

THESIS

RADIOCESIUM FATE AND TRANSPORT IN THE SOIL-WATER ENVIRONMENT IN THE PROXIMITY OF  
THE FUKUSHIMA DAI-ICHI NUCLEAR POWER PLANT

Submitted by

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## ABSTRACT

### RADIOCESIUM FATE AND TRANSPORT IN THE SOIL-WATER ENVIRONMENT IN THE PROXIMITY OF THE FUKUSHIMA DAI-ICHI NUCLEAR POWER PLANT

The objective of this research was to examine the dynamics of Cs-134 and Cs-137 in the aquatic ecosystems of the contaminated areas adjacent to the Fukushima Dai-ichi Nuclear Power Plant (FDNPP). Samples of water, suspended and bottom sediments from rivers, ponds, and a reservoir were collected. Three rivers (Niida, Abukuma, and Maeda), four ponds (Inkyozaka, Suzuuchi, and Funasawa 1& 2), and Sakashita Reservoir located in the Fukushima contaminated area were the focus of the study. Bottom sediment cores were taken from the Sakashita reservoir to analyze vertical distribution of radiocesium (r-Cs). Water samples were collected and filtered using an in-situ filtration system in the field or by using a laboratory filtration system. Dissolved Cs-134 and Cs-137 were removed from water using one of two different sorbents, a cartridge with non-woven cloth impregnated with potassium zinc ferrocyanide or a column filled with granulated cellulose impregnated with Iron Ferrocyanide. Activity concentrations of Cs-134 and Cs-137 were measured via gamma-spectrometry on a high purity Germanium detector, and concentrations of major cations affecting r-Cs behavior were measured via Ion Chromatography and Inductively Coupled Plasma Mass Spectrometry (ICP/MS). R-Cs was concentrated in the sediment in all but two of the river samples. Suzuuchi pond r-Cs was primarily concentrated in the sediment in both samples, but in the other three ponds r-Cs switched between being primarily concentrated in suspended sediment in July from

being primarily dissolved in solution during June. Sakashita reservoir r-Cs was primarily dissolved in solution. Mean river  $K_d$  values (L/g) were five to ten times higher than the mean values from the ponds and reservoir. River  $K_d$  variance was also much higher due to three river sample points having  $K_d$  values over 1000 while the rest were below 500.

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## INTRODUCTION

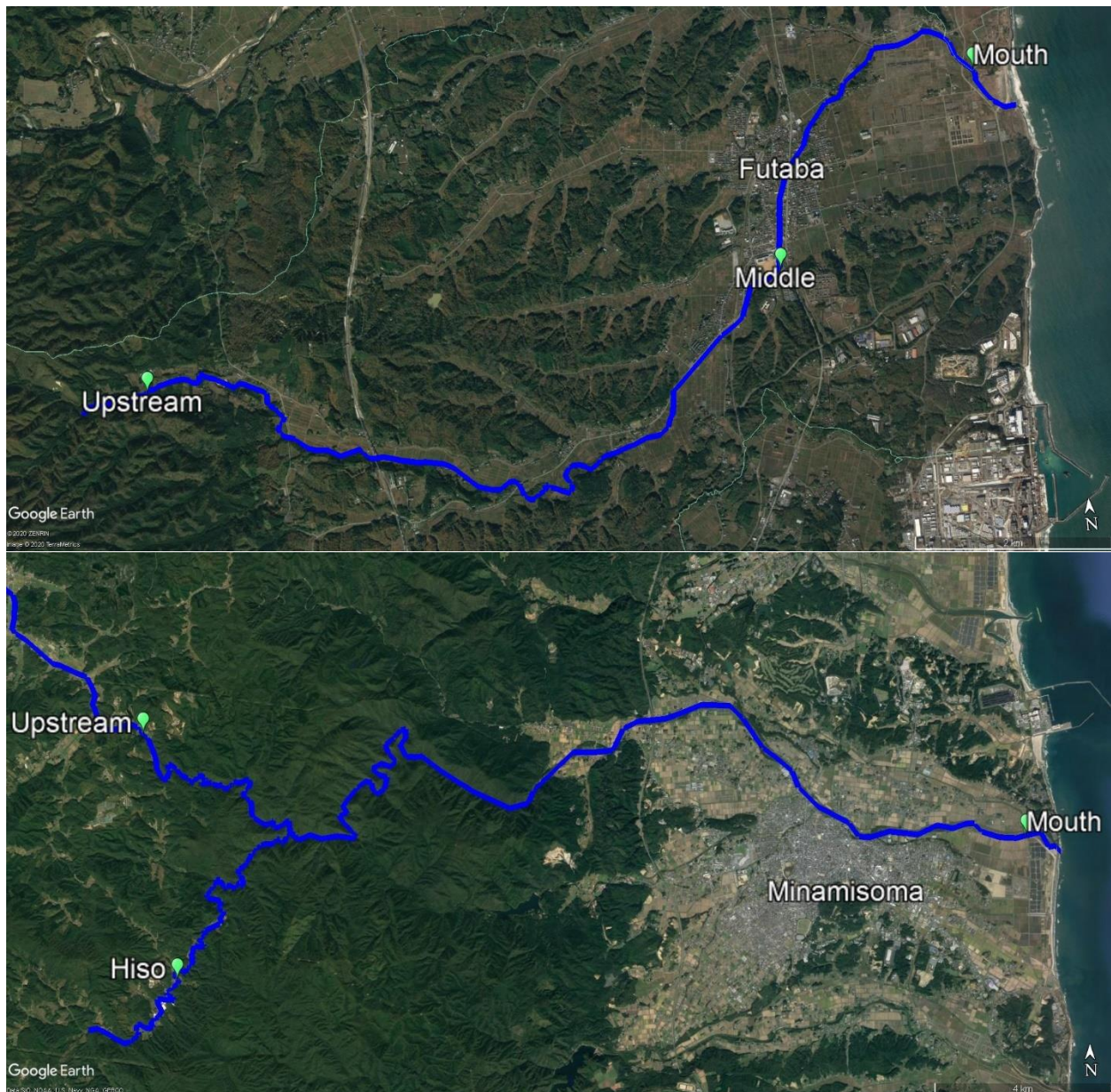
In March 2011 a major earthquake off the east coast of Japan and resulting 15-meter tsunami disabled the emergency power and cooling abilities of the Fukushima Daiichi Nuclear Power Plant (FDNPP) reactors. As a result, an area 50-70 km long and 20 km wide was primarily contaminated with I-131, Cs-134, and Cs-137 (Hirose, 2012). Initially the ratio of Cs-134 to Cs-137 was 1 (Hirose, 2012). Due to the shorter half-life of Cs-134 the ratio is approximately 8% of the radiocesium inventory as of 2019. The research objective is to examine the dynamics of Cs-134 and Cs-137 in aquatic ecosystems of the contaminated areas in proximity to the FDNPP. Analysis was accomplished by field sampling of water, suspended and bottom sediments in rivers, ponds, and a reservoir. By comparing activity concentrations of Cs-137 dissolved in the samples and from filtered sediment, analytical empirical and semi-empirical models can be used to assess fate and transport.

Previous studies found that r-Cs in ponds is primarily found on suspended and bottom sediments (Wakiyama et al. 2017). R-Cs attached to sediment largely binds cesium-selective sorption sites called frayed edge sites (FES) on clay minerals (Konoplev, 2015). Cesium competes for FES with other ions such as  $K^+$  and  $NH_4^+$ . The FES can collapse causing r-Cs to become fixed to the crystal lattice of the clay minerals. R-Cs migrates through rivers in both particulate and dissolved forms which is why the distribution coefficient,  $K_d$ , is used.  $K_d$  is defined as the ratio of the contaminant concentration associated with the solid to the contaminant concentration in the surrounding aqueous solution when the system is at equilibrium (EPA, 1999). In this study  $K_d$  is specifically the ratio of r-Cs attached to suspended sediment vs dissolved in the surrounding

water source.  $K_d$  values in the water bodies in Fukushima are 1-2 orders of magnitude higher than those that were found after the Chernobyl accident. The difference in  $K_d$  between the two accident zones is attributed to a high r-Cs binding capacity of the Fukushima contaminated area sediments and soils (Konoplev et al. 2016a). Analysis of the clay minerals found in Fukushima soils has shown that the soils which contain partially-vermiculitized biotite, termed “weathered biotite”, sorbed far more r-Cs than other clay minerals by approximately two orders of magnitude (Mukai et al. 2016). R-Cs in rivers that go through the Fukushima contaminated zone have been shown to correlate with the average r-Cs inventory in the catchment as well as depend on particle size composition (Yoshimura et al, 2014). Hot glassy particles have also been discovered in the FDNPP contaminated area that are insoluble and in addition to r-Cs contain other elements such as Uranium or other fuel and reactor materials (Adachi, 2013).

## MATERIALS & METHODS

Water samples were collected from three locations in each of three different rivers located in the contaminated region; Maeda, Niida, and Abukuma.



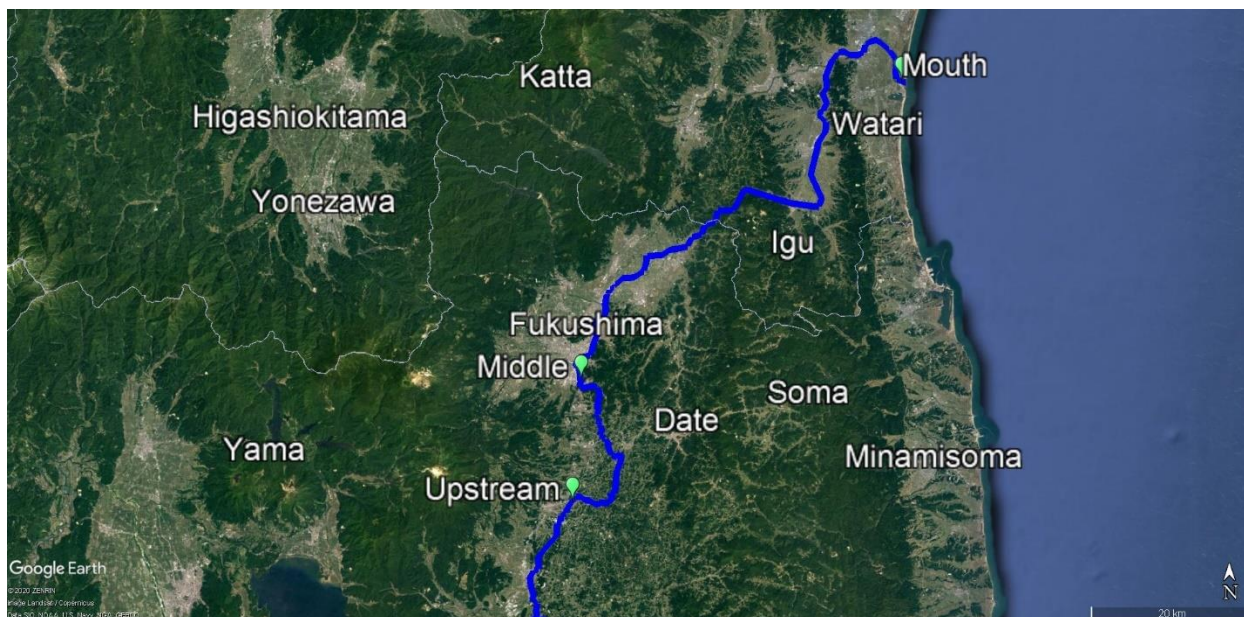


Figure 1. Map locations of sample points of Maeda, Niida, and Abukuma Rivers shown top to bottom respectively.

