NATURAL AND ARTIFICIAL MERCURY DECONTAMINATION – OTTAWA RIVER AND MINAMATA BAY (YATSUSHIRO SEA)

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ABSTRACT

Everyday, man creates new processes and materials whose properties are not fully known and are sometimes toxic to him and the environment. One of the worst cases was in Minamata Bay, Japan where a fatal accident occurred due to mercury pollution. The accident was unique in many aspects thus forcing the establishment of a new decontamination process for polluted areas. Two mercury polluted sites, the Ottawa River, Canada and Minamata Bay (Yatsushiro Sea), Japan, have been investigated to establish a fundamental approach for decontamination with special emphasis on natural and artificial processes and their problems. Artificial decontamination on bottom sediments has primarily cleaned-up Minamata Bay and the near-by Yatsushiro Sea considerably, an acceleration of the natural processes by 31.5 years. The surrounding area will be fully cleaned-up by natural forces, following the artificial work, by 2011 AD (or 20 years from now). This means that the natural decontamination has a half-life of 9.5 years in Yatsushiro Sea. For the Ottawa River, only natural processes were applied to clean-up the river system. It took 5 years for physical components to be decontaminated (a half-life of 1.20 years) while it took a longer period for biological components. This last finding was also true for Minamata Bay. This delay may be due to the longer life span of the biota and their efficient processes of bioaccumulation from diluted surrounding water. Based on the findings herein, a combination of both artificial and natural decontamination methods is recommended in consideration with the given environmental conditions.

KEYWORDS

Ottawa River, Minamata Bay, Yatsushiro Sea, Mercury Pollution, Mercury Decontamination.

INTRODUCTION

Man, forever industrious, has been creating new and more efficient processes and materials whose properties are not fully known and sometimes toxic to him as well as the environment. Lack of understanding in environmental behaviour may unintentionally allow toxins to exceed the limits of the natural self-purification processes and create serious environmental pollution. One of the worst cases of pollution was in Minamata Bay, Japan where a fatal accident occurred due to a release of industrial wastewater containing minute amounts of mercury.

The Minamata mercury accident was unique in many aspects: (a) it was the first known case of
numerous deaths as a result of natural bioaccumulation of a toxic material, methylmercury, in a diluted industrial wastewater. In other instances, deaths were followed by direct consumption of contaminated water. (b) The Minamata toxicant was partly produced from relatively harmless compounds (HgCl₂ and HgSO₄) by natural processes and the mechanisms for these reactions were entirely unknown to man until this accident (Jensen and Jernelov, 1968). (c) Methylmercury maintains an equilibrium concentration in the environment and is thus persistent, not disappearing quickly with time (Kudo et al., 1977), while most toxicants such as chlorinated phenols are biodegradable and gradually disappear from the scene. Hence, persistent hazardous materials, such as long half-life radioactive materials, cannot be handled by traditional methods in the environment.

The problem is how to clean up these polluted sites (areas) contaminated with persistent hazardous materials safely, efficiently, economically, and with agreed upon ways. The environmental, social and economical conditions, however, vary considerably from one site to another and therefore, there would be no single universal method for the clean-up. However, there must be some fundamental approaches for the decontamination based on the long term behaviour of the toxins in the environment. The knowledge and understanding of their behaviour, in turn, would establish the most safe, efficient, economical and acceptable means for the clean-up.

Two entirely different locations, the Ottawa River, in Canada, and Minamata Bay (Yatsushiro Sea), in Japan, Fig. 1, have been polluted by mercury but recently they are considerably cleaner. Fortunately, both sites have been investigated for the movement of the mercury for more than a decade. Of course, the two sites are completely different in almost every aspect such as geographically, geologically, climatologically, hydraulically and so on. Even the decontamination method was different: only a natural method was applied to the Ottawa River while a combination of artificial and natural methods were used for Minamata Bay.

