SOIL AND GROUNDWATER CONTAMINATION AND REMEDIATION IN JAPAN

T. Hirata
Depts. of Environmental Systems, Wakayama University, Japan

Keywords: soil pollution, groundwater pollution, remediation technologies, hazardous chemicals, volatile organochlorine, trichloroethylene, tetrachloroethylene, nitrate nitrogen, fertilizer reduction.

Contents

1. Introduction
2. Present State of Groundwater Pollution
3. Remediation for Organochlorines
3.1. Availability of Technology
3.2. Soil Excavation and Groundwater Extraction
3.3. Dual Extraction
3.4. Bioremediation
3.5. Maintenance Free Technology
4. Nitrogen Pollution
4.1. Natural Abundance of Nitrogen
4.2. Acidification due to Fertilizer Application
4.3. How to Identify Nitrogen Sources
4.4. Feasible Option for Nitrate Pollution
5. Conclusion
Glossary
Bibliography
Biographical Sketch

Summary

The nationwide groundwater pollution surveys of Japan began in 1982. Since then several tens of thousands of groundwater samples have been collected and analyzed. The results showed levels of organochlorines like trichloroethylene and tetrachloroethylene levels over drinking water standards. In addition, nitrate in groundwater was also detected across the nation at levels above the standard. Such anthropogenic chemicals tend to stay in the subsurface environment for many years, but various sorts of technologies, including some originally developed in Japan and others imported from overseas, have been applied to contaminated sites to remove and degrade hazardous contaminants. This chapter describes the present state of subsurface pollution and the remediation technologies applied so far in Japan.

1. Introduction

More than 20 000 chemicals have been industrially synthesized in order to produce fine chemicals, and parts for electrical devices, etc. and so many man-made substances have been created. A proportion of these chemicals, some of which are hazardous, are

released to the air and water around human habitations. When hazardous chemicals reach the ground they can penetrate the vadose zone and enter the groundwater. In this context, since the early 1980s, there has been a great deal of concern about contamination of public water supply with organic and inorganic substances. In particular volatile organochlorines like trichloroethylene and tetrachloroethylene were detected nationwide in groundwater taken from industrial and urban areas. These organic chemicals have been utilized as solvents in many hi-tech industries and laundry firms, as well as in domestic usage.

Agricultural practice is also a potential source of groundwater pollution. Many chemicals including fertilizer are applied to cropland to raise agricultural production, and consequently constituents of fertilizer are discovered in groundwater in such areas. In particular, nitrate groundwater pollution is becoming a major environmental issue in Japan as well other many developed countries.

Large-scale groundwater pollution due to hazardous chemicals was first discovered in a study by the Japan Environment Agency (JEA) in 1982. This revealed pollution by organochlorines like trichloroethylene and tetrachloroethylene. The survey was conducted to explore the groundwater pollution situation with regard to 18 substances in 1360 well waters sampled in 15 cities across the country. Following the results of the nationwide survey, many local governments carried out their own investigations on the state of groundwater pollution.

Systematic groundwater pollution surveys have been implemented regularly since 1989 on the basis of the Inspection Program of Water Quality under the Water Quality Control Law. This program comprises three types of studies:

1. a general inspection survey to investigate the state of pollution by dividing the survey area in a mesh grid, for example of 5km x 5km, and sometimes smaller,
2. a survey of the well waters in the surrounding locations to determine the spread of the pollution that has been discovered, and
3. groundwater pollution monitoring to follow up the variations in the pollution conditions over time.

Some hazardous chemicals, including halogenated hydrocarbons, are not easily decomposed in the environment and tend to remain for a long period in water, in particular in the subsurface zone. In this context extensive remediation technologies, including both conventional and imported innovative techniques, have been developed and evaluated for determining and optimizing remediation schemes. This chapter describes the present state of soil and groundwater pollution in Japan, and the results of remediation applied so far.

2. Present State of Groundwater Pollution

In Japan in 1974 the presence of trichloroethylene was first detected in well-water of the Tokyo Metropolitan area. In 1981, the serious state of groundwater pollution in the so-called Silicon Valley, California, USA was exposed. Considering the groundwater pollution in USA and other developed nations, the Japan Environment Agency (JEA)

started nationwide groundwater pollution surveys for organochlorines in 1982. The total number of water samples analyzed was 1499, of which 1083 samples were from shallow groundwater, 277 samples were from deep groundwater and 139 samples were from surface water. Chemical analyses were done mainly for volatile organochlorines and nitrate/nitrite nitrogen. Detection frequencies (see Figure 1) for the chemicals are as follows:

1. Of the eighteen chemical analysed for, nitrate/nitrite nitrogen was detected most frequently. The highest concentration of nitrate nitrogen was 80 mg/L. In addition, 10% of all well-water samples (1360) exceeded the drinkable limit for nitrate nitrogen, which is 10 mg/L in Japan and 11.3 mg/L according to the WHO (World Health Organization) guidelines.
2. In addition to nitrogen, trichloroethylene, tetrachloroethylene and 1,1,1-trichloroethane were also found with high frequency in well-waters,
3. Among these three organochlorines, trichloroethylene and tetrachloroethylene were detected in about one of every three well-water samples. Three to four percent of the 1360 well-water samples exceeded WHO guidelines for drinking water for both chemicals.