The samples from Niida and Maeda river were collected using a bucket tied to a rope that was lowered into the river from bridges. Samples from Abukuma river were processed using an in-situ system. The in-situ system consisted of a small pump placed approximately mid-depth of the river that fed into a container holding ten 14 cm diameter 0.45  $\mu\text{m}$  membrane Omnipore™ filters (Merck Millipore Ltd., Tullagreen, Carrigtwohill Co. Cork, IRL.). After 0.45  $\mu\text{m}$  membrane filtration, the sample was run through a nonwoven fabric cartridge filter impregnated with potassium zinc ferrocyanide (CS-13ZN by Japan Vilene Co. Ltd, Tokyo, Japan) to collect r-Cs dissolved in the water. The potassium zinc ferrocyanide sorbent cartridge filters have been shown to be effective for absorbing up to  $1 \times 10^{-6}$  g of ionic Cs at a flow rate of 2.5 L/min (Yasutaka et al. 2015). Error of 5% was assumed for the volume data returned from the in-situ system.



Figure 2. In-situ filtration system showing generator (1), pressure gauge (2), filter stack (3), sorbent cartridge (4).

Samples processed in the lab were passed through two 9 cm diameter 0.45  $\mu\text{m}$  membrane Omnipore™ filters (Merck Millipore Ltd., Tullagreen, Carrigtwohill Co. Cork, IRL.) with mesh followed by a column filled with granulated cellulose impregnated with Iron Ferrocyanide sorbent called Anfezh (EKSORB Ltd., Ekaterinburg, 620014, Russia). Lab filtration was driven by either vacuum or rotary pump. Three 5 cm deep soil samples were taken near the middle Maeda river sample point for estimating r-Cs deposition on Maeda River catchment.

River	Sample Size (L)	Filtration System
Abukuma	70	In-situ
Maeda	20	Laboratory
Niida	40	Laboratory



Figure 3. Map of the locations of ponds and Sakashita reservoir.

Water samples were collected from four ponds in the contaminated region; Suzuuchi, Inkyozaka, and Funasawa 1 and 2. Four liter sample containers were filled using a small bucket attached to a 2 meter stick. All the pond samples were filtered using the same method in the lab as for rivers. Funasawa 1 and Inkyozaka ponds had sediment traps installed in them to find the sediment and r-Cs distribution over the depth of the pond. The sediment traps were small containers with holes on the side of the top of the container that allow sediment in with the water and to over time settle out. The traps were evenly spaced along a two meter pole and placed into the ponds for a month (Figure 4). The ponds, however, were too shallow to have any sediment stratification. Previous data from a sediment trap in Abukuma river was obtained for activity and height. The Abukuma trap started at 150 cm from the bottom of the river and went

up to 340 cm. The individual traps had the captured sediment counted and afterwards checked for particle size distribution using a Laser Diffraction Particle Size Analyzer (Mastersizer 3000). Sakashita dam was the final location sampled where one hundred liters of water were filtered in-situ and 23 L were filtered in the lab.



Figure 4. Sediment traps deployed to Inkyozaka and Funasawa 1 ponds.



Figure 5. Sediment core sampling device(1), weight(2), and three sample containers(3).

Three bottom sediment cores were taken from the bottom of Sakashita dam using a weighted holder with a closing mechanism (Figure 5). The third of the cores split into two portions during the night resulting in half the sediment floating in the container and the sample becoming unusable for measurement. Two cores, one 43 cm long and the second 22 cm long, were the result. Cores were divided in the lab into 1-3 cm thick slices, dried at 50°C overnight, homogenized, and measured for r-Cs concentration. Using the cores, the activity of the bottom

sediment can be compared to r-Cs deposition in the surrounding soils. Since the reservoir is the only location where bottom sediments were sampled the reservoir is the only location where bottom sediments are compared to the surrounding environment levels. By comparing the activity to the depth, the peak activity was found and used to estimate a sedimentation rate in the reservoir from the time of the accident. Using the estimated sedimentation rate, reconstruction of the activity in the sediment over the time since the accident was made and compared to monitoring surveys performed by the Japanese Government Ministry of the Environment.



Figure 6. Sediment core #1.

After processing, the water sample, filters and sorbent were dried for >24 hours at 50° C. The filter before and after dry weight was used to find the dry sediment mass removed from the

water samples. After weighing, the filters were rolled up and cut into small pieces approximately 3 mm in width and pressed into the bottom a U-8 container for counting. The Anfezh dry weight was also obtained and counted in the same type of container. Activity of Cs-134 and Cs-137 were measured by gamma-spectrometry via a high purity Germanium detector.



Figure 7. Anfezh(blue) and filter samples(white) in U-8 containers for counting.

Each large water sample had a single small companion water sample taken simultaneously and kept chilled for hydrochemistry analysis, excepting the mouth of the Abukuma river where hydrochemistry samples were taken every 30 minutes during the in-situ filtering process to test for the effects of tide and seawater mixing. Hydrochemistry samples were filtered using syringes and 0.45  $\mu\text{m}$  membrane syringe filters. Concentrations of major cations affecting radio-caesium behavior, such as  $\text{Na}^+$  and  $\text{K}^+$ , were measured by Ion Chromatography. Stable Cs concentration was found by ICP/MS.

Cs-137 concentration in sediment was compared with Cs-137 dissolved in sorbent. The

comparison between Cs-137 in the sediment and in the sorbent was accomplished by normalizing the activities of the sediment and sorbent to the volume of water. Normalization was accomplished for sediment by dividing the activity counted from the filters by the total water filtered giving final units of Bq/L. The other parameter for comparing Cs-137 concentrations was  $K_d$  (L/g).  $K_d$ , or the distribution coefficient, is defined as the ratio of the quantity of the adsorbate adsorbed per mass of solid to the amount of the adsorbate remaining in solution at equilibrium. In this case a higher  $K_d$  value indicates that more Cs-137 is adsorbed to suspended sediment than is in solution.  $K_d$  was calculated by dividing the specific activity of Cs-137 in sediment by the activity concentration of Cs-137 in solution.

Soil types surrounding sample sites were determined in the field previously (Konoplev et al., 2016) and obtained from soil maps produced by the National Agricultural Research Organization of Japan and are based on the comprehensive soil classification system of Japan (Obara, 2015).

## RESULTS

### Maeda River

Three samples were taken from Maeda river. A large amount of construction on and around the bridge near the furthest downstream Maeda River sample point resulted in a large amount of r-Cs in sediment in the sample. As the river flowed downstream, the total Cs-137 inventory increased as well as stable Cs-133. Upstream and mouth samples found r-Cs primarily in the sediment while the middle sample point was primarily dissolved in solution.

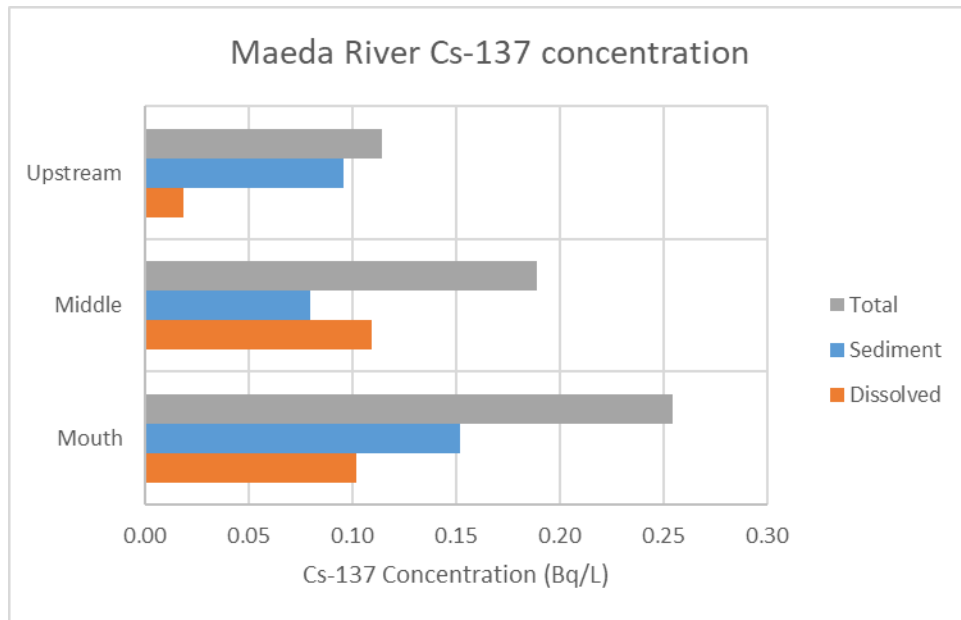


Figure 8. Maeda River Cs-137 concentration.

