

1 Present State of Groundwater Pollution

1.1 Organochlorines

Following the results of the nationwide survey, a large number of local authorities carried out their own investigations on the state of groundwater pollution with regard to three substances. Until 1995 fifty-nine thousand groundwater samples were collected, and the results made the temporal trends of excess rates for three substances over drinking water standards as illustrated in Fig. 13.1. Data are coming from a general inspection survey to investigate the state of pollution by dividing the survey area in a mesh grid, and investigation sites are not duplicated at all. Since 1989, the statistical incidence of excess rate for standards has reduced to be 0.3 through 0.6 % at present, however, this is totally attributed to the fact that at the initial stage the nationwide surveys focused on areas with a high risk of pollution such as those surrounding industrial-commercial users of the chemicals and now are extending to residential area and country side with a less potential of pollution.

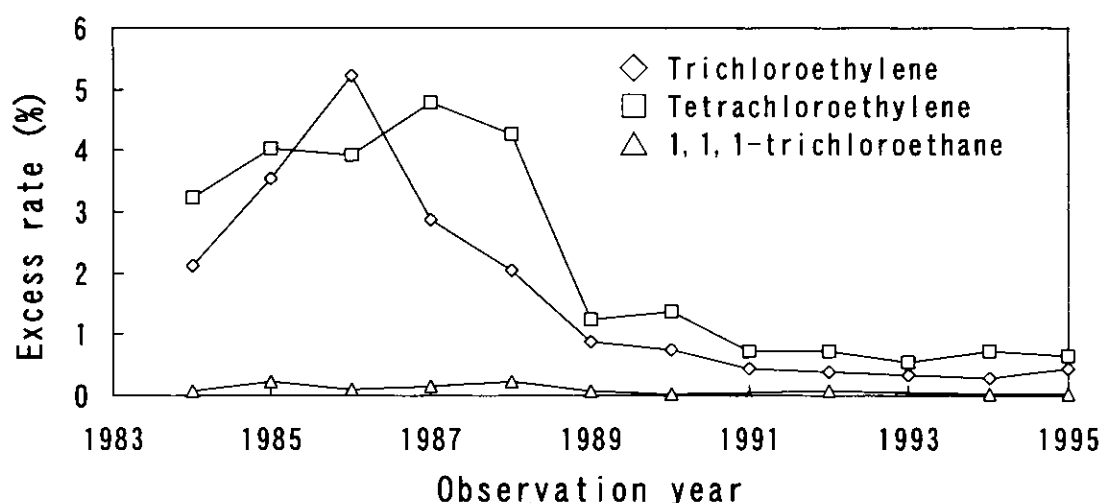


Fig.13.1 Temporal trends of excess rates for trichloroethylene, tetrachloroethylene and 1,1,1-trichloroethane over drinking water standards.

1.2 Heavy Metals

The groundwater pollution due to heavy metals has been systematically surveyed since 1989. The results show that except PCB, alkyl mercury and organic phosphorus, heavy metal pollution in groundwater was discovered. In particular the concentrations of arsenic and mercury over environmental quality ex-standards for groundwater was detected at high rate as listed in Table 13.1. The arsenic and lead controls were intensified, changing the standards from 0.05 and 0.1 mg/L respectively to 0.01 mg/L each. The subsurface pollution by arsenic and lead is thought to steadily increase in the future. Furthermore heavy metal soil pollution in urban area is also increasing, for example, chromium(VI), lead, mercury and cadmium in plating and chemical industries and research laboratory etc.

The elements of arsenic and chromium are basically contained in natural soil and rock, the average existing amounts of which count for 1.8mg/kg and 100mg/kg, respectively. Therefore

natural pollution are possible to originate from virgin soil and rock. The arsenic groundwater pollution is well known in Bengal district of India to be caused by natural geology. The intensive agriculture practice facilitated taking deep groundwater, which induced shallow groundwater penetration through arsenic mining to deep groundwater zone. Consequently deep groundwater use for drinking water produced human skin disease and cancer.

Table 13.1 Detection rate of heavy metals in groundwater

| Survey year | Detection rate over standards (%) | | | | | Standards (mg/l) |
|--------------|-----------------------------------|------|------|------|------|------------------|
| | 1989 | 1990 | 1991 | 1992 | 1993 | |
| Chromium(VI) | 0.0 | 0.03 | 0.03 | 0.0 | 0.04 | 0.05 |
| Arsenic | 0.26 | 0.16 | 0.14 | 0.18 | 1.44 | 0.01 |
| Mercury | 0.0 | 0.12 | 0.10 | 0.11 | 0.11 | 0.0005 |

Detection rate for arsenic until 1992 is based on ex-standards 0.05mg/l

1.3 Nitrate Nitrogen

The nitrogen origin in water environment is the atmospheric constituent, which is converted to fixed forms by biological or industrial fixation. The biological fixation amount on the globe is estimated to be 4×10^{10} - 1.4×10^{11} kg/y, and the industrial fixation to be 8×10^{10} kg/y, which is approximately equal to the natural abundance fixed. The excess nitrogen over the denitrification capability in the natural field results in nitrogen accumulation in water environment in many ways.

