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Remobilization by a major earthquake of DiPhenylArsinic Acid (DPAA) pollution at a site in Kamisu City, Japan

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Japan has a limited amount of readily developable land. Sites are used and reused with alternating phases of excavation and deposition of man-made strata (MMS). The Great East Japan earthquake of 11th March 2011 caused widespread damage due to ground waves and associated liquefaction-fluidization of MMS. A site at Kamisu City, Ibaraki Prefecture, was significantly affected. There, a gravel pit had been filled, partly excavated and then refilled. In 2003 it was found that a nearby drinking water well was seriously contaminated with arsenic. The site was sampled using a grid of boreholes and then excavated within a sheet pile enclosure at the source of the arsenic. The pollution came from large blocks that had been made by mixing cement with DiPhenylArsinic Acid powder and wastes that had been illegally dumped during refilling. The blocks were excavated and removed. Contaminated groundwater was pumped out and purified and the excavation was filled. The site was thought to be decontaminated, but the earthquake caused liquefaction-fluidization. Sand and water contaminated with arsenic were extruded onto the land surface through sand boils. After careful investigation of the site history and the depositional stages of the MMS it was found that arsenic had not been fully removed from MMS beneath the level containing the blocks because remediation works had been designed on the basis of grid sampling rather than an understanding the stratigraphy of, and discontinuities within and beneath, the MMS. The mechanism of DiPhenylArsinic Acid (DPAA) geopollution had not been properly understood. It is important to take history and stratigraphy of MMS into account when designing surveys and works.

Introduction

Japan has a limited amount of readily developable land. Therefore sites tend to be used and reused fairly frequently, with alternating phases of excavation and deposition. Deposition creates man-made (anthropogenic) strata (MMS which are referred to in North America as unengineered fill) which are often associated with geopollution and stability problems. The Great East Japan earthquake of 11th March 2011, with an epicenter off the coast of Tohoku, caused widespread damage due to ground waves (known in Japan as Jinami) and associated liquefaction-fluidization of MMS. Kamisu City, Ibaraki Prefecture, East Kanto Region (Fig. 1), was significantly affected.

The City had several former gravel extraction sites that, after extraction, had been backfilled for new uses. On March 20th, 2003, it was found that drinking water from a well used by occupants of collective housing at Kizaki, Kamisu City, close to a former gravel pit, was contaminated by arsenic. The concentration in the water was 450 times greater than that of the upper limit of the drinking water standard (0.01 mg/L) (Health Prevention Division and Environment Health Division of the Department of Health and Welfare, and the Environmental Measures Division of the Department of Civil Affairs and the Environment, Ibaraki Prefecture, 2003). Some citizens who used this water suffered health damage including cranial nerve disorders or mobility disorders (Ishii et al., 2004).

Following the discovery of the pollution, the Geopollution Survey Team of the Center for Water Environment Studies, Ibaraki University (Research Group for Kamisu Organic Arsenic Geopollution of the Center for Water Environment Studies, Ibaraki University, 2003a, 2003b) and the Survey Committee of the Ministry of the

Environment (Ministry of the Environment, Government of Japan, 2005) independently conducted surveys. The latter established that the pollution came from porous blocks that had been made by mixing cement with diphenylarsinic acid (DPAA) powder and waste (e.g., glass fragments etc.) (Fig. 2). These materials had been illegally used in the back filling of a fish pond (Fig. 3). The abbreviation used for

Figure 1. Location map of Kamisu, Japan, where the DPAA geopollution site is being cleaned up by the Ministry of the Environment. Main map: Location of the re-pollution site caused by liquefaction, fluidization and jinami (ground wave) in Kamisu.

Figure 2. Excavation of DPAA mixed concrete block (DCB), Kizaki, Kamisu (Ministry of Environment, Government of Japan, 2005).

these blocks in this paper is DCBs (DiPhenylArsinic Concrete Blocks).

The DCBs were excavated and removed, the excavation was back-filled and contaminated groundwater was pumped out and purified (Ministry of the Environment, Government of Japan, 2005). The site was severely disturbed by the 2011 earthquake during which the contaminant was mobilized by liquefaction-fluidization processes. This resulted in surface extrusion of sand contaminated with arsenic. This paper reviews the history of the site and reports results of observations and analyses.