![Fig. 1. Locations of the Ottawa River, Canada and Minamata Bay (Yatsushiro Sea), Japan. (Carrying a log boom to a pulp mill at the Ottawa River. Low tide at Minamata Bay.)](https://iwaponline.com/wst/article-pdf/26/1-2/217/16413/217.pdf)

This paper reports (a) the behaviour of mercury in the Ottawa River and Minamata Bay, (b) the decontamination processes used at both sites, and (c) a proposal of fundamental approaches for clean-up and identification of practical problems establishing safe, efficient, economical, and acceptable clean-up methods for a site once polluted by persistent hazardous materials.
Ottawa River and Study Section  The Ottawa River is approximately 1113 km long, and the average annual flow rate is 1700 m$^3$/s (about the same size as the Rhine River in Europe). The River originates in northern Ontario and eventually joins the St. Lawrence River. Its drainage area is 148,000 km$^2$, and the total precipitation in the area averages 864 mm/year. The snowfall contributes about 25% (or 212 cm/year) of the total precipitation. Formerly, the ratio of maximum to minimum recorded flow rate was as high as 12:1, but the construction of a series of dams in recent years has reduced this ratio significantly (at present, 4:1). The River has been used extensively to transport timber (log-driving) from upstream forests to pulp and paper mills. It, in turn, has been receiving wastes in the form of wood fragments (fibres, chips, and barks) from adjacent pulp and paper mills for nearly 100 years. This (along with a series of dam constructions) creates extensive organic deposits in the River (up to 4 m in depth). This feature of organic rich sediments, along with the brownish colour of the water due to humic acid contents, is an important characteristic of the Ottawa River.

The pulp and paper mills used various forms of mercury (Hg, HgCl$_2$, and phenyl mercuric acetate) in considerable amounts for the production of sodium hydroxide and for a slimicide. Furthermore, gold prospectors, in the past, used metallic mercury for easy extraction from ores. This mercury along with that of natural origin accumulated in the River system. Though the River has been used for recreation and water supply, it has never supported a significant commercial fishery. However, the use of mercury in pulp and paper producing processes was banned in 1969 because the concentration of mercury in fish frequently exceeded the legal limit of 0.5 ppm, and a warning was issued in 1970 discouraging the consumption of any fish caught downstream from the city of Ottawa.

For the detailed study on mercury, its distribution (geographical, compartmental, and anatomical), transport and transformation, a 5 km stretch (section) of the River was selected as a representative model river system, Fig. 1. This section began about 2 km downstream from the city of Ottawa, the Canadian capital, and was chosen especially for the complicated forms (2 islands make a complicated hydraulic flow pattern) and the varied environments it represented (Ottawa River Study Group, 1977). On average, the study section was covered by ice for a period of 4 months (December to April), and the frost-free period for plant growth was about 3 months in this area. In other words, this area is hot in the short summer (extreme maximum average air temperature is 33.5 °C) and cold in the long winter (extreme minimum average is -29.5 °C). Some of the environmental data which influenced general characteristics in the study section of the Ottawa River are shown in Table 1.

<table>
<thead>
<tr>
<th>Ottawa River (Study Section)</th>
<th>Minamata Bay</th>
<th>Yatsushiro Sea</th>
</tr>
</thead>
<tbody>
<tr>
<td>Location</td>
<td>45 N, 76 W</td>
<td>32 N, 130 E</td>
</tr>
<tr>
<td>Type of Water Body</td>
<td>Fresh water</td>
<td>Sea water</td>
</tr>
<tr>
<td>Surface Area (km$^2$)</td>
<td>5.0</td>
<td>3.0</td>
</tr>
<tr>
<td>Water Depth (average m)</td>
<td>4.6</td>
<td>10</td>
</tr>
<tr>
<td>Water Volume (million m$^3$)</td>
<td>23</td>
<td>30</td>
</tr>
<tr>
<td>Water Flow Rate (km$^3$/year)</td>
<td>53</td>
<td>4.4</td>
</tr>
<tr>
<td>Water Velocity (max., m/s)</td>
<td>2.7</td>
<td>0.05</td>
</tr>
<tr>
<td>Retention Time (day)</td>
<td>0.16</td>
<td>2.5</td>
</tr>
<tr>
<td>Water Temperature (°C)</td>
<td>0.5-23</td>
<td>15-20</td>
</tr>
<tr>
<td>Other Characteristics</td>
<td>humic acids</td>
<td>organic sludge</td>
</tr>
<tr>
<td></td>
<td>woodchip sediments</td>
<td>inland sea</td>
</tr>
</tbody>
</table>