The rainwater contains nitrogen oxides, however, the inorganic nitrogen concentration takes 0.52mg/L and the annual load is estimated to be 9.1kg/ha/y in the averaged value of the whole Japan. The main source of nitrate in groundwater in Japan is coming from the loss of fertilizer application and the soil treatment of the domestic wastewater.

The field measurement shows that the half amount of fertilizer applied in Japan appears to be leaching out to the groundwater. With respect to the arable lands, paddy field is usually in anaerobic condition and annual amount of nitrogen fertilizer applied is within 100kg/ha, so it is rather acting as the reduction of nitrate concentration, compared the input and output loads.

The intensive field surveys have been done in Miyakojima Isle, Okinawa Pref. and Kagamihara basin, Gifu Pref. The drinking water of Miyakojima totally depends on groundwater abstraction. During the past three decades, increase of chemical fertilizer application to the sugar cane caused the groundwater pollution. In addition, soil treatment of the domestic wastewater was accelerating the rise of the nitrate concentration. The steady increase of nitrate concentration is also observed in the whole isle to become nearly four times high from 1.92mg/L in 1966 to 7.58mg/L in 1989 as illustrated in Fig. 13.2.

1.4 Other Chemicals

The environment Agency's 1982 survey led to the detection of three types of aromatic hydrocarbons (benzene, toluene and xylene) and two types of phthalates (dibutyl and di(2-

ethylhexyl phthalates) in groundwater. Four of these substances, except for dibutyl phthalate, were added to the standard for water environment and guideline, but only one well showed the presence of benzene in excess level as compared with their standards.

Benzene, toluene and xylene were tested for in the 1989 inspection survey when only toluene was found to be present, and that was at levels below the standard values. In an incidence in which gasoline had leaked from a tank into ground, aromatic hydrocarbon pollution was observed to have spread, with both benzene and toluene clearly exceeding the standard levels.

In the 1992 inspection survey, 1,4-dioxane, one of the Designated Chemicals under the Chemical Substance Control Law, was discovered at a high incidence. This substance has also been detected at a high rate of incidence in surface waters. The maximum concentration found in groundwater is higher than that in surface water. The maximum level exceeded 0.07 mg/L, a concentration the USEPA considers as being equivalent to a 10^{-5} cancer causing risks. This chemical has a high solubility in water and is therefore difficult to separate from water.

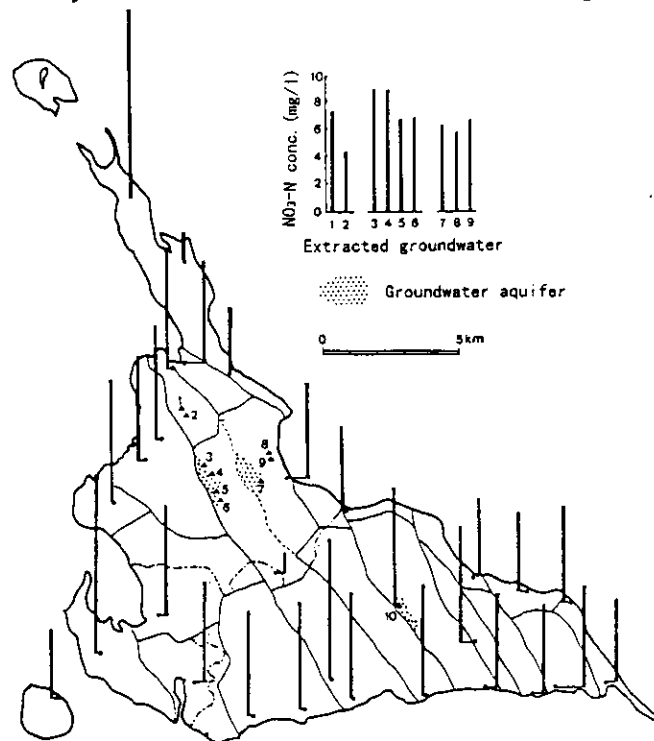


Fig.13.2 Areal distribution of nitrate nitrogen in the groundwater of Miyakojima Isle(1989)

2 Remediation Action for Groundwater Pollution

2.1 Prevention of Groundwater Pollution

The groundwater quality management comprises two parts of prevention from being polluted and pollution remediation. When the groundwater pollution is discovered in regional groundwater, at first the inspection survey is conducted to depict the groundwater pollution plume around the polluted well. In addition, emergency measure is taken place according to the groundwater use. In case that the groundwater is utilized for drinking water, the remediation for contaminant removal from polluted groundwater to meet the drinking water standard or alternative water resource is required. In addition, under the Water Control Law any water containing the

hazardous chemicals is prohibited to be infiltrated to the subsurface environment, and plus the effluent concentration of hazardous chemicals into public surface water is determined not to be over ten times the water quality standards.

