-N level was 37 mg/L and at the outlet from the lake, 10.1 mg/L.

Average values of various water quality parameters at Bay Mau Lake are as follows: temperature, 25.5 ℃; pH, 7.3; SS, 44 mg/L; turbidity, 34.8 NTU; EC, 592 μs/cm; TDS, 311 mg/L; DO, 6.3 mg/L; BOD_5, 21.1 mg/L; COD, 46.7 mg/L; NH_4-N, 3.8 mg/L; NO_2-N, 0.47 mg/L; PO_4^{3-}, 1.0 mg/L; Cl^-, 44.8 mg/L; total iron, 0.18 mg/L; Pb, 0.048 mg/L; SO_4^{2-} ,30 mg/L; and total coliforms, 4.56 MPN/100 ml.

The waters of rivers used as drainage canals in Hanoi City, such as the Tolich and Kimnguu Rivers, are heavily polluted. The levles of BOD and COD are 2 to 3 times higher than typical values for river water in Hanoi and coliforms are two to three orders of magnitude higher than regulatory limits.

Downstream from the city, the rivers are still used for fishing, however, the current state of these rivers, in particular for NH_4^+, COD, BOD and coliforms, has deteriorated significantly in comparison with water quality data of 1995 and 1996. Apparently due to natural purification, the pollution level in Bay Mau Lake is not excessive and it qualifies as a B-level surface water body. The water quality of the lake was stable during the monitoring period of 1995 and 1996.

Ⅲ-2 Groundwater contamination in Hanoi City

In Hanoi City, especially south of the Red River, there is intensive exploitation of groundwater and studies have shown that river water and groundwater in the region are hydraulically interconnected. Pollution of groundwater in the Hanoi urban area is especially evidenced by the presence of NH_4^+ in the Holocel and Pleistocen aquifers where the NH_4-N levels may be as high as 25 to 30 mg/L. This groundwater pollution is evidenced in some of the most developed areas in central Hanoi City and effects the water supply stations of Luong Yen, Tuong Mai, Bach Khoa and Ngo Si Lien. Furthermore, in the Ha Dinh and Phap Van areas, almost all wells drawing from the Pleistocene aquifer are strongly polluted with high levels of NH_4^+.

There are differing explanations as to why contamination of groundwaters with nitrogenous compounds is occurring. One proposal considers the transfer of polluted water from surface sources into upper aquifer zones, which then passes to the deeper zones through geological windows or discontinuities in subsurface impervious strata. In this scenario, pollution from anthropogenic origins is responsible for the nitrogenous compounds that ultimately enter the deep aquifer zones and contaminate these potable water sources. However, natural origins of the contamination are also considered. Regardless of the debated origins of the nitrogenous compounds, the pollution problem is a reality that must be dealt with.

Groundwater qualities in the Hanoi area have been investigated by the Hanoi Clean Water Business Company and considerable data exists for the period from 1994 to 2000. Most recently, research conducted by Kumamoto University and CEETIA have contributed to this data base the joint field survey conducted in November of 2000 and on-going efforts by CEETIA in 2000 and 2001. The joint findings of these inquires are discussed below.

Analyses conducted by CEETIA demonstrated that NH_4-N levels at the inlet to the Phap Van Water Treatment Plant were from 17.5 to 23.4 (average, 21.6) mg/L during August and September 2000, at the end of the rainy season. During April and May, 2001, at the middle of the rainy season, the concentration of NH_4-N was 20.8 mg/L. From fieldwork conducted by the Kumamoto team in November, 2000, groundwater NH_4-N levels at Phap Van wells 1, 2 and 3 ranged from 15.4 to 28.0 (average 20.8) mg/L and at Ha Dinh wells 5, 6 and 8, from 11.6 to 13.1 (average 12.3) mg/L. In the south and south-west

zones of Hanoi City where the Phap Van and Ha Dinh Water Treatment Plants are located, hydrogeological windows linking the higher Holocene and lower Pleistocene aquifers with polluted surface waters are believed to exist.