Minamata Bay and Surrounding Yatsushiro Sea  Minamata Bay is a small inlet (about 3 km$^2$) located on the southwestern coast of Kyushu, Japan, Fig. 1. It does not have any major fresh water inflow from the land except a small creek with a flow rate of about 1 m$^3$/s, most of which is discharged from a chemical factory. The tidal effect in the area including the Bay is considerable. The maximum tidal difference (between high and low tide) is 3.40 m with an average of 2.23 m (Kumamoto prefecture, 1976). The maximum surface water velocity caused by the tidal action is 5 cm/s for inflow and 4 cm/s.
for outflow at the Bay. With these low water velocities, no bed sediment transport can be expected, while suspended sediment transport is possible. Because the total volume of sea water at the Bay is about 30 million tons, 20% of the volume or 6 million tons of sea water is exchanged twice a day by the tidal action with Yatsushiro Sea.

Yatsushiro Sea is a small inland sea facing the East China Sea with a surface area of 636 km² (about the size of Lake Leman in Switzerland). The maximum depth of the Sea is 47 m and the total volume of water is 13 km³, about 5% of which is exchanged by unpolluted outside water twice a day by the tide. The maximum surface water velocity caused by the tide is 0-20 cm/s within the Sea and could reach 2 m/s at the edge of the Sea. Therefore, some bed sediment transport can be expected in a very limited area, but not in the majority of the Sea.

Because the Minamata Bay is deep (up to 30 m) and is surrounded by a protective island called Kojijima (means a romantic island), it has been a good fishing ground as well as a natural harbour for fishermen for many centuries. Historically, people in this region have lived exclusively on fishing due to the accessibility to the good fishing grounds of Yatsushiro Sea (which includes Minamata Bay) and unavailability of agricultural lands (steep hills come up to the sea shore). In 1973, well after the outbreak of Minamata disease, 144 families were still living as commercial fishermen around this small inlet. (Fishing in the Bay has never been banned officially but, in 1957, fishermen voluntarily announced the ban of fishing inside of the Bay due to the lack of sale for their catch based on a rumour that fish in Minamata Bay were toxic (Nomura, 1968)).

The accident at Minamata Bay surfaced because of human misery. On April 21, 1956, an infant girl was hospitalized with paralysed hands and legs; soon many similar cases were found and it reached an epidemic proportion. In fact, there were strange phenomena since 1953 in Minamata Bay; a number of people began to suffer from progressive numbness of their fingers, lips and tongue; dead fish, dead sea shells, unhealthy aquatic plants, etc. were frequently observed while birds and cats died violently with a nervous disorder. By 1960, at least 111 patients were identified with this strange Minamata disease. The mortality of the disease was high (20%) and the surviving patients were permanently disabled. By 1987, the Japanese Government officially recognized 1742 patients whose compensation amounted to billions of dollars (US funds).

MOVEMENT OF MERCURY

Ottawa River Study Section: Most mercury (96.7%) was associated with the bottom sediments at the study section of the Ottawa River. The total amount of mercury (22.6 kg) associated with bed sediments was obtained by measuring the surface (0-4 cm) bed sediment concentration at various locations (n = 1153), Fig. 2. The average concentration of total mercury for the entire study section was 80.6 ppb (dry) and sediments considered weighed 280,000 tons. The total mercury of 420 g was contained in the suspended solids of the study section. The average total mercury concentration (n = 104) was 1140 ppb (dry) and the mass was considered to be 3700 tons (or 16.1 mg/l). Of course, the suspended solids concentration in the water varied considerably ranging from 0.5 mg/l in winter to over 100 mg/l immediately after a shower in summer. The average mercury concentration was obtained considering seasonal variations as well as the contents of mercury. The filtered water of the River contained 300 g of mercury and this was 1.3% of mercury existing in the study section.