History of the site

Prior to 1978, the Kizaki site was covered by pine forest growing on sandbars and an upper layer of alluvium (Kikuchi, 1968) (Fig. 4). In 1983 the site was quarried for sand and gravel but, by 1988, had

Figure 3. Fish pond used for disposal of DPAA contaminated concrete blocks, Kizaki, Kamisu, March, 1991.

Figure 4. Aerial photographs showing the history of the sand quarry and excavation site of DPAA mixed concrete blocks in Kizaki, Kamisu City.

been backfilled with sand, believed to have come from the Omi River, Chiba Prefecture, and was leveled flat. In 1991, part of the site was again excavated to form a rectangular fish pond which was infilled in 1993 (Fig. 3) and again in 1997 (Fig. 4). A well to the northwest of the pond (Fig. 3) supplied drinking water to nearby collective housing.

The 1993 infill included some large DCBs that were dumped illegally (Ministry of the Environment, Government of Japan, 2005). The fill included some large DCBs that were dumped illegally. In 2003 contamination was identified in the domestic supply well. After a site

investigation using the standard grid method recommended by the Geo-Environmental Protection Center of the Ministry of the Environment, remedial action began in 2004. In 2005, the main contaminated area was isolated in a rectangular steel sheet pile enclosure. The ground within the enclosure was excavated to expose the concrete blocks and after these had been removed, was backfilled. The sheet piles were left in place to isolate groundwater from the rest of the site while pumping and cleaning of groundwater at a purpose-built facility were carried out.

By the time of the 2011 earthquake, pumping has stopped because it was thought that the site had been decontaminated to acceptable levels. However, when material extruded by liquefaction-fluidization was checked it was found to be contaminated. Therefore decontamination was continued until, in 2012, it was estimated that about 90% or more of DPAA around well A (Fig. 2) had been removed. Therefore the pumping operation ended, the treatment facility was dismantled, the sheet piles were removed and the site surface was restored by spreading fresh fill.

Man-made strata and subsurface pollution

When the gravel pit was infilled with sand (MMS1), the base of that fill was the contact between man-made and natural strata. This anthropogenic/natural junction is known in Japan as the Jinji Unconformity (Nirei et al., 1996a; Edgeworth et al., 2015). When the fish pond was excavated in 1991, part of MMS1 was removed. Soon after, the pond was infilled with muddy silt to form a second man-made stratum (MMS2) that rested on a new basal surface (Jinjin Unconformity 1) (Nirei et al., 2015). The term Jinjin Unconformity is used in Japan for the depositional breaks within MMS. The DCBs that were the source of the contamination were disposed of illegally during this backfilling operation.

After pollution was discovered, a U sheet piled Enclosure was constructed and excavated to a depth of about 4.0 m. After removal of the DCBs, the enclosure was filled with sand and clay (MMS3) on a third basal surface (Jinjin Unconformity 2) within the Enclosure (Fig. 5). However, the excavation depth from side B to 6m away was only 2.5 m because no DCBs were in that part of the enclosure. Accordingly a

Figure 5. Man-made strata in the Enclosure and Excavation of DPAA mixed concrete block (DCBs), Kizaki, Kamisu (Source from Ministry of Environment, Government of Japan, 2005, Revised by Hiyama, 2013).

Figure 6. Excavating a DCB outside the enclosure along the line A-C (Ministry of Environment, Government of Japan, 2005).

part of fishpond sediments (MMS2) overlay the Jinjin Unconformity I in this area. On the other hand, as the result of making the enclosure and excavating the U sheet pile cut down to DCB Block Number II (Fig. 6). By excavation along the west side of the Enclosure, one part of Block Number II and another block were moved with retaining wall. At the time of the 2011 earthquake, the Enclosure with U sheet piles had been set up but the retaining wall had not been removed. As DCBs outside the Enclosure had overlain MMS1 directly with Jinjin Unconformity II, DPAA seems to have diffused into MMS1.