The three Maeda river soil cores had an average Cs-137 content of  $2.27 \times 10^4 \pm 2.67 \times 10^3$  Bq/kg which is a higher concentration than the suspended sediment found at the two upstream sites, but lower than the mouth sample. The Cs-137 inventory found from the three samples came out to be 1101 kBq/m<sup>2</sup>, 1788 kBq/m<sup>2</sup>, and 1242 kBq/m<sup>2</sup> with a mean inventory of  $1377 \pm 362$  kBq/m<sup>2</sup>.  $K_d$  decreased as the Maeda river flows downstream. Ion concentrations rose as the

Maeda river flows downstream with a large increase at the mouth where sea-water mixing occurs.

Table 2. Maeda River Sample Masses and Activities.			
Sample Point	Upstream	Middle	Mouth
GPS Location	N37°26.343 E140°56.113	N37°26.685 E141°0.463	N37°27.671 E141°1.901
Surrounding Soil type	Gravel Ordinary Gray Lowland (F3z1p2)	Gravelous Wet Brown Lowland (F4a3p2)	Coarse-Grained Normal Brown Lowland (F4z1t4)
Sample Date	17-Jun-19	17-Jun-19	17-Jun-19
Sediment Mass (g)	0.0304 ± 0.0007	0.0948 ± 0.0007	0.4441 ± 0.0007
Dissolved Cs-137 (Bq/L)	0.018 ± 0.0013	0.109 ± 0.0058	0.102 ± 0.0055
Sediment Cs-137 (Bq/L)	0.096 ± 0.0054	0.08 ± 0.0042	0.152 ± 0.0078
Sediment Cs-137 (Bq/kg)	69445 ± 883	17633 ± 254	6850 ± 70.4
Total Cs-137 (Bq/L)	0.114 ± 0.006	0.189 ± 0.007	0.254 ± 0.010
K <sub>d</sub> (L/g)	3771 ± 267.7	161 ± 8.9	67 ± 3.7

Sample Location	Stable Cs (ppt)	Na	NH <sub>4</sub>	K	Mg	Ca	Cl	SO <sub>4</sub>
Upstream	4	5.39	0.08	0.76	1.53	8.73	2.88	1.70
Middle	5	6.61	0.08	1.56	2.62	13.33	4.06	9.37
Mouth	7	67.35	0.13	4.53	9.67	17.70	105.27	26.70

### Niida River

Niida river had 3 points sampled the mouth, upstream Niida, and the tributary Hiso River. Hiso had the lowest total Cs-137 concentration while the upstream Niida sample had the largest Cs-137 inventory due to an extreme amount of Cs-137 in the sediment. The mouth of the Niida river was sampled twice during the day to ascertain tidal effects on measurements. Cs-137 was primarily found in the suspended sediment, except for the morning mouth sample where Cs-137 was primarily dissolved in solution.  $K_d$  of the Hiso and upstream samples was higher than both river mouth samples, the afternoon sample being higher than the morning. Ion concentrations were lower in the Hiso and upstream samples than both river mouth samples due to sea water mixing. The morning Niida river mouth sample was taken an hour into the ebb tide while the afternoon sample was taken during the low tide of the day. The afternoon Niida river mouth samples contained approximately half the ion concentrations of the morning sample likely due to increased sea water mixing prior to the morning sample.

Sample Point	Stable Cs (ppt)	Na (ppm)	K (ppm)	Mg (ppm)	Ca (ppm)	F (ppm)	NO <sub>3</sub> (ppm)	SO <sub>4</sub> (ppm)
Hiso	2	4.720	1.032	1.230	6.526	0.456	0.892	2.373
Upstream	2	6.251	1.066	1.569	8.711	0.418	1.052	2.587
Mouth AM	15	462.782	18.274	58.163	29.431	0.200	4.014	129.840
Mouth PM	11	192.845	8.860	23.903	17.829	0.118	4.145	57.684

Table 5. Niida River Sample Masses and Activities.				
Sample Location	Hiso	Upstream	Mouth AM	Mouth PM
GPS Location	N37°36.830 E140°48.061	N37°39.751 E140°47.550	N37°38.532 E141°0.939	N37°38.532 E141°0.939
Surrounding Soil Type	Gravelous and Humid Andosol (D3z1p2)	Fine Grain Ordinary Gray Lowland (F3z1t1)	Medium Grain Normal Gray Lowland (F3z1t2) Lower Inorganic Decay Peat Soil (B1e4w2)*	Medium Grain Normal Gray Lowland (F3z1t2) Lower Inorganic Decay Peat Soil (B1e4w2)*
Sample Date	10-Jun-19	10-Jun-19	6/10/2019 10:00	6/10/2019 15:20
Sediment Mass (g)	0.233 ± 0.0010	0.3597 ± 0.0009	0.1627 ± 0.0010	0.2563 ± 0.0009
Dissolved Cs-137 (Bq/L)	0.015 ± 0.0010	0.018 ± 0.0012	0.09 ± 0.0048	0.026 ± 0.0015
Sediment Cs-137 (Bq/L)	0.043 ± 0.0024	0.572 ± 0.0294	0.006 ± 0.0005	0.042 ± 0.0023
Sediment Cs-137 (Bq/kg)	7830 ± 168	57257 ± 685	1413 ± 76.6	7043 ± 152
Total Cs-137 (Bq/L)	0.059 ± 0.003	0.59 ± 0.029	0.096 ± 0.005	0.068 ± 0.003
K <sub>d</sub> (L/g)	507 ± 33.2	3208 ± 226.1	16 ± 1.2	275 ± 17.5
*Soil types are listed north bank followed by south bank of river.				

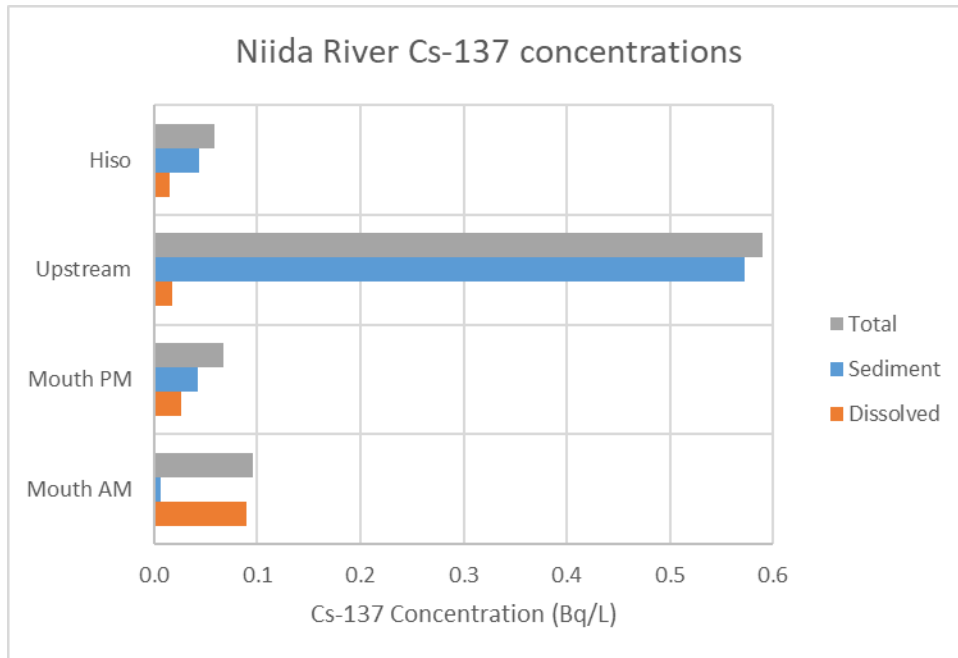


Figure 9. Niida River Cs-137 concentration.

### Abukuma River

Abukuma River samples were collected in-situ over several hours by pumping water through the sample system shown in Figure 2. Total Cs-137 concentration can be seen on figure 10 and was highest at the mouth sample point followed by the upstream, and lowest at the middle. Abukuma river sediment contained the majority of the Cs-137. Approximately 0.004 Bq/L Cs-137 was consistently found dissolved in solution at all sample points.

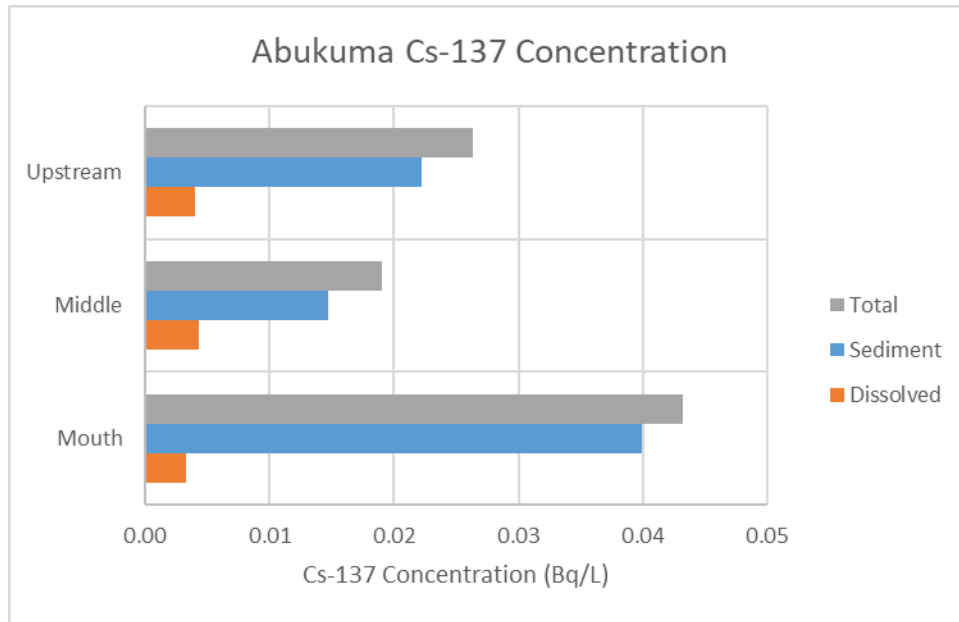


Figure 10. Abukuma River Cs-137 concentration.