The Yen Phu treatment plant is located in the northern part of Hanoi City and the groundwater for this plant is drawn from two groups of wells. One group is located inside and the other outside of a dike adjoining the Red River. Field work conducted by personnel of Kumamoto University during November, 2000, revealed an NH_4-N concentration of 1.1 mg/L at Yen Phu well 12 which is located outside of the dike. Inside the dike near the location of the treatment plant at Yen Phu well 20, a higher NH_4-N concentration of 4.6 mg/L was observed.

The well at the HUCE Water Treatment Plant has a depth of 65 m and extracts groundwater from the Pleistocene aquifer at a rate of 15 m^3/h. An NH_4-N level of 10 mg/L was determined at this well by CEETIA. Two private wells that draw water by hand-pump from the Holocene aquifer were also analyzed by CEETIA. The well at Dong Tam is situated only 10 m from the Set River and has a depth of 30 m. The NH_4-N concentration at this well was 25 mg/L. The well at Minh Khai is located about 300 m from the Kim Nguu River and has a depth of 25 m. The NH_4-N concentration at this well was 7.6 mg/L. These results suggest that groundwater wells closer to the river sustain higher NH_4^+ concentrations due to surface water and groundwater interactions.

The following characteristics can be drawn form the investigation on groundwater pollution in the urban areas of Hanoi City:

1. Groundwaters in the aquifers of Hanoi are polluted with nitrogenous compound predominately in the form of NH_4^+ and can be divided into the following categories:

(1) Very heavily polluted areas in which NH_4-N levels are greater than 10 mg/L. Areas included in this category include Kim Lien, Bach Khoa, Ha Dinh, Tuong Mai and Phap Van.

(2) Heavily polluted areas in which NH_4-N levels range from 5 to 10mg/L. Only Quynh Mai is included in this category.

(3) Moderately polluted areas in which NH_4-N levels range from 1 to 5mg/L. This category includes Luong Yen, Yen Phu, Ngo Sy Lien and Don Thuy.

(4) Slightly polluted areas in which NH_4-N levels are only from trace to 1 mg/L. Areas included in this category are Ngoc Ha, Mai Dich and Thuy Loi University.

2. NO_2-N and NO_3-N pollution is not significant in the groundwater of Hanoi City and is consistently below regulatory limits (data not shown). However, there are some irregular appearances of these contaminants and attention should be given to their occurrence. The places of concern are the south side of La Thanh Dike in the Thanh Tri District, Yen So, Dong Da Hill, the Foreign Language University,

Thuong Dinh, Army Hosital 103, the Quang Trung Mechanical Factory, the Phosphate Factory and the Van Dien Battery Factory (with NO_3-N levels ranging up to 6 or 7 mg/L).

3. The concentrations of nitrogenous compounds are higher during the dry season than the rainy season.

4. Pollutant levels at various locations can often be explained by hydrogeological conditions, the structure of soil layers and the degree of groundwater extraction.

Ⅲ-3 Abatement measurers adopted in Hanoi City

Countermeasures adopted to reduce levels of nitrogenous compounds in the groundwaters of Hanoi have included treatment by strong oxidation such as chlorination. While this has the drawback of high treatment costs, it includes the beneficial formation of chloramines with long lasting disinfection potential. However, the formation of harmful chlorinated organic compounds, while reduced by the formation of choramines, is still a possibility. Associated health concerns have restricted the use of this method. Microbiological conversion methods are also being considered. Biological methods have the potential of converting nitrogenous compounds to non-toxic, environmentally safe forms and are thus worth further pursuing.

Ⅲ-4 Future research plans

Faculty members of CEETIA at HUCE and the Department of Civil Engineering at Kumamoto University plan to engage in joint research on the development of appropriate nitrogen removal process for groundwater. The two universities also plan to arrange academic exchanges of research staff and to install a bench scale experimental apparatus in CEETIA.

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