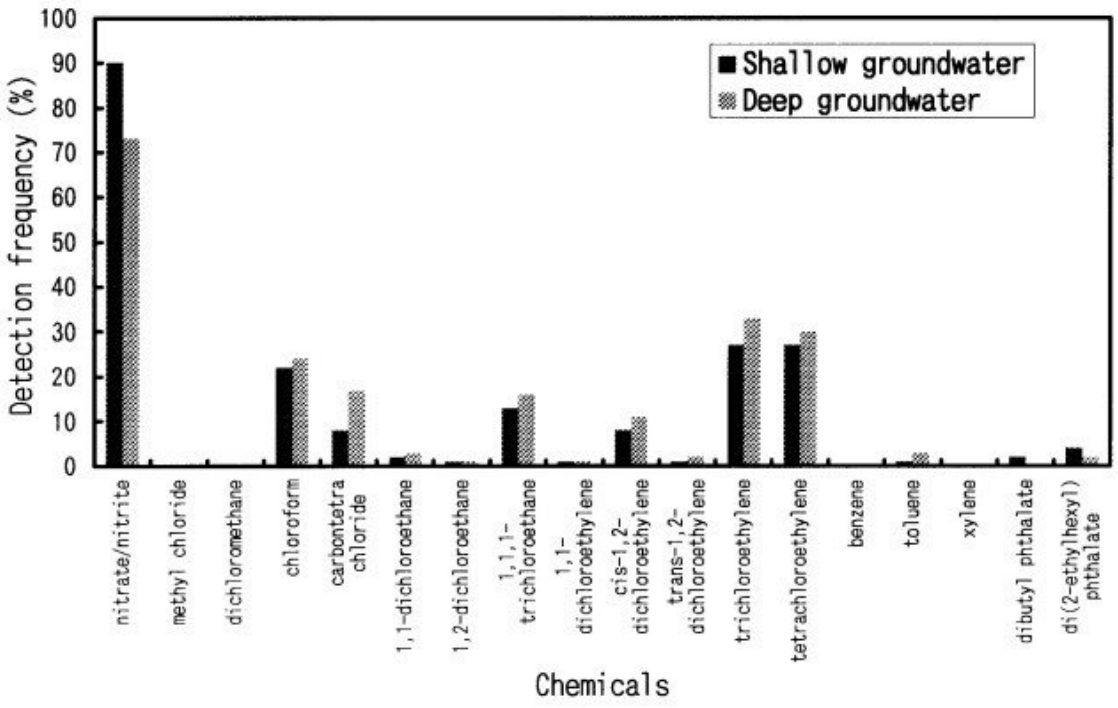

Figure 1. Detection frequency of chemicals discovered in nationwide groundwater survey in 1982

The results of the nationwide survey in 1982 became a trigger to accelerate the partial amendment of the Water Quality Control Law by JEA in 1989 (see *Protection of Freshwater Resources*). Consequently any water containing hazardous chemicals including trichloroethylene and tetrachloroethylene is now prohibited from being injected and recharged into the subsurface environment. In addition the environmental quality standard for groundwater was established by JEA in 1997, and now 26

chemicals are designated. For example, the permissible levels for trichloroethylene, tetrachloroethylene and nitrate nitrogen are 0.03, 0.01 and 10 mg/L, respectively.

A general inspection survey for groundwater pollution has been conducted across the nation since 1984. By 1997, 67 000 groundwater samples had been collected. Results for three chemicals exceeding groundwater standard are illustrated in Figure 2. Data came from a general inspection survey, where the sampling sites in the mesh grid and the mesh grid itself are basically different each year.