$K_d$  at the upstream and middle sample points were both approximately 400 L/g while the mouth was over double at approximately 1100 L/g. Ion concentrations at the mouth of Abukuma river were much higher than the two upstream sample points due to seawater mixing. Tidal changes occurred during the Abakuma river mouth sampling. In-situ sampling began around 13:30 (Sample 3) three hours into the flood tide, just below the mid-point for the day. Sampling was completed at 16:00, approximately an hour before high tide (Sample 8). In contrast to the Niida river having an ebb tide, the Abukuma river flood tide resulted in mixing between sea and fresh water and inconsistent ion concentrations. Figure 11 shows the results from the sediment trap that was installed in the Abukuma river comparing Cs-137 activity to distance from the bottom of the river. Activity levels are higher in the upper middle portion of the river, 220-260 cm, than the rest of the river. Particle size distribution is approximately the same at all depths with the highest volume of small particles (<50  $\mu\text{m}$ ) being at 290 cm and the highest volume of large particles (>1000  $\mu\text{m}$ ) being found at 150 cm, the lowest point sampled.

Table 6. Abukuma River Sample Masses and Activities.			
Sample Location	Upstream	Middle	Mouth
GPS Location	N37°35.648 E140°27.681	N37°43.623 E140°28.387	N38°3.094 E140°54.998
Surrounding Soil Type	Medium Grain Normal Brown Lowland (F4z1t2)	Fine-Grained Normal Brown Forest (I1z1t1)	Fine-Grain Reduced Gray Lowland (F2a1t1) Fine-Grained Peaty Lowland (F2e6t1)*
Sample Date	20-Jun-19	19-Jun-19	18-Jun-19
Sediment Mass (g)	0.948	0.5872	0.8085
Dissolved Cs-137 (Bq/L)	$4.04 \times 10^{-3} \pm 2.02 \times 10^{-4}$	$4.32 \times 10^{-3} \pm 2.16 \times 10^{-4}$	$3.28 \times 10^{-3} \pm 1.64 \times 10^{-4}$
Sediment Cs-137 (Bq/L)	$2.23 \times 10^{-2} \pm 1.11 \times 10^{-3}$	$1.47 \times 10^{-2} \pm 7.35 \times 10^{-4}$	$3.99 \times 10^{-2} \pm 1.99 \times 10^{-3}$
Sediment Cs-137 (Bq/kg)	1645	1752	3990
Total Cs-137 (Bq/L)	$2.63 \times 10^{-2} \pm 1.32 \times 10^{-3}$	$1.90 \times 10^{-2} \pm 9.51 \times 10^{-4}$	$4.32 \times 10^{-2} \pm 2.16 \times 10^{-3}$
K <sub>d</sub>	407	406	1084
*Soils listed in order of west bank followed by east bank of river.			

Table 7. Abukuma River Hydrochemistry									
Sample Point	Stable Cs (ppt)	Na (ppm)	NH <sub>4</sub> (ppm)	K (ppm)	Mg (ppm)	Ca (ppm)	Cl (ppm)	NO <sub>3</sub> (ppm)	SO <sub>4</sub> (ppm)
Upstream	18	13.43	0.47	3.40	3.67	17.61	13.86	4.00	16.20
Middle	14	11.03	0.17	3.20	3.39	16.65	11.75	3.68	14.76
Mouth Avg	21	79.78	0.14	5.40	10.86	16.53	132.52	3.58	31.76

Sample Time	Cs-133 (ppt)	Na (ppm)	NH <sub>4</sub> (ppm)	K (ppm)	Mg (ppm)	Ca (ppm)	Cl (ppm)	NO <sub>3</sub> (ppm)	SO <sub>4</sub> (ppm)
11:30	16	65.53	0.10	5.05	8.96	15.56	97.29	3.27	28.14
12:00	15	51.15	0.11	4.38	7.87	15.66	77.77	3.38	25.21
13:30	23	128.12	0.38	7.04	16.20	18.89	219.18	3.60	40.82
14:00	23	119.21	0.12	6.75	15.45	18.67	204.36	3.64	40.49
14:30	22	31.00	0.10	3.67	5.69	15.52	42.72	3.88	20.73
15:00	22	67.81	0.10	5.00	9.73	16.54	104.95	3.33	28.36
15:30	22	68.83	0.09	5.03	9.30	15.13	117.15	3.49	30.79
16:00	25	106.62	0.09	6.29	13.66	16.26	196.76	4.05	39.53

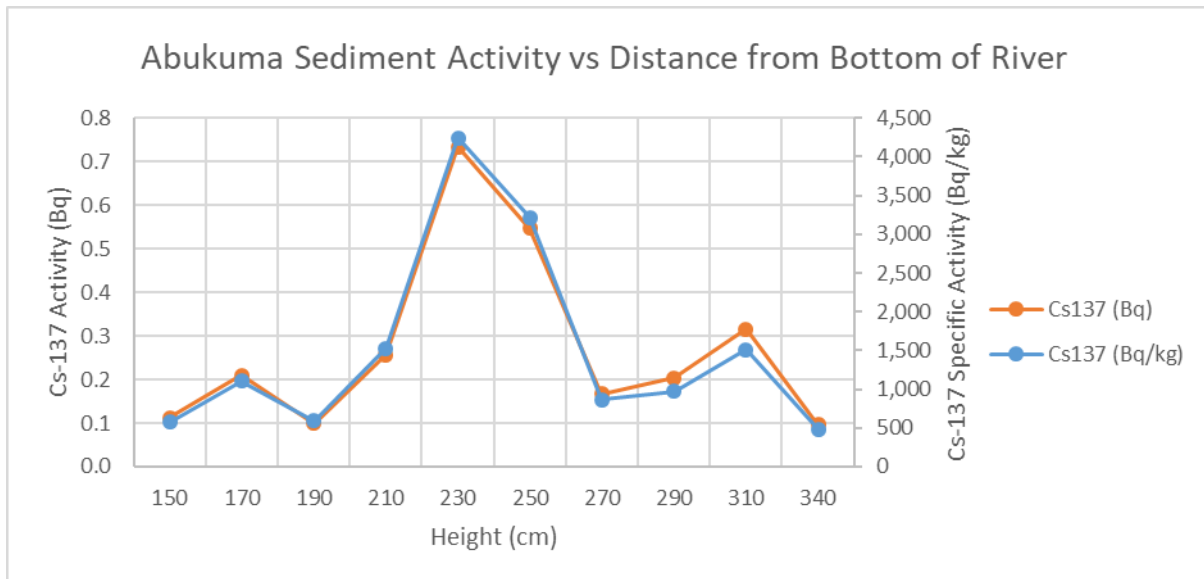


Figure 11. Abukuma River sediment activity vs distance from bottom of river obtained from sediment traps.

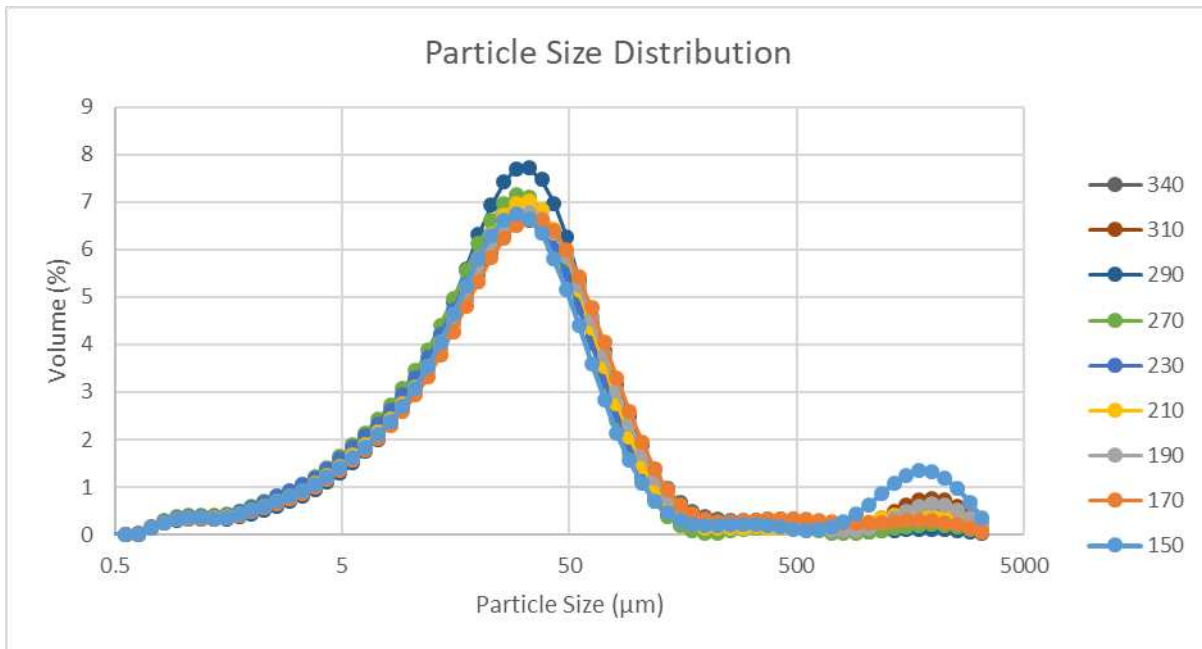


Figure 12. Particle size distribution from Abukuma River sediment traps. Legend values are height of trap from bottom of river.

## Ponds

Four ponds (Inkyozaka, Suzuuchi, Funasawa 1 & 2) were sampled twice occurring on 6 June 2019 and 10 July 2019 (Table 9). The total Cs-137 inventory in the ponds was approximately the same between the two dates, but the portion of activity in sediment vs dissolved switched between the two sample times for all the ponds except Suzuuchi on both accounts. During the June sample Cs-137 was primarily found in solution, while in July the Cs-137 was primarily found in sediment in all ponds except Suzuuchi where r-Cs was primarily in sediment in both samples. Sediment mass increased from June to July in Funasawa 1 & 2, and a near sevenfold increase in Suzuuchi, but stayed the same in Inkyozaka pond. Inkyozaka pond samples were taken near vegetation that may have contributed to the consistent amount of suspended sediment due to vegetation zones having higher sedimentation rates (Tsujiimoto, 1999). Remediation work near Suzuuchi pond was postulated to explain the large increase in sediment

mass. Funasawa 2 is fed from overflow of Funasawa 1 via a canalized channel, and unsurprisingly, Funasawa 1 & 2 had similar changes in measurements between the June and July sample dates. Ion concentrations in solution decreased between the June and July samples for Funasawa 1 and Inkyozaka ponds. In Funasawa 2 pond all measured ion concentrations decreased except potassium which increased from June to July. In Suzuuchi pond stable cesium, potassium, chlorine, and sulfate ion concentrations increased while the remaining ions decreased in concentration from June to July. The sediment traps in both ponds had activities higher than the sediment filtered from the samples. Inkyozaka trap sediment was  $1.30 \times 10^5$  Bq/kg while the June 6 and July 10 samples were  $8.51 \times 10^4$  Bq/kg and  $1.17 \times 10^5$  respectively. Funasawa 1 trap sediment specific activity was  $7.76 \times 10^4$  Bq/kg while the June 6 and July 10 samples were  $7.06 \times 10^4$  Bq/kg and  $7.03 \times 10^4$  Bq/kg respectively.