2.2 Classification and Availability of Technologies

The pollutant entering into the soil and groundwater results in the following conditions, depending on its chemical feature: (1) adsorption on the soil particles; (2) dissolution in the water; and (3) vaporization in air of soil pore if the pollutant is volatile substance. When pollutant enters the soil in large quantity as undiluted liquid, another possibility exists, namely (4) that the pollutant occupies the pore spaces in undiluted liquid form. In any of existing forms, the determination of concentrations is totally attributed to partitioning characteristics among the gas, liquid and solid phases. When the distribution is biased toward any particular phase, the most effective way to repair the pollution situation is to eliminate the high-concentration medium.

Remediation technology for subsurface pollution consists of removal of pollutants from the soil or groundwater and their detoxification. Generally it is classified into two categories i.e. (1) diffusion control of the pollutants and (2) removal and decomposition of pollutants as collected in Fig. 13.3. In the pumping up of groundwater, diffusion control comes about with the use of a barrier well. When it takes place close to the pollutant source, the element of pollutant removal technology predominates. These measures are thus closely interrelated and should be used in a proper combination to achieve an improved remediation effect.

Solidification technology as one of these techniques is not applicable to such liquid pollutants as trichloroethylene. Chemical reaction and vitrification techniques, however, are primarily reserved for heavy metals. Microbial decomposition is in widespread use as a decontamination and detoxification technique for hydrocarbons such as petroleum and petroleum products which are relatively readily degradable. Recently, however, it has become possible to discover microorganisms capable of decomposing at high efficiency volatile organochlorines. Their use is now being developed for commercial-base application, with the technology having reached the stage of site verification tests.

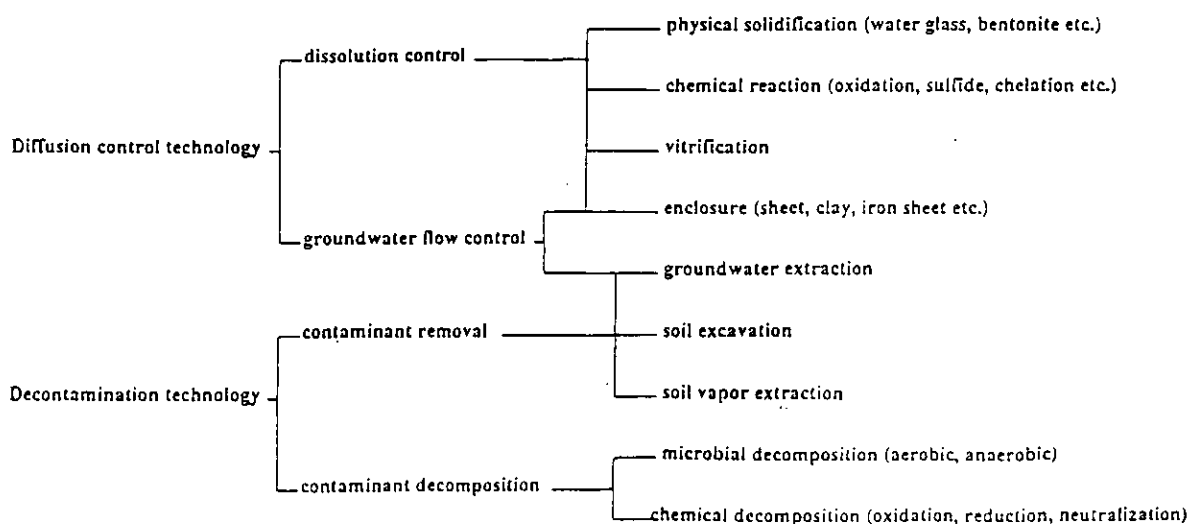


Fig. 13.3 Classification of remediation technology for subsurface pollution

3. Remediation Technology for Groundwater Pollution

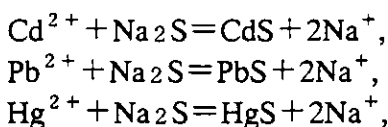
3.1 Heavy Metals

(a) Physical treatment

Various sorts of heavy metals are detected from soil environment, for example, copper, cadmium, arsenic from arable land, and mercury, cadmium, lead, arsenic from urban area. Basically heavy metals in soil are not easily soluble with water. In addition the cation like cadmium is likely to be adsorbed on the surface soil, so physical treatments of ploughing with deeper soil, placing clear soil, in-situ solidification, enclosing into water-tight container are used so far. In the arable land, the area of 6140 ha was planned to be treated mainly for cadmium pollution, and 80 % of the area is already established until 1994. The inquiries of Japan Environment Agency in 1994 showed that the technology of solidification and enclosure occupies 30% of 318 soil remediation practices, and soil placing is up to 16%.