The biological components such as aquatic plants, invertebrates, and fish in the Ottawa River contained very little (0.2%) mercury in their systems. The average mass of aquatic plants in the study section was 1200 tons (wet) with 55 species identified. The mass included roots, stems, leaves, and so on and it was obtained by harvesting (or digging) all biomass at an area of 0.5 m². More than a dozen sites (shores or in water) were sampled every year to obtain a reasonable average. The mercury concentration varied considerably with geographical locations, species, anatomical locations, seasons,
The average total mercury concentration (n=472) was 14.2 ppb (wet) for the plants in the study section. More difficulty was encountered to obtain the mass for the benthic invertebrates in the study section. The bottom sediments with a depth of 40 cm were collected and sieved to catch all invertebrates in a surface area of 0.25 m². The average mass obtained was 180 tons (wet) with 23 species identified for the study section and the average mercury concentration (n=141) was 233 ppb (wet).

The precise estimation of the mass of fish living in the study section was almost impossible. Three methods were applied for the estimation; (a) catching fish by placing a number of nets at various locations, (b) catching fish and releasing them with identification tags (total 5000 fish) and attempting recatching tagged fish at a later day, and (c) using published fish mass for a unit area. The best possible estimate was 13 tons (wet) for the study section. The average total mercury concentration (n=535) for fish was 162 ppb (wet) and this amounted to 2.1 g of mercury in fish in the study section. The mass of fish, 13 tons, was 2.6 tons per km² of the water surface.

The organic mercury contents in the biological samples were higher than those of physical components of water, suspended solids and bed sediments. The aquatic plants, benthic invertebrates, and fish contained 20, 40, and 85% of organic mercury, respectively, while water and bed sediments contained 24 and 3.8%, respectively. The value of bed sediment, 3.8%, was not obtained by direct measurements but by an equilibrium concentration experiment between inorganic and organic mercury in various bed sediments in the study section.

A considerable amount of mercury, 1687 kg, was flowing through the study section every year, Fig. 2. The suspended solids carried more than a half, 982 kg (58%), of mercury transported downstream in the Ottawa River and water carried 689 kg (41%). Reported data supported the significant role of suspended solids in the Ottawa River, the River Thames in England 92%, the Rhine River (Germany) 60%, the Rhine River (Netherlands) 56%, the Colombia River in the USA 52%. Bed sediments played a negligible role (1% or 16.1 kg) for the geographical transport of mercury, though 96.7% of mercury existing in the study section was associated with bed sediments. Again, the biological components' roles were extremely small (0.003%) for the geographical transport of mercury. Only 7, 40, 2 g of mercury were transported by aquatic plants, invertebrates, and fish, respectively.