Figure 13. Inkyozaka pond sampling point.

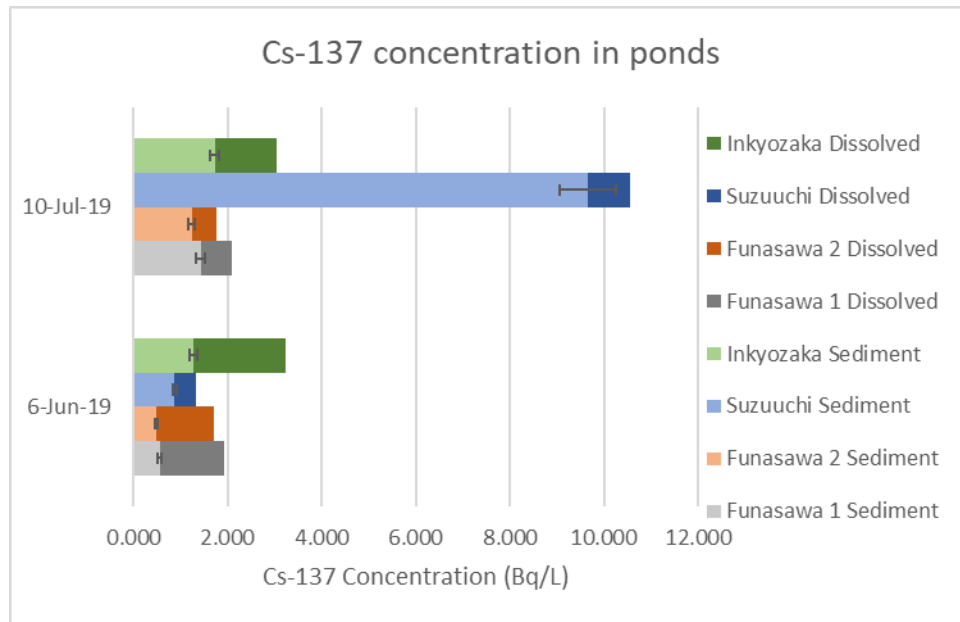


Figure 14. Inkyozaka, Suzuuchi, Funasawa 1 & 2 pond Cs-137 concentration.

Sample Location	Sample Date	Cs-133 (ppt)	Na (ppm)	K (ppm)	Mg (ppm)	Ca (ppm)	F (ppm)	Cl (ppm)	SO <sub>4</sub> (ppm)
Funasawa 1	6-Jun-19	15	6.723	2.328	4.099	27.122	0.411	5.027	26.094
	10-Jul-19	13	4.518	2.111	2.569	16.099	0.363	3.959	18.887
Funasawa 2	6-Jun-19	15	7.102	2.077	4.257	21.963	0.409	5.322	22.691
	10-Jul-19	12	4.390	2.266	2.361	14.145	0.375	4.045	16.855
Suzuuchi	6-Jun-19	5	5.921	0.783	5.275	15.511	0.132	2.370	14.607
	10-Jul-19	10	4.886	2.151	3.791	11.183	ND	3.863	42.767
Inkyozaka	6-Jun-19	19	7.048	1.476	1.601	4.483	0.792	10.714	10.584
	10-Jul-19	17	6.040	1.254	1.457	4.452	ND	8.312	9.330

Sample Location	Funasawa 1		Funasawa 2		Suzuuchi		Inkyozaka	
<b>GPS Location</b>	N37°24.363 E140°59.173		N37°24.10104 E140°59.46636		N37°24.9500 E140°58.7910		N37°25.4990 E141°01.05	
<b>Surrounding Soil Type</b>	Typical Ordinary Humid Adosol (D3z1y1)		Fine-Grained Normal Brown Forest (I1z1t1)		Fine Grain Brown Forest (I1k2t1) Typical Ordinary Humid Adosol (D3z1y1)*		Fine Grain Ordinary Gray Lowland (F3z1t1)	
<b>Sample Date 2019</b>	6-June	10-July	6-June	10-July	6-June	10-July	6-June	10-July
<b>Sediment Mass (g)</b>	0.0318 ± 0.0005	0.0811 ± 0.0005	0.0277 ± 0.0005	0.0814 ± 0.0005	0.0833 ± 0.0003	0.5384 ± 0.0003	0.0597 ± 0.0005	0.0593 ± 0.0005
<b>Dissolved Cs-137 (Bq/L)</b>	1.358 ± 0.0697	0.677 ± 0.0360	1.232 ± 0.0633	0.539 ± 0.0289	0.457 ± 0.0247	0.887 ± 0.0465	1.961 ± 0.1002	1.303 ± 0.0670
<b>Sediment Cs-137 (Bq/L)</b>	0.561 ± 0.0404	1.425 ± 0.1000	0.484 ± 0.0288	1.236 ± 0.0689	0.878 ± 0.0452	9.664 ± 0.5937	1.27 ± 0.0896	1.733 ± 0.0966
<b>Sediment Cs-137 (Bq/kg)</b>	70582 ± 3504	70268 ± 3439	69836 ± 1951	60748 ± 1455	42148 ± 505.1	71800 ± 2562	85116 ± 4187	116920 ± 2732
<b>Total Cs-137 (Bq/L)</b>	1.919 ± 0.081	2.102 ± 0.106	1.716 ± 0.070	1.775 ± 0.075	1.335 ± 0.052	10.551 ± 0.595	3.231 ± 0.134	3.037 ± 0.118
<b>K<sub>d</sub> (L/g)</b>	52 ± 3.7	104 ± 7.5	57 ± 3.3	113 ± 6.6	92 ± 5.1	81 ± 5.1	43 ± 3.1	90 ± 5.1
*Listed in order of northwest side followed by southeast side of pond.								

## Sakashita Reservoir

Sakashita dam was sampled twice, filtered using both the in-situ and lab systems. The total Cs-137 filtered in the lab system was nearly double that filtered using the in-situ system. Cs-137 was primarily found in solution from the two samples.  $K_d$  was higher in the in-situ sample. The in-situ system filtered 80 more liters of water than the lab system and resulted in more sediment mass at a slightly higher concentration than the lab sample. The in-situ system sediment concentration was 3.34 mg/L, and the lab filtration was 3.04 mg/L. Figure 16 shows the Cs-137 activity concentration compared to depth of the sediment core. If we assume the peak activity concentration that occurs around 20.5 cm is where the r-Cs from the FDNPP was introduced into the system then we can estimate the mean sedimentation rate by dividing the depth by the years since the incident, approximately  $\frac{20.5 \text{ cm}}{8 \text{ y}} = 2.56 \text{ cm/y}$ . Using the estimated sedimentation rate the activity concentration at each depth was correlated to a time and compared to monitoring survey results from the Japanese Ministry of Environment.

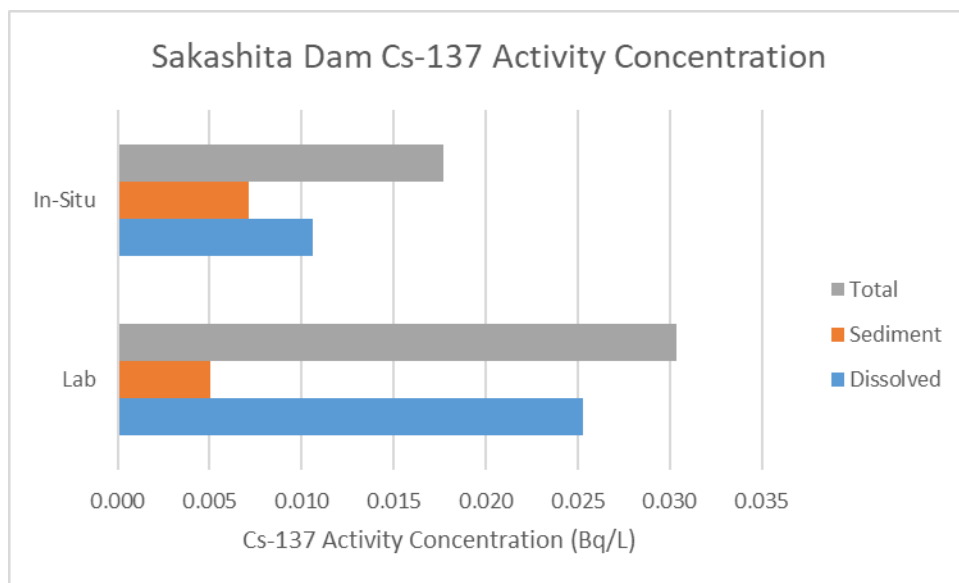


Figure 15. Sakashita Dam Cs-137 concentration.