(b) Chemical treatment

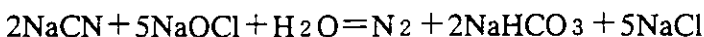
The solubility with water is quite different with chemical structure of heavy metals, and this property was utilized for remediation as oxidation or reduction technologies. For example, the sodium sulfide is added to cadmium, lead, mercury compounds in order to reduce the water solubility,



chromium(VI) is reduced to chromium(III) with iron sulfate, lignite, poultry manure etc.



Cyanide compounds is oxidized with sodium hypochlorite as follows.



The success of chemical treatment mentioned above depends on how long time the chemical reaction continues. In this context after the chemical treatment, extraction test with water from treated soil is conducted, and when the solution concentration is over the standards, the treated soil should be controlled in the watertight container.

After the soil treatment by physical enclosure and chemical reaction, the contaminants still stay in the in-situ soil. Taking an example on the chromium pollution in Tokyo Metropolitan, the soil and groundwater pollution reappeared after the effective period of chemical reaction. In place of these conventional methods, innovative technologies for contaminant detoxification and removal from polluted site have been developed. The vitrification technique of contaminated soil is used, mainly in America, for heavy metals and radio active material, controlling high current electricity in the soil. The vitrification technique is capable of closing the contaminants within glass solidification. The soil washing and classification are thought to remove highly contaminated parts from soil excavated. In addition, when the extraction test with water meets the

environmental standards, the treated soil is able to get back to the original position.

There exists a remediation practice in in-situ groundwater contaminated with chromium(VI). Fig.13.4 shows a remediation system, where the remediation wall of 6 m depth was installed, and lignite and acid clay were placed in the bottom of the wall in order to reduce chromium(VI) in perched groundwater. This groundwater treatment was performed as an emergency measure until contaminated soil was removed.

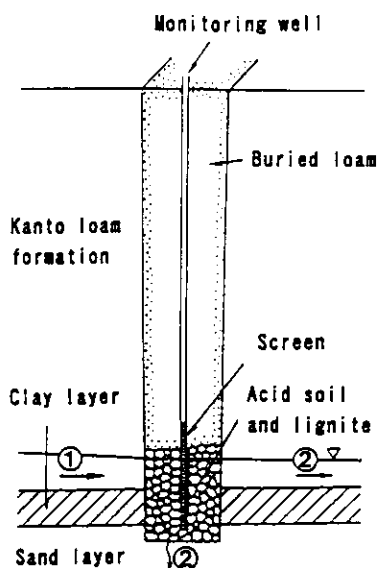


Fig. 13.4 Remediation wall for chromium(VI) treatment in perched groundwater. The number in a circle ① is for inflow of contaminated groundwater and ② for outflow of treated groundwater.

3.2 Organochlorines Pollution

(1) Pollution features

The volatile organochlorines like trichloroethylene and tetrachloroethylene are widely and effectively used in cleansing process by many and various industries producing huge amounts of fine products. They tend to be little adsorbed on soil. In addition the organochlorine liquids are heavier in weight and less in surface tension and viscosity than water, therefore, they are rather mobile in unsaturated soil compared to water.

Table 13.2 gives the maximum values for the soil and groundwater concentrations collected so far. In some cases with concentrations in the range of a few tens to a few hundreds of thousands of mg/kg, the pollutant is present in the soil in the form of undiluted liquid. The vertical soil concentration profile exhibits that while maximum pollutant concentrations occur at different depths, in many cases the pollutant tends to raise its concentration at or near the gravel/clay layer boundary at the bottom of the aquifer.

Also in groundwater the concentrations for trichloroethylene and tetrachloroethylene overshoot their solubility in water of 1100 and 150 mg/l respectively. It is reasonably assumed that the organochlorines form the pools of undiluted liquid mainly in the vicinity of the pollutant source and are present in groundwater at high concentration nearly at their solubility in water. In the

presence of undiluted liquid, the concentrations measured for the soil gases should be equal to their vapor pressure, i.e. trichloroethylene:76300ppmv and tetrachloroethylene:18400 ppmv. Actual measurements for soil gas concentrations have shown evidence of high concentration of soil gas in excess of 10000 ppmv.