Minamata Bay: Surprisingly, very little information was available concerning the movement of mercury in Minamata Bay. This was due to the fact that the Japanese Government had held a position for over 2 decades of "Mercury associated with bed sediments does not move within Minamata Bay" without any published scientific documents to support this position. No one from the Japanese scientific community had supported their Government's position independently despite the fact that there are many excellent scientists and engineers with considerable resources in Japan and Japan is a free democratic country with a guarantee of academic (scientific) freedom. The understanding of mercury movement in the Bay is most urgent and critical to protect the people and their surrounding environment. Furthermore, knowledge from Minamata Bay can help not only in the Minamata problem itself but in similar problems around the world.
In 1960, Goto et al. measured mercury concentration in bed sediments using the dithizone method (Takeuchi, 1969). This was the first measurement of mercury 7 years after the outbreak of Minamata disease. To their surprise, bed sediments in a creek from the chemical factory to Minamata Bay contained 2010 ppm of mercury in wet weight base (it could be well over 3000 ppm (or 0.3 %) in dry weight base). In January of 1961, the factory installed a wastewater treatment facility to reduce mercury releases in the effluent water. In 1960, mercury was not spread massively all over the Bay sediments. Four hundred metres from Hiyakken-mon (where the creek and the Bay meet), mercury concentration in bed sediments was 133 ppm (wet), at 1 km it was 59 ppm (wet), at 2.5 km 40 ppm (wet), at 3 km 19 ppm (wet) and 35 km 12 ppm (wet). In 1973 (13 years after the first measurements), Kumamoto prefecture launched an intensive program of measurements of mercury concentration in bed sediments at 342 locations within the Bay (Kumamoto prefecture, 1976). The maximum concentration was 558.6 ppm (dry) at 0.2 km from Hiyakken-mon and it decreased with an increase of distance; at 1 km it ranged from 100 to 500 ppm; at 2 km, 25-100 ppm; at 3.5 km, 3-20 ppm. It was clear with these data that mercury moved from the effluent point, Hiyakken-mon, to the Bay and beyond to Yatsushiro Sea, despite the position of "No Movement". No comprehensive investigation on Minamata Bay mercury was conducted at this time and since. Kudo et al., in 1980, had shown a dispersion curve of mercury concentration of bed sediments using Kumamoto prefecture's data and their own measurements including the Yatsushiro Sea. Again, the data had clearly shown a movement (dispersion) of mercury, or consistent pattern of decrease in concentrations of surface bed sediments with an increase of distance from the effluent mercury source, Hiyakken-mon.

**Yatsushiro Sea:** In 1975, 24 sampling stations were marked in Yatsushiro Sea (636 km²) in order to answer whether mercury was moving (dispersing) into Yatsushiro Sea. Usually it took 3 days to collect surface sediments (0-4 cm depth with an Ekman sampler) from all stations using a Nagasaki University research ship. Only total mercury was analyzed for each station. Since then (15 consecutive years), mercury concentration was analyzed for each sampling station every year (total 291 samples). It was impossible to pinpoint each sampling station every year in the open sea (modern radar equipment in the ship could locate a point with an error of ± 100 m). Therefore some variation in texture of bed sediments (hence the content of mercury) was unavoidable.

![Fig. 3. Time history of mercury in Yatsushiro Sea sediments showing transport of mercury from Minamata Bay and effect of artificial and natural decontamination. (A in 1982 was caused by a historical rainfall, 400 mm in 3 hours, a 200-year event.)](https://iwaponline.com/wst/article-pdf/26/1-2/217/16413/217.pdf)

Over the years of sampling and analysis, shifting trends in mercury behaviour in Yatsushiro Sea have been found. Between 1975 and 1984 mercury concentrations gradually increased, with sampling stations close to Minamata Bay showing the greatest rise. At a station 8 km from Hiyakken-mon, the increase was dramatic, with concentrations rising from 0.6155 ppm (dry) in 1975 to 3.478 ppm in 1978 and continued increasing to a peak of 10.273 ppm over the following years. At another, more remote station (52 km), concentrations rose from 0.167 ppm in 1975 to 0.218, 0.250, 0.244, 0.261, 0.300, 0.100,
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0.218, 0.284, and 0.215 ppm over the ensuing years. The background level of total mercury in the surface sediments of Yatsushiro Sea was considered to be 0.100-0.200 ppm.

Total amounts of mercury in the surface sediments were calculated using concentrations obtained each year, Fig. 3. Amounts in the Sea increased from 11.28 tons in 1975 to 16.78, 22.39, 33.35, 38.37, 37.71, 26.83, 43.05, 44.86, 23.49, 25.92, 33.39, 24.04, and 20.35 tons from 1977 to 1989. Between 1975 and 1984, except 1982, the total amount of mercury increased from 11.28 to 44.86 tons. This could be considered to be a continuous dispersion of mercury from Minamata Bay outward into Yatsushiro Sea. On average, an increase of 3.7 tons of mercury per year was observed over this period. More detailed information was available elsewhere (Kudo et al., 1980, 1991, Miyahara et al., 1988).