Table 11. Sakashita Dam Sample Masses and Activities.		
Filtration Location	Laboratory	In-Situ
GPS Location	N37°22.95774 E140°56.46108	N37°22.95774 E140°56.46108
Sample Date	16-Jul-19	16-Jul-19
Surrounding Soil type	Humic Normal Allophane Andosol (D6z1v4)	Humic Normal Allophane Andosol (D6z1v4)
Sediment Mass (g)	0.0714 ± 0.0007	0.3339
Dissolved Cs-137 (Bq/L)	0.025 ± 0.0017	0.011
Sediment Cs-137 (Bq/L)	0.005 ± 0.0003	0.007
Total Cs-137 (Bq/L)	0.03 ± 0.002	0.018
K <sub>d</sub> (L/g)	66 ± 5.1	201

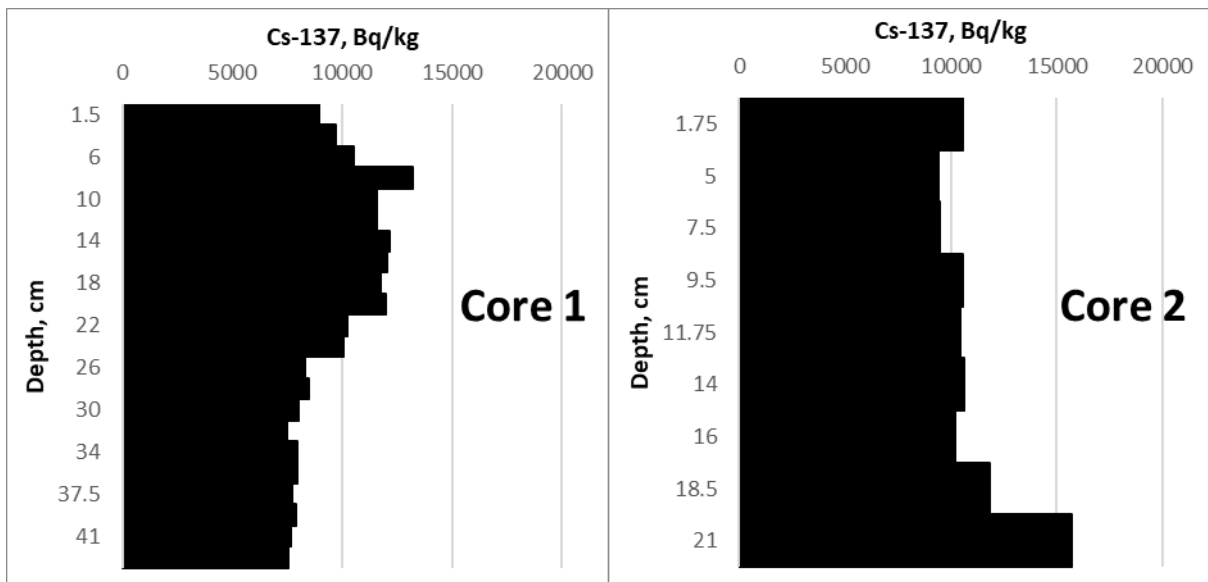


Figure 16. Sediment core Cs-137 activity concentration by depth. Core 1 total Cs-137 inventory was 986 kBq m<sup>-2</sup>, Core 2 total inventory was 515 kBq m<sup>-2</sup>.

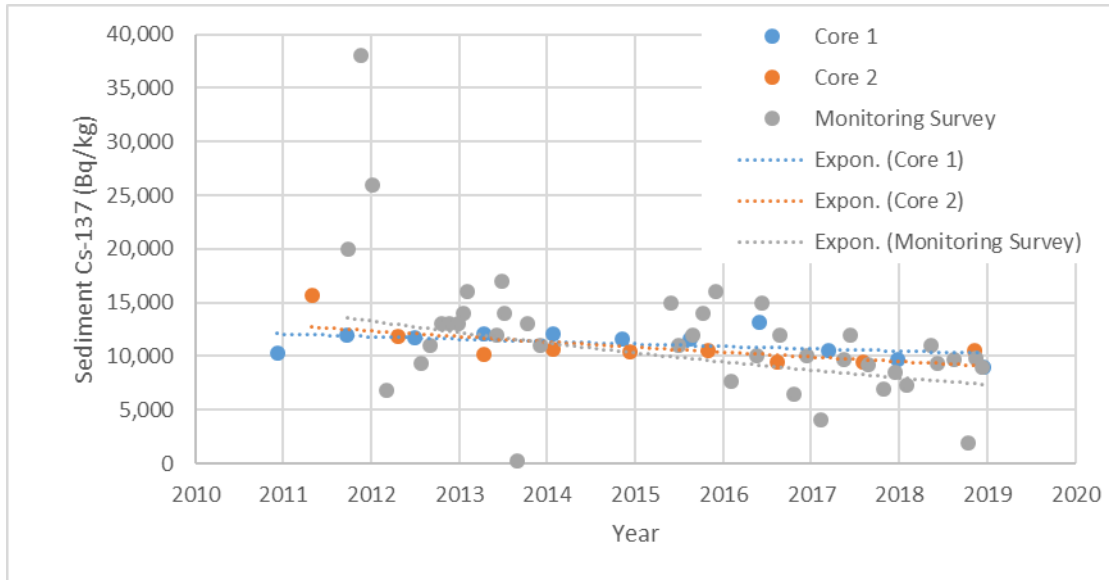


Figure 17. Reconstructed bottom sediment Cs-137 activity concentrations over time compared against Japanese Government Ministry of the Environment monitoring surveys.

### K<sub>d</sub> Comparison

Ponds had relatively similar  $K_d$  values on the same day of sampling, while differing between the two sample dates except for Suzuuchi pond. Suzuuchi was similar between the two sample dates (having a higher  $K_d$  than the other three ponds on the June samples). River  $K_d$  is largest for Maeda river at the upstream sample point at 3771 L/g, for Niida river at the upstream sample point at 3208 L/g, and for Abukuma river at the mouth sample point at 1084 L/g. Maeda and Niida rivers  $K_d$  were lowest at the mouth sample points of the rivers, 67 L/g for Maeda and 16 L/g in the morning and 275 L/g in the afternoon for Niida. Comparing the ponds to the rivers the mean pond  $K_d$  are lower than the mean river  $K_d$  values. The mean pond  $K_d$  values are comparable to the bottom end of the Maeda river distribution since the Maeda mean  $K_d$  is driven up by the upstream sample point so much.

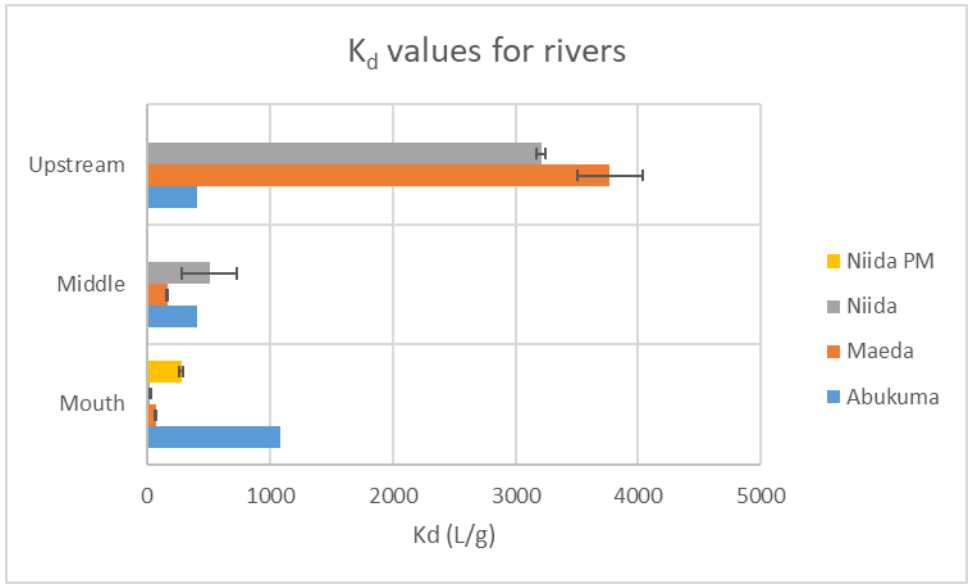


Figure 18. K<sub>d</sub> values from rivers. Niida middle point is the Hiso sample point.

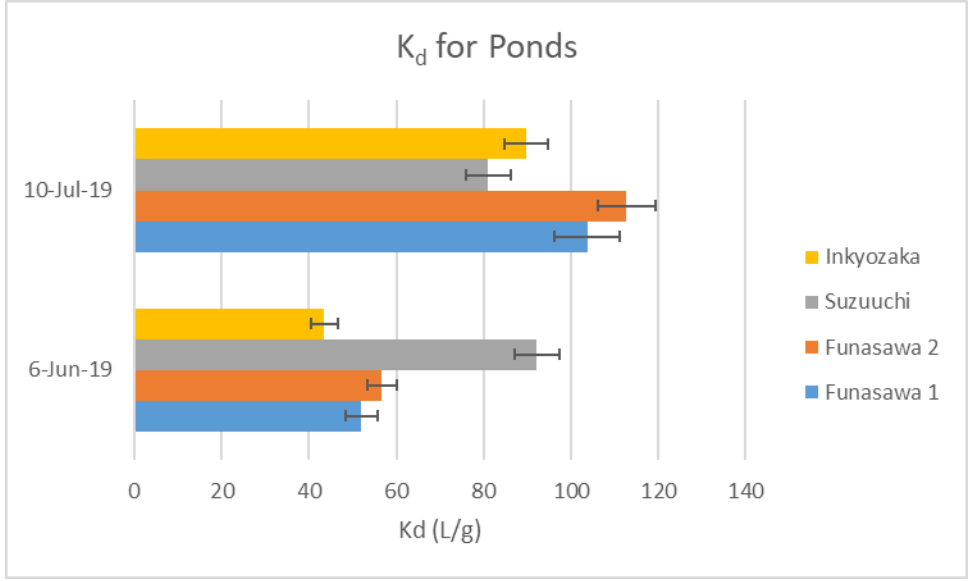


Figure 19. K<sub>d</sub> values from ponds.

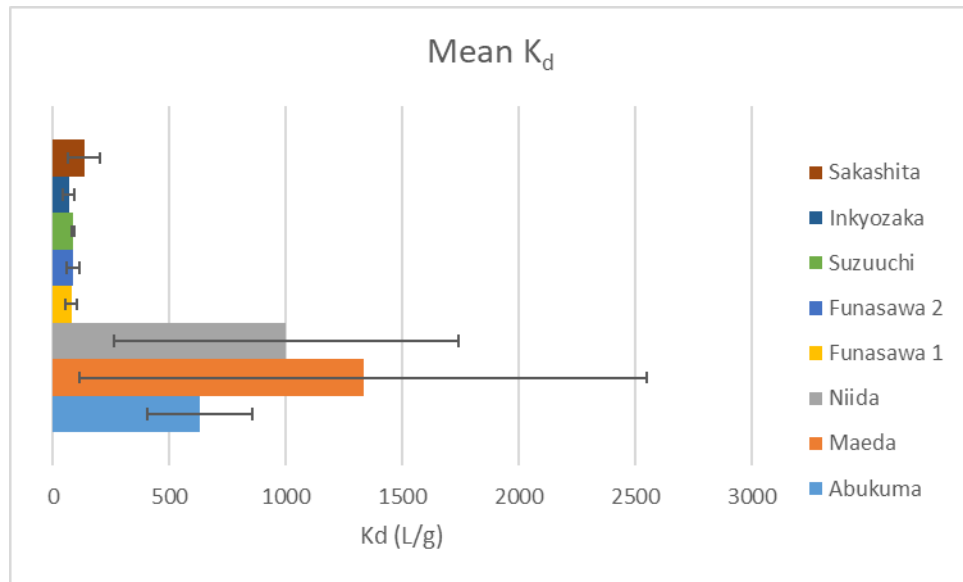


Figure 20. Mean  $K_d$  values for ponds and rivers.