Table 13.2 The maximum concentration of organochlorines in soil and groundwater in pollution incidents of Japan with their soil feature

| Locations | Solvent | Depth (m) | Soil feature | Soil conc. (mg·l ⁻¹) | Groundwater conc. (mg·l ⁻¹) |
|-----------|---------|-----------|--------------|----------------------------------|---|
| TK-A | TCE | 7 | Clay | 64 | 11 |
| | cis-DCE | 7 | | 39 | 19.2 |
| TK-B | TCE | 11.5 | Silt | 0.2 | 1.9 |
| | cis-DCE | 6.3 | | 3.43 | 5.5 |
| T1 | TCE | 2 | Silty sand | 6.600 | 42 |
| | TCE | 37 | | 5.4 | 103 |
| KT-U | TCE | 25-27 | Silt | 40 | 360.000 |
| KT-K | TCE | 0.7 | Surface soil | 10 | >10 |
| KM-K | PCE | 5.5 | Silt | 360 | 80 |
| KM-T | TCE | 46 | Silty sand | 138 | 294 |
| SZ-A | PCE | 2.1 | Coarse sand | 8.100 | 8.6 |
| SZ-B | PCE | 0.8 | Clay | 25.000 | 22 |
| IT-S | TCE | 3 | Coarse sand | 232 | 1.390 |
| IT-M | TCE | 7-8 | Silty sand | 210.000 | 40 |

Solvents are: TCE – trichloroethylene, PCE – tetrachloroethylene, and, Cis-DCE – cis-1,2-dichloroethylene.

(2) Remediation technology

(a) Soil Excavation

In many pollution incidents of shallow groundwater, the countermeasure of soil excavation is first considered. In 1983 during monitoring the quality of groundwater, in particular used for public water supply, volatile organochlorines were found here and there in regional groundwaters. At the upper part of the region, an electric firm, one of the biggest world enterprises, is in operation, utilizing trichloroethylene to clean the component parts of television and semiconductor. Immediately after detecting trichloroethylene in water for the public supply, surrounding shallow well-waters of 427 points for domestic usage were investigated, revealing that the maximum concentration in the shallow groundwater was up to 10mg/L just down from the firm, and the trichloroethylene plume over 0.03mg/L covered the area of more than three kilometers toward the down stream gradient.

In order to solve the pollution mechanism, borehole investigations were conducted inside the firm site, and consequently highly polluted groundwaters were collected around the building installing the trichloroethylene groundwater pollution storage tank situated at the southeast part of the firm area. The trichloroethylene concentration in groundwater rose to 40mg/L at 7m depth and 4mg/L at 10m depth.

At first the firm shifted the solvent to 1,1,1-trichloroethane, and evacuated polluted soil of more than 1mg/kg in concentration underlying the storage tank, of which amount counted for 1,000m³ in volume. The trichloroethylene highest concentration in soil reached 6,600mg/kg at 2m depth. Immediately after that, the concentration in shallow groundwater taken from the well near the

building began to markedly decline, however, no discernible decrease was gained in deep groundwater, as illustrated in Fig.13.5. The implication is that the soil removal operation was forced to terminate due to groundwater appearance at the depth of 7m, so appreciable quantity of contaminant is still present within the deep aquifer. Consequently seventeen tons of trichloroethylene were removed from 1984 to 1989 by means of groundwater extraction.