DECONTAMINATION RATES (HALF-LIVES)

Both natural and artificial decontamination processes were effective. For the Ottawa River, the mercury in the bottom sediments was removed rather rapidly by flowing water, ranging from 0.95 to 2.6 years for the decontamination half-life, Table 3. In this case, the study section of 5.038 km² was divided into 5 sub-sections; (a) north channel below a pulp and paper mill (0.696 km²), (b) north channel above the mill (0.550 km²), (c) main channel (3.522 km²), (d) Kettle Island bay and shores of Upper Duck Island (0.130 km²), and (e) Ontario shore (0.130 km²). In 1972, the average mercury concentrations for each sub-section were 380, 123, 58, 403, and 420 ppb (dry), respectively, for the 4 cm surface sediments. One year after they were reduced to 312, 20, 27, 62, and 129 ppb, respectively. More than 40 samples were taken for each section for every year. In 1976, they were reduced further to 170, 5, 10, <5, and 50, for sub-sections (a), (b), (c), (d), and (e), respectively. From these observations taken over 5 consecutive years, the decontamination rate (or half-life) of the surface bed sediments were calculated as 2.6, 0.95, 0.98, 1.05, and 0.95 years for the sub-sections of (a), (b), (c), (d), and (e), respectively. For the study section as a whole, the decontamination rate (half-life) was calculated as 1.20 years by weighting sub-section areas for the total mercury removal rate from the surface bed sediments.

The artificial decontamination process applied in Minamata Bay was impressive, Fig. 4. A series of steel columns filled with clean sands sealed a portion, 582,000 m² (or 176,000 tsubo or 144 acres), of Minamata Bay, whose bed sediments were contaminated by 50-559 ppm of mercury. The contaminated (25-100 ppm) bed sediments existing outside of the sealed area were vacuumed with large vacuum pipes into the sealed area, as shown in Fig. 4. The total amount of contaminated bed sediments vacuumed was 1.5 million m³ (or the equivalent volume of a highrise building of 100 m x 100 m with 150 m height). With this artificial decontamination process, more than 80 % of mercury associated with bed sediments was placed in the sealed area of the Bay where it was further covered with clean, mercury-free soils to create an area of reclaimed land. The decontamination was conducted carefully, avoiding any disturbances on the surface of the remaining contaminated bed sediments. All contaminated sediments with mercury concentration above 25 ppm were sealed under the reclaimed land, but bed sediments containing mercury less than 25 ppm remained in the Bay and outside. An interesting point was that fish living in and outside of the Bay still contained the same level of mercury before and after this massive artificial decontamination effort (Kumamoto prefecture, 1989). Nevertheless, the efforts for this artificial decontamination prevented the dispersion of mercury from the Bay and accelerated the natural clean-up processes in the surrounding environment.

Yatsushiro Sea has been cleaned-up with the natural decontamination process with a half-life of 9.5 years. If this trend of natural processes continues for the foreseeable future, the Sea will be fully cleaned-up by 2011 AD (or 20 years later from 1991). The effects of the artificial decontamination process conducted in Minamata Bay were dramatically observed in Yatsushiro Sea where levels dropped from 44.86 to 23.49 tons. This decontamination rate was equivalent to an acceleration of the natural processes by 31.5 years.
The natural forces can be as effective as the massive artificial process applied in Minamata Bay as shown in Fig. 3 (marked A). There was a historical rainfall (400 mm in 3 hours (a 200-year event)), which flooded a region of Yatsushiro Sea. Millions of tons of fresh clean soils were washed out from the surrounding steep mountain slopes and lands, covering the surface of bed sediments throughout Yatsushiro Sea. The fresh sediments, thus, reduced the mercury concentration in the surface sediments of the Sea. One month after this historical rainfall, the surface sediment samples were collected from 24 stations. The reduced mercury concentration in 1982 reflected the natural "purification" or decontamination processes.