## Discussion

### **Maeda River**

The Maeda river largely flows through the Fukushima contaminated zone so erosion and wash-off are the likely cause of the total Cs-137 concentration increase as the river flows downstream.  $K_d$  decreases as the river flows downstream which is likely in part because cation concentrations, such as  $K^+$  and  $Ca^{2+}$ , increase resulting in more competition for selective binding sites on the suspended sediment; different soil types surrounding the river as the river flows also likely contribute to the decline in  $K_d$  due to the different soils having varying structures and different numbers of selective binding sites.  $K_d$  at the upstream sample point is very high likely due to the surrounding soil type of gravel ordinary gray lowland, which is the same soil type for all the samples with  $K_d$  values above 1000 L/g.

### **Niida River**

Unlike Maeda river in the Niida river the mouth sample point had the second highest total r-Cs concentration. The upstream Niida sample had the highest total r-Cs concentration due to very high sediment activity concentration. The upstream sample site is surrounded by fine grain ordinary gray lowland soil which likely has a larger number of binding sites for r-Cs compared to other soils. Similarly, the Maeda river  $K_d$  is highest in the area with the ordinary gray lowland soil type.  $K_d$  at the mouth was lower than either the Hiso or upstream sample point for similar reasons to Maeda river, increased concentration of competing ions and different sediment composition. The increased r-Cs dissolved in solution and low  $K_d$  from the morning sample near the river mouth were likely caused by ion concentrations being nearly

doubled in the morning sample compared to the afternoon. The afternoon sample had approximately five times as much activity in the sediment as well as a higher sediment mass when compared to the morning sample. Suspended sediment from the ocean may also have contributed to a different sediment make up between the morning and afternoon mouth samples lowering the capacity of r-Cs adsorption on sediment. The sediment traps in Abukuma River showed an increased concentration in the middle depths of the river. It is possible that the higher water level from the high tide during the morning sample resulted in higher activity levels not being sampled since the Niida river mouth was sampled near the surface. The afternoon sample taken during the low tide may have captured the potential higher activity region if the lower water level resulted in the higher activity region being closer to the surface. The upstream sample point is near the start of the Niida river, so the cause of the lower total r-Cs concentrations downstream is likely from tributary dilution. The  $K_d$  value of  $507 \pm 33$  L/g obtained at the Hiso sample point is in the range of 92 to 970 L/g found by Ueda et al., 2012. The consistency of  $K_d$  over the years is likely due to the sediment composition not changing so r-Cs distribution maintains similar proportion in sediment compared to dissolved in solution. While the r-Cs distribution in the Hiso river is approximately the same the r-Cs activity concentrations found in sediment and solution were one to two orders of magnitude lower than the results of Ueda et al., 2012.

### **Abukuma River**

The middle sample point for the Abukuma river was taken from the output of a permanently installed sampling station. The lower sediment mass found at the middle sample point may have been a result of getting the water from the permanent sampling station while

the other river sites were sampled directly. If there was a loss of sediment from using the sample station that would explain why the middle sample site has lower total r-Cs concentration than the other sites. The general trend seen in this study is for total r-Cs concentration to increase as the rivers flow downstream. Since r-Cs is primarily in the sediment of the Abukuma river and the specific activity of Cs-137 in the sediment increases as the river flows downstream it seems likely that a portion of sediment was lost during the sampling process. Assuming that some sediment was lost then Abukuma river should also increase in total r-Cs concentration as the river flows. Maeda river has similar surrounding soil types to Abukuma where both the rivers are surrounded by majority brown lowland or forest soil. The sample with the highest  $K_d$  from Abukuma river was the mouth which is surrounded the grey lowland soil that was seen at the other high  $K_d$  river locations. The mean  $K_d$  found in the Abukuma river of 633 L/g is within the previously found mean value range of  $660 \pm 50$  (Konoplev et al. 2016a). Tidal effects on Abukuma river hydrochemistry showed large variation as the incoming seawater mixed with the fresh river water. Due to the in-situ system filtering over several hours and the hydrochemistry showing seemingly random changes throughout the sampling period it is difficult to make any specific claims as to the effects. Hydrochemistry samples were taken from the surface water while the in-situ filtration sampled several meters underneath as well further compounding discrepancies. Further examination into the effects of tidal change and mixing across the depths of the river is warranted. The sediment trap distribution showed an increased Cs-137 activity concentration in the upper central portion of the river compared to the other depths. Analysis of the particle size distribution of captured suspended sediment at each depth failed to yield an explanation for the increased activity between the different depths. More research into the

cause of this phenomenon is required.

## **Ponds**

There was no precipitation during the days leading up to June 6<sup>th</sup> and temperatures averaged in the low 20's (Celsius). July 7<sup>th</sup> had a large amount of rain, but weather was dry afterwards to July 10<sup>th</sup> with temperatures averaging around 17°C. The weather data was gathered by the Japan Meteorological Agency in Namie, Fukushima, the closest monitoring station to Okuma town and the ponds. Each pond had approximately the same total r-Cs activity concentration between the two sample dates, except Suzuuchi. In Funasawa 1 and 2 the driving factor for the switch between r-Cs being concentrated primarily in solution to sediment seems to be the increase in suspended sediment in each sample. July samples from the Funasawa ponds had over double the sediment mass of the June counterparts. The change in suspended sediment may have been a result of the rainstorm on July 7<sup>th</sup>. The specific activity of the sediment was the same in Funasawa 1 between the two samples and a slight decrease in Funasawa 2. Suzuuchi pond was the only pond to not change where r-Cs was primarily concentrated. Sediment activity concentrations increase of over ten times is a result of the increase to sediment mass in the sample and a specific activity increase of approximately 70%. Again, the rainstorm on July 7<sup>th</sup> may have resulted in a large amount of sediment being washed into the pond from the surrounding steeply sloped banks as well as sediment becoming detached from the floating plant life that covers the pond. Inkyozaka pond was the only pond to not have increased sediment mass between the two sample dates having the same mass for both. The change of r-Cs being primarily concentrated in solution to sediment comes from the specific activity of the sediment increasing by approximately 37% between the two sample

dates. The pond sediment trap in Funasawa 1 had a slightly higher sediment specific activity than the filtered sediment. Inkyozakas trap sediment specific activity was on the high end of the July sample filtered sediment specific activity and higher than the June sample filtered sediment specific activity. In the case of both ponds the higher specific activities follows the pattern of increasing around May to August shown when Wakiyama et al. (2017) sampled the same ponds. The two pond sediment traps were installed and retrieved after the July round of pond samples was taken. Table 12 lists a comparison of r-Cs activities in Inkyozaka, Suzuuchi, and Funasawa 1 to values found by Wakiyama et al. (2017). Wakiyama et al. sampled the ponds 13 times during 2015 and 2016. Values from this study were decay corrected back four years to 2015 for comparison using effective half-lives found by Hayes et al. 2020. Inkyozaka and Funasawa were corrected with the undisturbed effective half-life of 3.22 y and Suzuuchi with the disturbed effective half-life of 1.77 y. Activity of Cs-137 in the suspended sediment of Inkyozaka was similar in both studies. Dissolved Cs-137 in Inkyozaka was significantly higher than the previous study, but since  $K_d$  was lower it's expected that more of the r-Cs would be found in solution. Suzuuchi suspended sediment Cs-137 activity concentration in the June sample was one standard deviation lower than the previous study while the July sample was the same as the previous study mean. Suzuuchi dissolved Cs-137 activity and  $K_d$  were within one standard deviation of the previously found mean. Total Cs-137 activity concentration of the June Suzuuchi sample was very low compared to the previous study while the July sample was similar. Funasawa 1 suspended sediment activity concentration was over one standard deviation lower in this study than the previously found mean. Funasawa 1  $K_d$  was like Inkyozaka, lower than the previous study. Dissolved Cs-137 is over two standard deviations higher in the June sample than

the previous study mean, while the July sample is similar to the previous study. Total Cs-137 activity concentration has been shown to have temporal variation (Wakiyama et al. 2017) Suzuuchi pond specifically has a massive difference between just the two samples.

Table 12. Comparison of pond data with Wakiyama et al. 2017.				
Sample year*	Dissolved Cs-137 (Bq/L)	Sediment Cs-137 (Bq/g)	Total Cs-137 (Bq/L)	K <sub>d</sub> (L/g)
Inkyozaka				
2015/2016	1.6 ± 0.22	263 ± 136	9.5 ± 5.0	170 ± 80
2019 June/July	4.6/3.1	201/277	7.6/7.2	43/90
Suzuuchi				
2015/2016	3.0 ± 1.4	333 ± 79	40.6 ± 20.8	130 ± 50
2019 June/July	2.2/4.2	202/344	6.4/50.5	92/81
Funasawa 1				
2015/2016	1.8 ± 0.51	337 ± 120	13.3 ± 9.4	210 ± 93
2019 June/July	3.2/1.6	167/166	4.5/5.0	52/104
*2019 samples decay corrected four years to 2015 equivalent.				