The Ministry of the Environment, Government of Japan (2005) surveyed DPAA geopollution of subsurface inside the Enclosure taking many As polluted cores below the excavation level 4 m (Table 1).

Relationship of Jinami (ground waves) and liquefaction-fluidization phenomena

The importance of Jinami in Japan was established in various studies such as those of: the 1978 earthquake off Miyagi Province (Nirei et al., 1986); the 1987 east Chiba earthquake; and southern Hyogo Prefecture earthquake in 1995 (Nirei et al., 1996b); as well as other events (Kazaoka, 2003). These established links between Jinami (an example of which is shown in Fig. 7), liquefaction, fluidization and man-made strata. Liquefaction of the ground is a precursor to the more serious devastation caused by fluidization of the ground (Kazaoka, 2003) and is in turn related to the morphology of the deposit that is affected.

Fluidization was described by Lowe (1975). Kazaoka (2003) proposed that fluidization occurs after liquefaction and, because the two are closely linked, used the term liquefaction-fluidization. Three mechanisms have been described in relation to the connection between Jinami and liquefaction-fluidization (Nirei, 2011):

(1) A detailed description of the liquefaction which occurred around Niigata City in the Great Niigata Earthquake of 1964 was provided by the Department of Geology and Mineralogy, Niigata University (1964). In a triaxial test apparatus, pore water pressure in an isotropically consolidated sand was found to rise when subjected to a cyclic deviator stress of constant amplitude resulting in liquefaction at shallow depths according to effective overburden pressure (Seed and Lee, 1966).

(2) Pore water pressure rises because of increasing ground water pressure associated with the resonance between the natural frequency of the aquifer and the seismic motion. Rising water pressure leads to liquefaction following fluidization of the sand layers and, eventually, a ground wave forms (Working Group on the Problem between Earthquake and Groundwater and Tatsuo Shibasaki, 1980).

Table 1. As concentration in each core sample in DCBs in the enclosure determined by X-ray fluorescence spectrometry (Ministry of Environment, Government of Japan (2005))

DPAA Mixed Concrete Block Number	Sampling Depth (m)	Material Sample	All As (mg/kg)
Block Number I	I-1-1	Core Of DPAA Mixed Concrete Block	porous concrete 450
			porous concrete 630
			gravelly sandy clay 560
			gravelly sandy clay 300
		Soil Core	sandy clay 75
			sandy clay 48
			fine~medium sand 92
			fine~medium sand 48
			fine~medium sand 320
			gravelly sand 170
	I-2-1	Core Of DPAA Mixed Concrete Block	porous concrete 930
			porous concrete 980
			gravelly clay 980
			gravelly clay 1,100
			gravelly clay 1,100
			porous concrete 5,800
			concrete including asphalt 100
		Soil Core	clay including gravels 3,400
			clay including gravels 2,600
			clay including gravels 1,700
			clayey sand 5,300
			clayey sand 120
			clayey sand 440
			fine~medium sand 360
			fine~medium sand 43
			fine~medium sand 64
	I-3-1	Core Of DPAA Mixed Concrete Block	porous concrete 11,000
			porous concrete 4,200
			porous concrete 8,200
			sandy clay 460
			porous concrete 130
			porous concrete 520
			porous concrete 650
			porous concrete 440
		Soil Core	porous concrete 820
			cemented concrete 84
			cemented concrete 160
			cemented concrete 110
			gravelly clay 38
			gravelly clay 120
			gravelly clay 100
			fine~medium sand 170
			fine~medium sand 110
			gravelly sand 92
			gravelly sand 64