![Fig. 4. Decontamination Work at Minamata Bay (Vacuuming contaminated bed sediments into sealed area).](image)

The decontamination rate for the flowing river water was also impressive. The concentration of total mercury in water was 38 ppt in 1972, 18 ppt in 1973 and 13 ppt for 1974. The concentration was not available for the subsequent years because it was below the detection limit of 12 ppt. The more precise analytical method was developed for mercury in 1978. This method gave values of 4.6 ppt on April 14, 1978 and 9.8 ppt on May 31, 1978 in sub-section (b). For sub-section (e), it was 6.1 ppt for total mercury in water on both April 14 and May 30, 1978, though methylmercury concentrations were different at 2.8 ppt and 1.6 ppt, respectively (Kudo, et al. 1982). Nine years later in 1987, mercury concentrations in Ottawa River water were also measured as 9.0 ppt for sub-section (b) and 7.7 ppt for sub-section (e) (Schintu et al., 1989). The mercury concentration in the Ottawa River was measured as 6.8 ± 1.3 ppt in 1990 as one of the National Research Council Standards Program (NRCC, 1990). In this case, the water sample was collected about 100 km upstream from the study section. Using these available data, the decontamination rate (half-life) for Ottawa River water was considered to be 1.25 years for total mercury.

Interestingly, decontamination rates between water (1.25 years) and bed sediments (1.20 years) in the Ottawa River were close to each other. This was further evidence that an equilibrium concentration of total mercury between water and surface bed sediments could be established rapidly (matter of minutes or hours) (Kudo et al., 1978).

Although the amount of total mercury in the physical components (water and bed sediments) of the
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Ottawa River system was removed rapidly and reached a background level within several years, it was difficult to obtain a precise decontamination half-life for the biological components such as fish, aquatic invertebrates and plants. It was clear that the decontamination rate for the biological components was slower. The average concentrations for aquatic plants, benthic invertebrates and fish were 14.2, 233 and 162 ppb (wet weight), respectively, for total mercury in 1976. For fish, assuming the average concentration in 1969 was 0.4 ppm, this then led to a decontamination half-life of 5.1 years. The decontamination half-life for other biota would be somewhat greater than 5 years. The organic mercury fractions for aquatic plants, benthic invertebrates, and fish was 20% (number samples =472), 40% (number of samples =141), and 85% (number of samples =535), respectively. Two reasons were considered for the slower decontamination rates; (a) life span of most biota was longer than one cycle of the seasons (b) most biological components contained more organic mercury than those of physical components and organic mercury seemed more stable in a humic acid river water system.

Table 2: Mercury in Ottawa River, Minamata Bay and Yatsushiro Sea

<table>
<thead>
<tr>
<th>Source</th>
<th>Ottawa River</th>
<th>Minamata Bay</th>
<th>Yatsushiro Sea</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemical Forms</td>
<td>Pulp and Paper</td>
<td>Chemical</td>
<td>from Minamata Bay</td>
</tr>
<tr>
<td>Amount of Mercury</td>
<td>23.379 kg</td>
<td>150 tons (†)</td>
<td>45-60 (†) tons</td>
</tr>
<tr>
<td>Mercury Content</td>
<td>Water (total)</td>
<td>38 ppb, 1972</td>
<td>12.3 ppm in 1985</td>
</tr>
<tr>
<td></td>
<td>6.6 ppb, 1978(avg)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Water (organic)</td>
<td>2.2 ppb, 1978(avg)</td>
<td>5.1 ppm in 1985</td>
</tr>
<tr>
<td></td>
<td>Sediments</td>
<td>80.6 ppb, 1975-76(avg)</td>
<td>2010 ppm in 1976-77</td>
</tr>
<tr>
<td></td>
<td>Plasma (total, wet)</td>
<td>14.2 ppb, 1975-76(avg)</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Fish (total, wet)</td>
<td>162 ppb, 1975-76(avg)</td>
<td>15.7 ppm, 1960-61</td>
</tr>
</tbody>
</table>