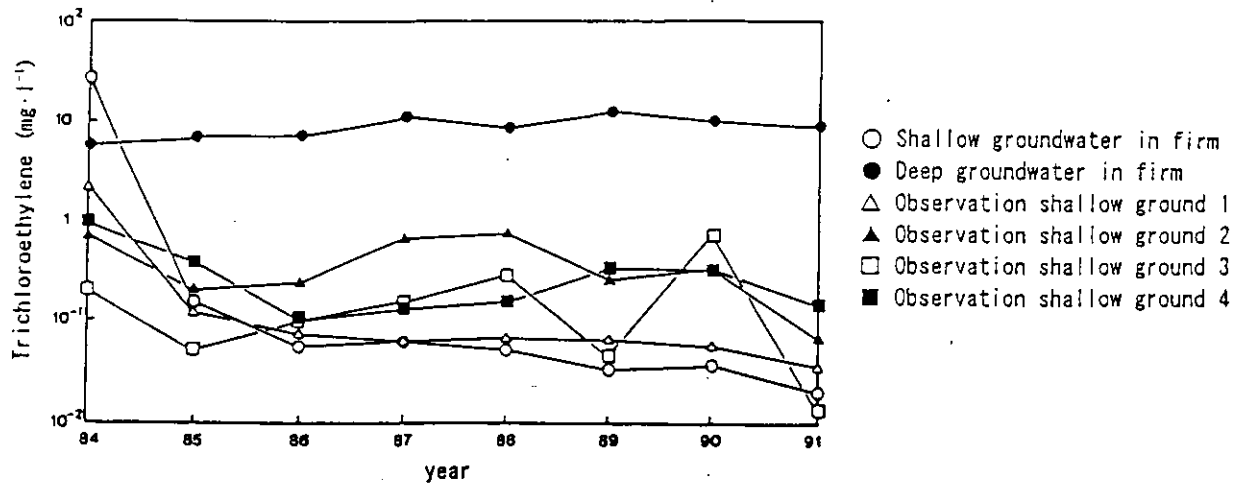


Fig. 13.5 Time-varied changes of trichloroethylene concentration in shallow groundwater after polluted soil excavation and subsequent groundwater extraction for remediation

(b) Dual Extractions of Soil Gas and Groundwater

Soil vapor extraction technique is to remove pollutants, which have vaporized in the soil vapors in the vadose zone. The remediation technique is reported to have removed a few hundred kilograms to one ton volatile organochlorines in operations for a period of a few months to a year. However it is available only for vadose zone, so that when the groundwater is also polluted, the operation to pump up groundwater is inevitable.

In a shallow groundwater region where the highly residual part of trichloroethylene is existing in both sides of vadose and groundwater zones, the soil vapor extraction method was employed for vadose zone remediation with pumping up the groundwater at the same time. In order to extract both of soil gas and groundwater from a same remediation well, four dual extraction wells were constructed around the hotspot for the soil gas flow reach to cover the highly polluted area (Hirata and Nakasugi, 1993). The remediation operation using two technologies resulted in removing the trichloroethylene amounts of 472 kg by soil vapor extraction and 1764 kg by groundwater extraction during the period of 27700 hours (1154 days).

Fig. 13.6 compares the time-varied changes of the trichloroethylene removal rate in the unit of kg per hour. Both removal rates indicate the total rate of the four extraction wells. It appears that at the initial stage of the remediation, the soil vapor extraction operation has been successful in removing by suction 1 kg/hr of trichloroethylene, which shows one order higher than the removal rate achieved with groundwater extraction. Yet the removal rate due to soil vapor extraction does begin to decline much earlier than in case of groundwater extraction, so that the removal rates of

these two methods develop inversely with the progress of remediation. In particular the removal behavior of pollutant totally depends on the chemical feature of trichloroethylene liquid being highly volatile and little soluble in water (Hunt and Sitar, 1988) and the change of existing form of pollutant affects the effectiveness of applied remediation technologies with the operation time going on. The groundwater extraction requires so many years to cleanup the polluted aqueous phase, contrary to this, it is likely to eliminate much more pollutant with groundwater extraction than other technologies.

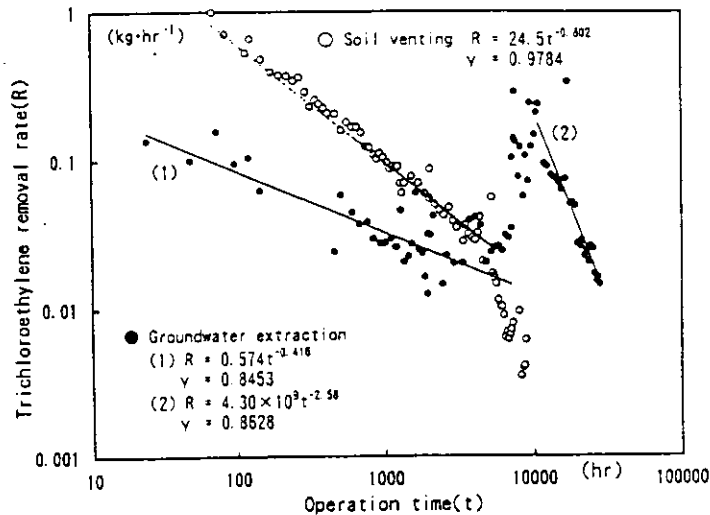


Fig. 13.6 Comparison of trichloroethylene removal rates due to soil vapor and groundwater extractions. The numerals a and b in the figure denote the constants, when the removal rate R is formulated with the operation time t by $R = at^b$.