Table 1. Continued

DPAA Mixed Concrete Block Number	Sampling Depth (m)	Material Sample	All As (mg/kg)
Block Number I	I-3-3	Core Of DPAA Mixed Concrete Block	porous concrete 650
			porous concrete 1,100
			porous concrete 630
			porous concrete 1,500
			porous concrete 2,100
			porous concrete 1,700
			sandy clay 1,100
			sandy clay 1,300
			porous concrete 730
			porous concrete 590
			cemented concrete 93
		Soil Core	sandy clay 420
			sandy clay 1,700
			fine~medium sand 710
			fine~medium sand 53
			fine~medium sand 98
			sandy clay 1,100
			sandy clay 900
			clayey sand 270
Block Number II	II-1	Core Of DPAA Mixed Concrete Block	porous concrete 560
			porous concrete 400
			porous concrete 480
			gravelly clayey sand 860
			porous concrete 710
			porous concrete 1,400
			porous concrete 980
			gravelly clayey fine sand 320
			gravelly clayey fine sand 230
			gravelly clayey fine sand 350
		Soil Core	porous concrete 960
			porous concrete 1,400
			porous concrete 1,500
			silty sand 140
			silty sand 2,100
			fine~medium sand 230
			fine~medium sand 260
			fine~medium sand 170
			fine~medium sand 260

(3) Liquefaction-fluidization commonly takes place in sand layers in MMS overlying the Jinji Unconformity. It is speculated that the process is similar to a seiche within the MMS. Pore pressures fluctuate as surface wave vibrations, Jinami, moved through the Jinji Unconformity, initiating liquefaction-fluidization (Nirei, 2011).

Figure 7. An example of a 1993 Jinami (ground wave) that caused liquefaction and fluidization of a road at Oshamanbe, Hokkaido (Morisaki et al., 1993).

Figure 8. Distribution of erupted sand and the crest of the Jinami (ground wave). Note the wavy line of the retaining net wall along the site of DPAA groundwater pollution cleanup by the Ministry of the Environment, Japan.

Figure 9. Jinami ground wave at the site of DPAA geopotential.

Effects of the 2011 earthquake at the Kizaki site

The Kizaki site after the earthquake is shown in Figures 8 and 9. A fence leading from the left hand side to a large depression in the centre of the site can be seen. The enclosure from which DCBs were removed and groundwater was pumped was behind that fence. The building and two tanks seen behind were the facility for treatment of the DPAA-polluted groundwater. The net fence at the left was originally level but was left undulating by the Jinami. Details of the site are shown diagrammatically in Figure 10. These include:

- the lines of poles marking the extent of the former fish pond areas where, due to liquefaction-fluidization, sand boils and jetted sand occurred;
- areas where water was jetted in association with sand boils;
- a ground crack outside and others inside the fences of A–A', B–B' and A–B and in the enclosure; and
- two crest lines of Jinami that are associated with two parallel rows of sand boils and jetted sand inside the area where the original gravel working was backfilled.

Leveling was undertaken to describe the morphology of the Jinami at the Enclosure. The tips of fencing poles around the former fish pond were used as benchmarks (○ in Fig. 10). The area surrounded by a zigzag broken line indicates the Enclosure from which DCBs were excavated. The black area in the plan at Figure 10 shows the positions of DCBs. The fence X–X' indicates the crest line of a Jinami extending eastwards from the western end of the steel sheet pile area toward east. The Jinami deformed the poles of the X–X' fence but not the steel sheet piles. Inside the enclosed area ground cracks occurred in a striped pattern and some sand boils were seen (Fig. 11). The former fish pond area also suffered cracks and sand boils extending parallel to the length of the pond (Fig. 12).

Results of analyses for arsenic in erupted sand

Seven samples were taken from sand boils and analyzed for arsenic. These were: (a) five samples from the southwestern part of the Enclosure; (b) one sample each from one meter underground at a point one meter southwest of groundwater observation wells F-26 and F-27 respectively (Fig. 13).

The results of leaching ion tests for arsenic, including DPAA, are shown in Table 2 (Hiyama et al., 2013). All samples exceeded the upper limit of the Japanese soil environmental standard.