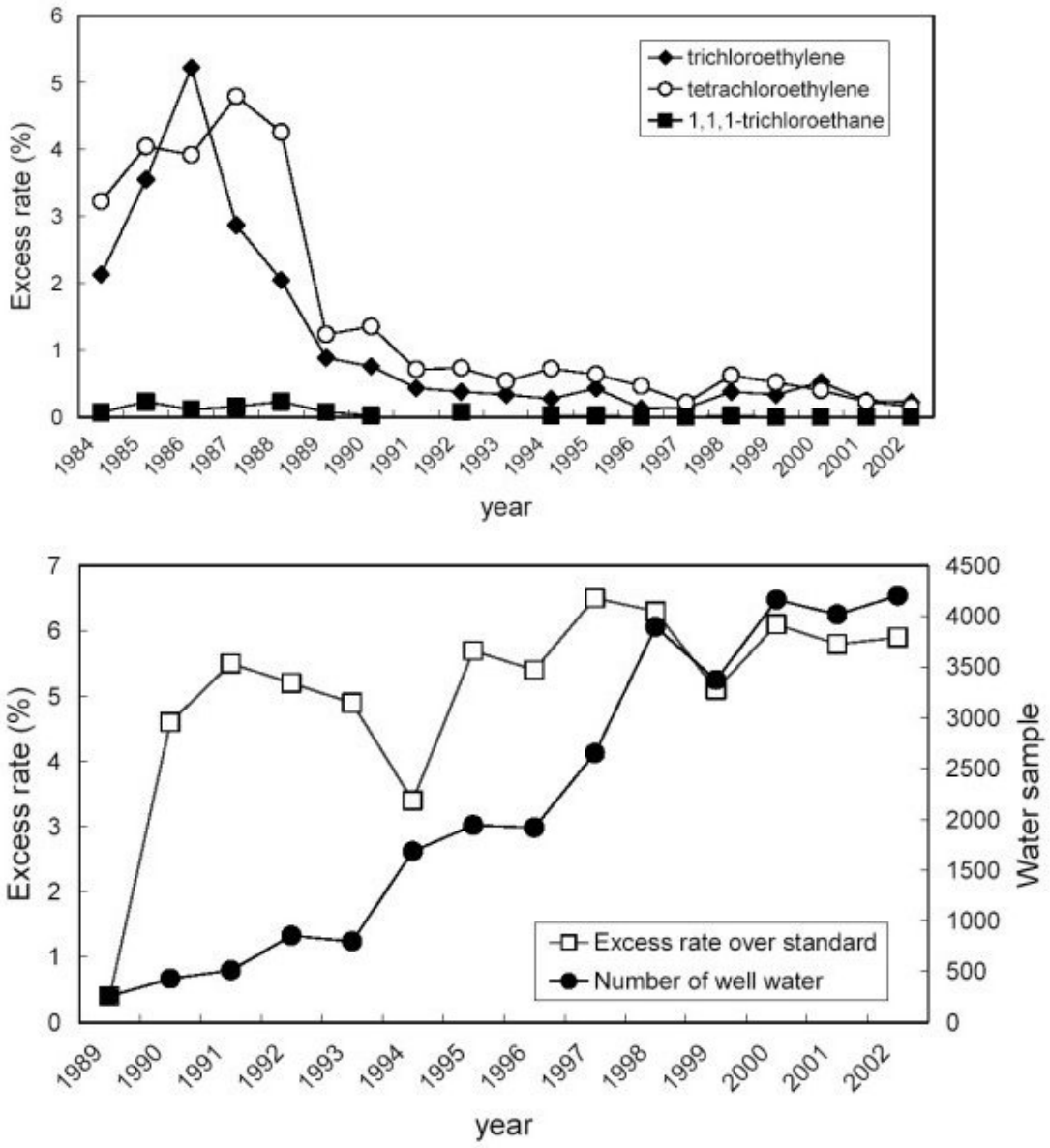

Figure 2. Temporal trend of excess rate of well water samples over the environmental quality standard for groundwater

No investigation sites were duplicated at all. Since 1989, the incidence of excess rates over the standard has been reduced to 0.3 through 0.6% of groundwater samples. However, this is totally attributed to the fact that the original nationwide surveys

focused on areas with a high risk of pollution, such as those surrounding industrial-commercial users of the chemicals. Now the surveys have been extending to residential areas and the countryside with a less potential pollution. Thus, the results have been diluted by changes in the monitoring design, rather than any real improvement in the pollution levels.

Figure 2 also depicts the excess rate of nitrate nitrogen, the sample size of which is considerably less than organochlorines. This is because nitrate nitrogen is a very important substance for the drinking water standard; however, until 2002 it was designated as a provisional substance for the groundwater standard. The result shows that the excess rates keep steadily around 5% every year with seriously high values compared to the results of organochlorines at the initial stage of the nationwide groundwater survey. This is totally attributed to the nitrogen supply source being dispersive as a result of fertilizer application on arable farmland.