Table 3: Decontamination Rate (Half-Life) of Mercury

<table>
<thead>
<tr>
<th>Processes</th>
<th>Ottawa River</th>
<th>Minamata Bay</th>
<th>Yatsushiro Sea</th>
</tr>
</thead>
<tbody>
<tr>
<td>Natural Processes</td>
<td>Water</td>
<td>1.25 years</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Sediments</td>
<td>1.20 years</td>
<td>9.5 years</td>
</tr>
<tr>
<td></td>
<td>Woodchips, sludge</td>
<td>2.6 years</td>
<td>1961-70</td>
</tr>
<tr>
<td></td>
<td>Woodchips, sands</td>
<td>0.95 years</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Coarse sands</td>
<td>0.98 years</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Fine sands</td>
<td>1.05 years</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Cohesive clay</td>
<td>0.95 years</td>
<td>—</td>
</tr>
<tr>
<td>Aquatic Plants</td>
<td>&gt; 5 years</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Benthic Inverte</td>
<td>&gt; 5 years</td>
<td>1.3 years (shell)</td>
<td>— (fish) 1961-70</td>
</tr>
<tr>
<td>Fish</td>
<td>5.1 years</td>
<td>2.1 years (1961-70)</td>
<td>47 years 1971-89</td>
</tr>
<tr>
<td>Historical Rain</td>
<td>—</td>
<td>—</td>
<td>2.76 years</td>
</tr>
<tr>
<td>Artificial</td>
<td>—</td>
<td>over 80%</td>
<td>1.83 years</td>
</tr>
</tbody>
</table>

As stated previously, over 80% of mercury associated with bed sediments was removed from Minamata Bay by this artificial decontamination. However, mercury concentration in fish did not decrease by an appreciable amount and remained around 0.4 ppm. This meant the decontamination half-life for fish, in Minamata Bay, was long (or 47 years) between 1971-89. The reason for the long decontamination half-life of the biological components was not fully understood yet.

In conducting investigations both at the Ottawa River and Minamata Bay (Yatsushiro Sea), it was found that the most important factor to obtain knowledge in the long term behaviour of a persistent toxin, mercury, in the environment was time, that is years of study. The study section of the Ottawa River was investigated intensively for 5 years with 50 professors, scientists, technicians and graduate students at the peak. On the other hand, Yatsushiro Sea was studied with meagre resources (probably less than 1% of the budget used for the Ottawa River) for 15 years. At present, a similar degree of information was obtained from these two investigations. In fact, the natural environment which we have been studying has a long history, probably more than thousands of years. If man tries to understand the long term behaviour of persistent toxic materials in the natural environment, he may need at least decades (not years) of observation. Unfortunately, the present society is not structured to accommodate such long term observation of the behaviour of persistent toxic materials, while society is urgently requesting the answers. This contradiction may be the biggest difficulty to solve in dealing with problems of persistent toxic materials in the environment.

CONCLUSIONS

Based on the investigations at the Ottawa River and Minamata Bay (Yatsushiro Sea), the following conclusions were obtained.
(1) The natural decontamination processes were effective for physical components of the surface sediments and the flowing water, where the turn-over of the flowing water was as high as several times per day.

(2) The artificial decontamination used at Minamata Bay (completely sealing contaminated sediments from surrounding environment) was effective and it accelerated the natural processes by 31.5 years at the surrounding Yatsushiro Sea.

(3) The biological components such as fish, invertebrates and aquatic plants took more time than that of the physical components to be decontaminated, especially when the physical components reached an equilibrium.

(4) The lack of understanding in the long term behaviour of persistent toxicants in the environment delays an establishment of rational decontamination methods.

(5) A combination of both artificial and natural decontamination methods is recommended for polluted sites with persistent toxic materials in consideration with the given environmental conditions.

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