A leaching ion test from No. 4 sampling point, 1.2 m west of the western corner of the enclosure, showed the highest value of 4.0 mg/L which was 400 times greater than the upper limit of the Japanese environmental soil standard (Fig. 14). The second highest result (2.9 mg/L) was from No. 2 located 5 m from No. 4 along the line of the sheet piles. Of the two samples taken from 1 meter below the surface, the one from location No. 7, closer to the western corner of the enclosure, showed a higher value than that from No. 8. The two samples taken one meter underground near the F-26 and F-27 groundwater observation wells located near the steel sheet piles were compared, and the value of F-26 closer to the western corner of the Enclosure showed the higher concentration. The results from the samples were used to draw an arsenic

Figure 10. Tops of the ranging poles (lines X–X' and Y–Y'), and the site of excavation of the large concrete blocks (black) containing DPAA (DCBs).

concentration contour map (Fig. 13) which shows the arsenic concentration decreasing westwards from the western corner of the Enclosure. This suggests that arsenic pollutants were advected from the subsurface principally near the western corner of the enclosure. It is thought that some sand and water polluted with arsenic was extruded from MMS1 outside the Enclosure towards the west.

Discussion and conclusions

Following site investigation, enclosure of the contaminated area, excavation and removal of the blocks and pumping and cleaning of groundwater the site was thought to have been successfully purified. However, during the 2011 earthquake, Jinami and liquefaction-fluidization led to extrusion of contaminated sand and water onto the ground surface within, and near the western end, of the Enclosure. Subsequently, further cleaning of groundwater continued until, in 2012, it was judged to be safe, and contaminated sand was removed before new fill was spread to restore the site.

The analytical results demonstrated that the remobilization was due

to advection of arsenic pollutants from MMS1 contaminated underlying the Jinjin Unconformity II outside the Enclosure. Possible scenarios for the advection mechanism are that:

- (1) pollutants in MMS1 western outside the Enclosure were not fully removed and were fluidized and advected from one part of MMS1 western outside the Enclosure onto the ground surface;
- (2) the first site survey did not use a stratigraphic method therefore the mechanism of DPAA geopollution was not sufficiently clarified. Therefore, MMS1 and MMS2 in the Enclosure, as well as all layers under the cultivated soil layer outside the Enclosure were contaminated. The remaining pollutants then moved up to the ground surface because of fluidization;
- (3) arsenic which was contained in the groundwater that erupted as sand boils as a result of fluidization was eluted.

Although the Kamisu study site has now been adequately remediated, there are investigation procedures that need further research. For example, why was the site thought to be successfully decontaminated when significant pollution was still present? We now find that the internal stratigraphy of with the enclosure was not sufficiently understood. The original survey used a standard grid for sediment sampling;

Figure 11. Details of the area used for research into liquefaction and fluidization causing with the jinami (ground wave) at the DPAA geopolymerization site being cleaned up by the Ministry of the Environment, Japan. Sampling points are shown.

Figure 12. Cracks in ground and sand remediation boils inside and outside the remediation site.

but this failed to account the inherent, complex internal stratigraphy of the MMS.

A second issue is why it was thought that the site had been successfully decontaminated when significant pollution still remained at the time of the earthquake. It seems that the ground within the Enclosure was not properly understood. The original survey used a standard grid method that did not take account of the stratigraphy of, and discontinuities within, the man-made strata. Thus it seems that the area enclosed by steel sheet piles was not placed on sufficiently clear scientific grounds. A proper understanding of the MMS stratigraphy is important in properly assessing sites and pollution mechanisms and designing remedial measures.

Japan has a legacy of polluted sites from previous land uses, many of which are associated with MMS. It also has fairly frequent large earthquakes that cause Jinami and liquefaction-fluidization is also commonly seen in those strata. There is, therefore, a significant risk of further incidents of this type occurring in future. In the case of the Kizaki site contamination was already known therefore the extruded

Figure 13. Concentrations (mg/L) of total As in erupted sand at each sampling point, including two sampling points 1m deep near observation wells F-26 and F-27. The concentration of total As was highest in the corner of the excavated site (sampling point no. 4). The concentration gradually decreased from this point westward.

Table 2. Total As concentrations in each sample

Sample No.	As concentration (mg/L)
1	1.0
2	2.9
3	2.5
4	4.0
5	1.1
7 (-1.0 m near F-26)	2.9
8 (-1.0 m near F-27)	1.1

Figure 14. Sample no. 4 used for total As analysis.

sand and water was considered to be suspect and was tested. However there are likely to be many other, as yet undiscovered, contaminated sites that could cause ground surface pollution. It would be prudent to check extruded materials for any problems.

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