-
-
-

Bibliography

Egusa, N, Hirata, T., Fukuura K. and Matsushita, T. (1998). Effect of air migration on recovery contaminant concentration in groundwater. *J. Japan Ass. Groundwater Hydro*., 40(4). 417-428 (in Japanese). [This paper describes air sparging coupled with soil vapor extraction and proves its effectiveness as a remediation technology for organochlorine contamination]

Hirata, T., Egusa, N. , Nakasugi, O., Ishizaka, S. and Murakami, M. (1998). Cost efficiency of subsurface remediation using soil vapor extraction and groundwater extraction. *Wat. Sci.Tech*., 37(8). 161-168. [This paper compares cost efficiency, taking as an example 1 kg trichloroethylene removal, between groundwater extraction and soil vapor extraction]

Hirata, T., II, H., Tase, N., and Nishikawa, M. (1997). Nitrogen isotopes in groundwater as indicator of land use. *27th IAHR Congress, US, California.* 271-276. [This paper provides characteristics of stable nitrogen isotopes in groundwater observed under various sorts of land use]

Hirata, T. and Nakasugi, O. (1992). Groundwater pollution by volatile organochlorines in Japan and related phenomena in the subsurface environment. *Wat. Sci. Tech*., 25(11). 9-16. [The migration features of organochlorines in subsurface environment are described on the basis of a nation-wide groundwater pollution survey in Japan]

Hunt, J.R. and Sitar, N. (1988). Nonaqueous phase liquid transport and cleanup. 1. Analysis of mechanisms. *Wat. Resour. Res.* 24(8). 1247-1258. [This paper describes the migration process and mechanism of undiluted solvents in porous media]

II, H., Hirata, T., Matsuo, H., Tase, N. and Nishikawa, M. (1998), pH and chemistry of nitrogen, phosphate, sulfur and aluminum in surface water near tea plantation. *J. Environ. Systems and Eng.* JSCE 597. 57-63. (in Japanese). [Migration of inorganic substances like nitrogen was observed and analyzed during acidification process due to inorganic fertilizer application to a Japanese tea plantation]

Imamura, S, Negishi, M., Shimomura, M., and Lee, C.S. (1998). Groundwater remediation by reactive

barrier using zero-valent iron. *Proceedings of the Seventh International Symposium on Semiconductor Manufacturing.* 484-487. [Capability of groundwater remediation by reactive barrier is investigated using numerical model]

Kohl, D.H., Sheare, G.B. and Commoner, B. (1971). Fertilizer nitrogen: Contribution to nitrate in surface water in a corn belt watershed. *Science.* 174. 1331-1334. [This work was a pioneer in isotope analyses, taking nitrogen as an example and verifying its effectiveness]

O'Hannesin, S.F. and Gillham, R.W. (1998). Long-term performance of an in situ "iron wall" for remediation, *Ground Water*, 36(1):164-170. [This paper describes a field experiment with granular iron for in situ degradation of dissolved organochlorines and provides an available record of five yeas performance]

Oyamada, H., Kitagawa, M., Miyake, Y., Higuchi, T., Kadokura,. N., Nakayama, F., Oosawa, T., Numata, M. and Mizukami, H. (1995). In situ bioremediation in trichloroethylene contaminated site. *7th Symposium on groundwater and soil pollution.* 75-78. (in Japanese) [This paper describes bioremediation for trichloroethylene in groundwater on the basis of field experiment and numerical calculation]

Senzaki, T. and Kumagai, H. (1988). Elimination of organic chlorinated compounds by reductive process. *Kogyo Yosui.* 257: 2-7. [This paper describes a mechanism of organochlorines degradation by a reductive process]

Terao, H. (1996). Field measurements and management in Kagamihara basin. *Soil and Groundwater Pollution and Management edited by T. Hirata*, Japan Environ. Measurement & Chemical Assoc. 239-245 (in Japanese). [This work describes a possibility of remediation of nitrate nitrogen groundwater pollution by reduction of inorganic fertilizer application]

Uchiyama, H., Nakajima, T., Yagi, O. and Tabuchi, T. (1989). Aerobic degradation of trichloroethylene by a new type II methane-utilizing bacterium, strain M. *Agric. Biol. Chem*. 53. 2903-2907. [This paper experimentally investigates the capability of aerobic biodegradation using methane for trichloroethylene]

Biographical Sketch

Dr. Tatemasa Hirata is professor of Department of Environmental Systems, Wakayama University. He received master and doctor degrees from Faculty of Engineering, Osaka University. Originally he was Civil Engineer. After he joined the National Institute for Environmental Studies which belongs to Ministry of Environment of Japan, he started the career of environmental scientist, specializing water control management. Especially he worked as project manager for soil and groundwater pollution surveys and development and evaluation of remediation technologies. He moved to Wakayama University in 1995 to establish new Faculty of Systems Engineering, and has worked as a member of National Committee of Environmental Standards.