(c) Bioremediation

Physical remediation technologies such as soil vapor extraction and groundwater extraction involve the removal of pollutant substances from the subsurface zone and their ultimate vaporization and recovery by adsorption on activated carbon. Though these techniques are ideal for the treatment of low-boiling point chemicals, the issue still remains that the solutions desorbed from the activated carbon and the activated carbon itself will need to be eventually disposed of by incineration treatment. This implies that in some sense, the situation is that of a merry-go-round of the pollution burden which is shifted from the subsurface to the atmospheric environment. In contrast to this, bioremediation technology holds out significant hopes for the possibility and practical availability of a process for the complete decomposition of the pollutant substances to carbon dioxide.

Biodegradation is already in practical use in the industrialized Europe and America as a remediation technology for relatively easy-to-decompose hydrocarbons such as gasoline. Similarly, for substances such as trichloroethylene, it has been possible to discover microorganisms that are capable of decomposing pollutants at high efficiency under anaerobic conditions. In situ Bioremediation gives rise to certain problems, which need to be resolved such as the issues of the toxicity of the intermediate products and the public acceptance of the technique. Bioremediation is a promising measure in the final stage after the implementation of

physical remediation procedures such as soil vapor extraction and groundwater extraction technologies. In Japan, site verification of bioremediation technology for volatile organochlorines has now commenced with a view to developing it for practical application. Fig. 13.7 illustrates a result of the test for in situ bioremediation for trichloroethylene groundwater pollution, injecting oxygen and methane into polluted groundwater. It proves that the groundwater concentration for trichloroethylene keeps nearly zero value, even after the injection of oxygen and methane stopped. During 40 days through 40th day to 80th day from the onset of the test, the amount of 1.2kg trichloroethylene is estimated from the mass balance calculation of groundwater flow to be decomposed.

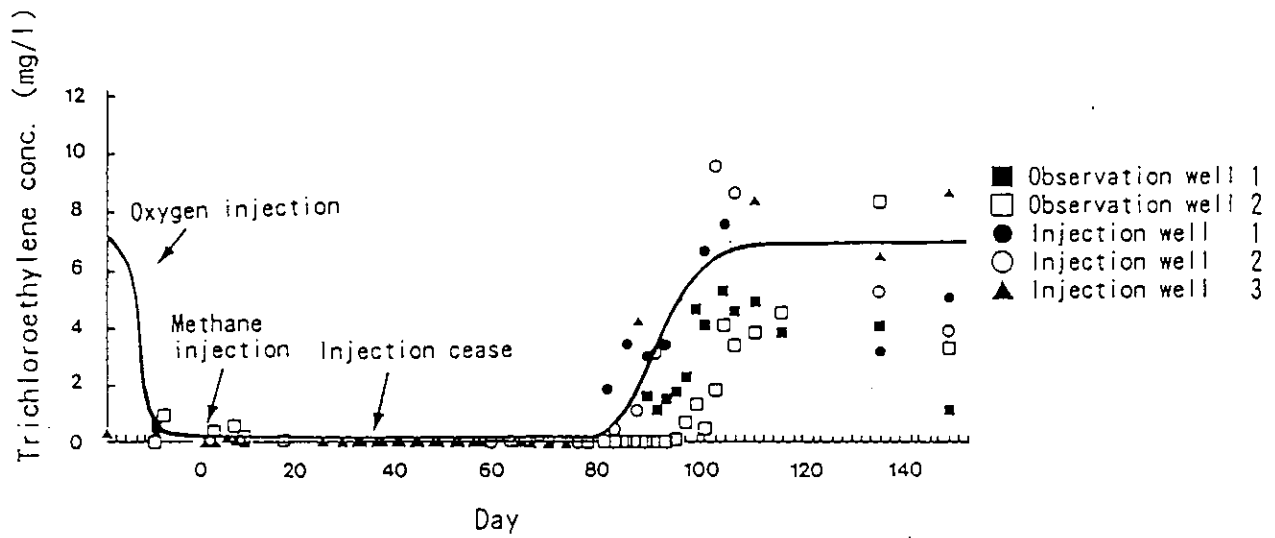


Fig. 13.7 In situ bioremediation for trichloroethylene pollution in shallow groundwater

3.3 Nitrate Pollution

When the nitrate concentration in groundwater is in steady upward trend and about to approach or exceed the drinkable limit (10mg/L in Japan), the easiest measure is to close the groundwater abstraction. In addition, making deeper borehole to take clean groundwater and blending with less contaminated water etc. are introduced. In case of no proper water resource except polluted groundwater, the drinkable water treated with the techniques like ion exchange method is supplied.

In order to conserve and prevent the groundwater from being polluted, it is urgently desired to reduce the leaching loss of nitrogen in agricultural practice and how to reuse and repair the groundwater with high nitrate concentration. In many cases of the nitrate pollution in groundwater, however, it initiates in leaching out the fertilizer constituent from non-point source. The definite measures applicable to the various sorts of nitrate problems are not easy to be discovered and determined. Several options about this are picked up as follows;

- 1) predictions of nutrient need and timing of inorganic and organic fertilizer use, and application of proper amount of fertilizer,
- 2) avoiding the unnecessary irrigation which promotes the leaching out of fertilizer applied in cropland field,

- 3) establishing the crop rotation introducing pea and bean,
- 4) utilizing organic fertilizer and introducing slow-acting fertilizer in place of some parts of chemical fertilizer,
- 5) reuse of groundwater from upgradient to downgradient,
- 6) careful management of ploughing-in in wet season, and
- 7) use of uncultivated paddy field to remove the nitrate due to the denitrification capability.

In Kagamihara City, the whole potable water of which is coming from groundwater, the nitrate concentration in shallow groundwater tends to rise since 1970s. The city authority, known as carrot production, has studied the management of fertilizer use to recover the groundwater quality. The field experiment showed that the 25% reduction of fertilizer application(300kg-N/ha/y) compared to the usual one(400kg-N/ha/y) is able to produce nearly the same amount of carrot without damaging the carrot quality and harvest amount. As a result of improvement of fertilizer use commenced at early 1990s, the groundwater in carrot field located in the east part of the city, where the nitrate nitrogen concentration overshoot 25mg/l, began to exhibit a bit decline in the nitrate concentration and heavily polluted area as illustrated in Fig. 13.8.

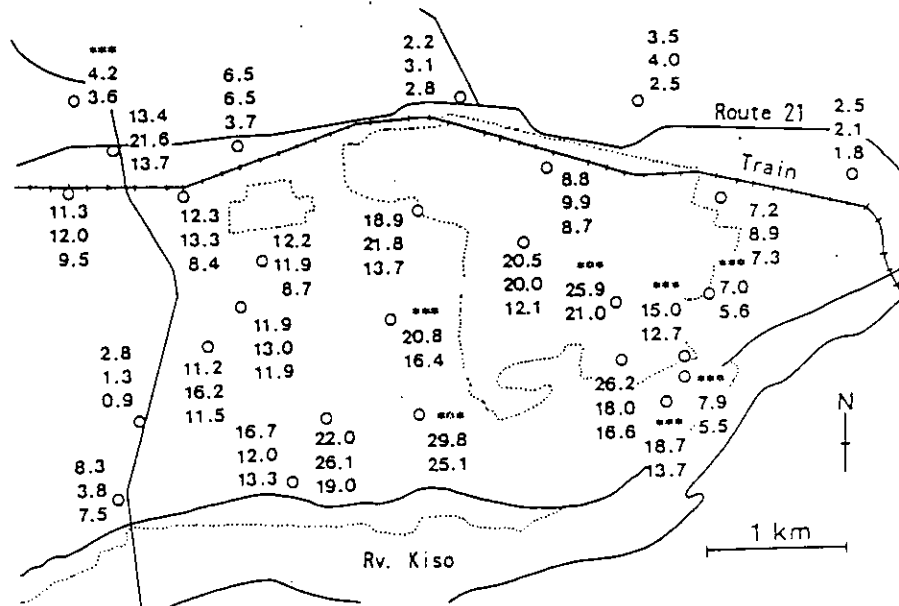


Fig. 13.8 Changes of nitrate nitrogen concentration in shallow groundwater observed in Kagamihara basin. Three values in the figure denote the nitrate nitrogen concentrations observed in July 1984, July 1990 and July 1994 respectively from the top.

4 Summary

For the remediation of soil and groundwater pollution by volatile organochlorines, innovative technologies are being developed and introduced, and their effectiveness in pollution removal has been demonstrated. Some technologies are naturally still in the in-situ or indoor verification stage as is the case with bioremediation using microbial activity. In general, however, it is emphasized that remediation technology has been approximately established. It is definitely possible to achieve polluted soil and groundwater remediation and restoration with the expenditure of large amounts of time and money. Remediation measures have thus been enforced in practice on large-

scale operating sites. Yet, in many pollution cases, the operating sites are small in scale, and cost is the major problem in the implementation of remediation.

To facilitate positive advances in remediation, it is necessary to develop suitable survey methods and efficient pollutant removal technologies. With the benefit of soil and groundwater pollution surveys, it should definitely be possible to carry out effective measures on the basis of preliminary pollution survey. The fact does remain, however, that there are limits as to the funds that can be expended in remediation. It will therefore also be significant to strive for a proper balance in the whole package of remediation operations. The need is to establish a completely integrated package of remediation measures starting with survey programs, including measures such as the planning of feasible remediation operations consistent with the use of the groundwater and also including a clear definition of the objectives of the remediation program.

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