### Sakashita Reservoir

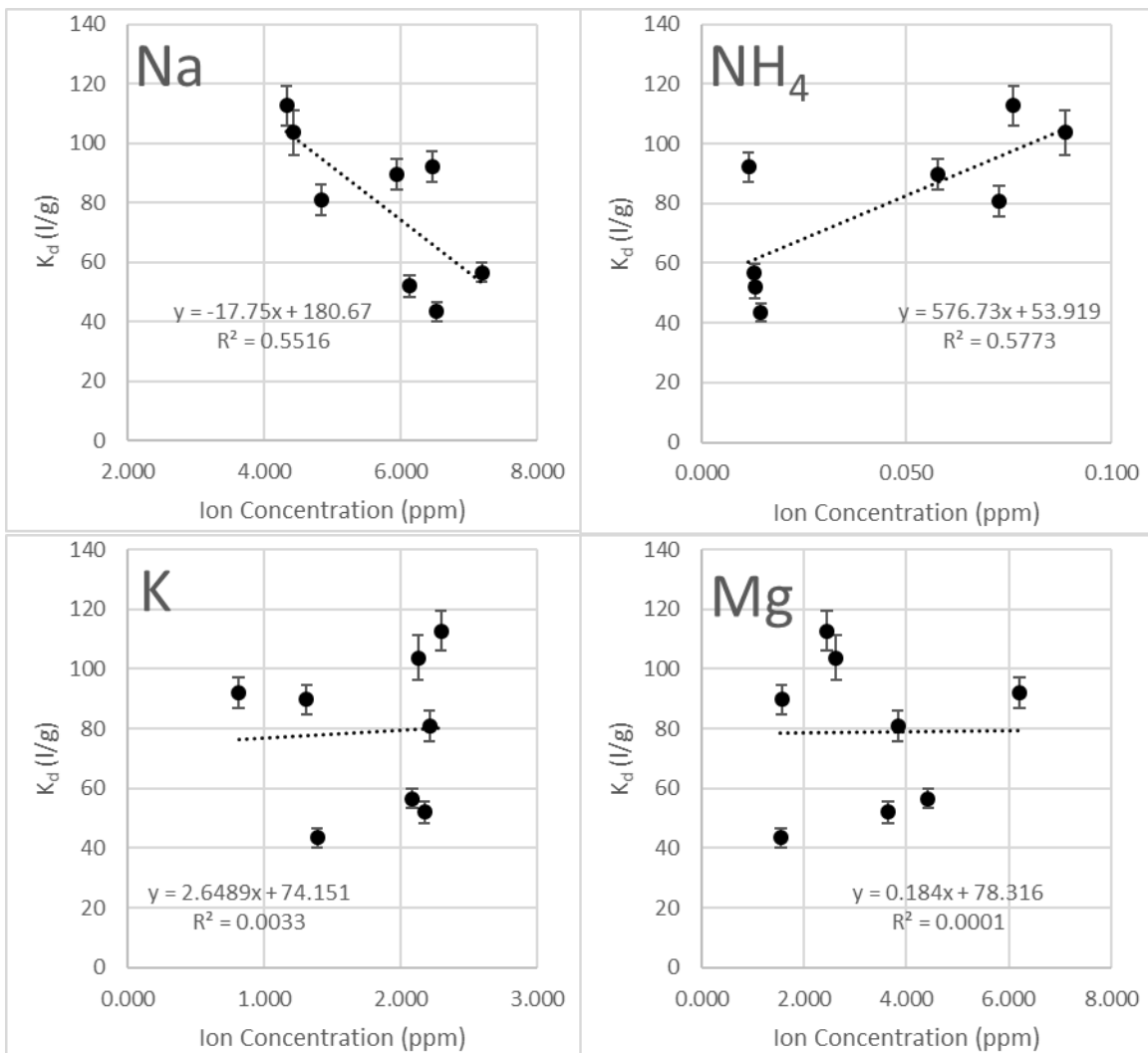
Sakashita Reservoir had total r-Cs concentration levels similar to those found in Abukuma River. The total Cs-137 activity concentration from the lab was higher than the in-situ sample. The lab sample was obtained from the shore with a bucket, while the in-situ system pump was drawing the sample several meters from shore and several meters underwater. The difference in sample depth between the in-situ and laboratory samples likely contributes to the difference in total activity as well as the difference in sample time, the in-situ system sampled over two hours while the lab sample was collected in minutes. Interestingly, the primary contributor to the difference in total Cs-137 activity concentration was from the lab sample containing over twice the concentration of dissolved Cs-137. Both samples the Cs-137 was primarily dissolved in solution. K<sub>d</sub> from the reservoir samples was on the high end of the range

of the ponds, but lower than the river  $K_d$  values. R-Cs inventory from the sediment cores can be calculated and compared with deposition on the soil in the surrounding area of  $672 \text{ kBq/m}^2$  (Konoplev et al., 2016b) which decay corrects to  $594 \text{ kBq/m}^2$  at the time of sampling. Any difference between the r-Cs inventory of the sediment cores and the r-Cs inventory of the surrounding land is due to sedimentation of suspended sediments. Total Cs-137 inventory of the two cores was  $986 \text{ kBq/m}^2$  and  $515 \text{ kBq/m}^2$  for the first and second core respectively. The top slice of core 1 (3 cm) had a Cs-137 inventory of  $57 \text{ kBq/m}^2$  and core 2 (3.5 cm) had  $60 \text{ kBq/m}^2$  the average of the two slices was  $58 \text{ kBq/m}^2$  showing that the topmost reservoir bottom sediments were made up by less of the surrounding soil than from other sources or the r-Cs is being leached out of soil into the water. Average specific activity of suspended sediment was  $1.9 \text{ kBq/kg}$  of suspended sediment and average specific activity of the bottom sediment was  $9.77 \text{ kBq/kg}$  of sediment. The difference between the suspended and bottom sediments is possibly due to difference in composition between the two. It is possible that higher Cs affinity sediments are more likely to be deposited on the bottom of the reservoir. The specific makeup of suspended and bottom sediments should be analyzed to investigate this. The core slice measurements from figure 16 show that after the increase around 20.5 cm the shallower activity levels remain relatively stable around the increased value. The relatively stable activity levels across depth indicate potential for mixing of layers between the bottom sediments or that a continuous source of radioactive sediment is being deposited. The sediment core reconstruction (Figure 17) has a much lower peak than was found from the initial Japanese government monitoring survey data. The remainder of the reconstruction matches the monitoring data with a flatter slope. The small peak, and general flatness of the core slices

supports that a high amount of mixing occurred amongst sampled bottom sediment.

### **K<sub>d</sub> and Hydrochemistry**

Change in pond K<sub>d</sub> between the two sample dates may be in part driven by hydrochemistry in addition to potential effects from weather. Ions that occur in hydrated form are too large to approach and compete for binding at FES. For ponds the flat trend of Mg, Ca, and SO<sub>4</sub> suggest that r-Cs K<sub>d</sub> is driven by adsorption to the selective FES and not outer surface regular exchange sites.



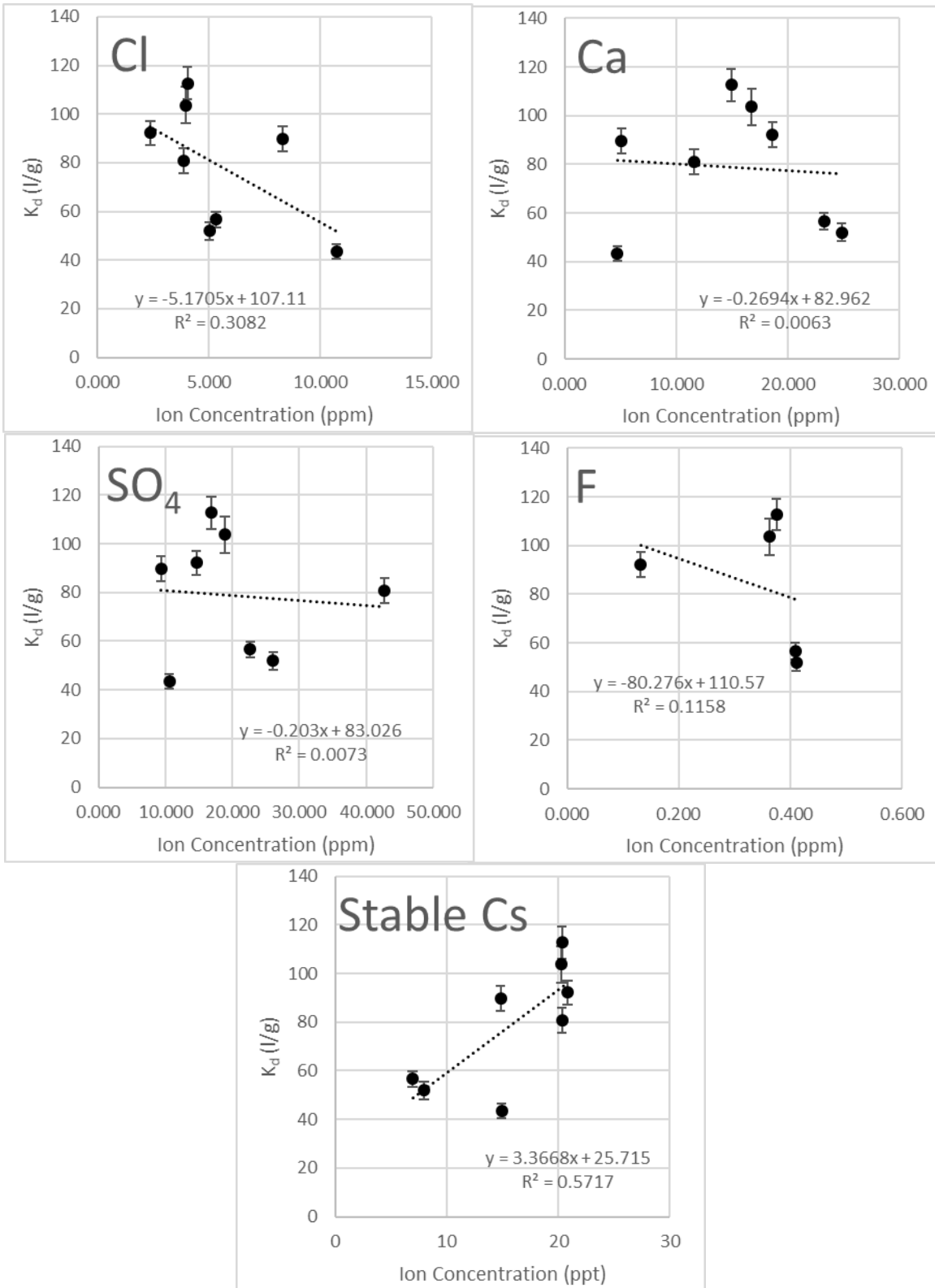
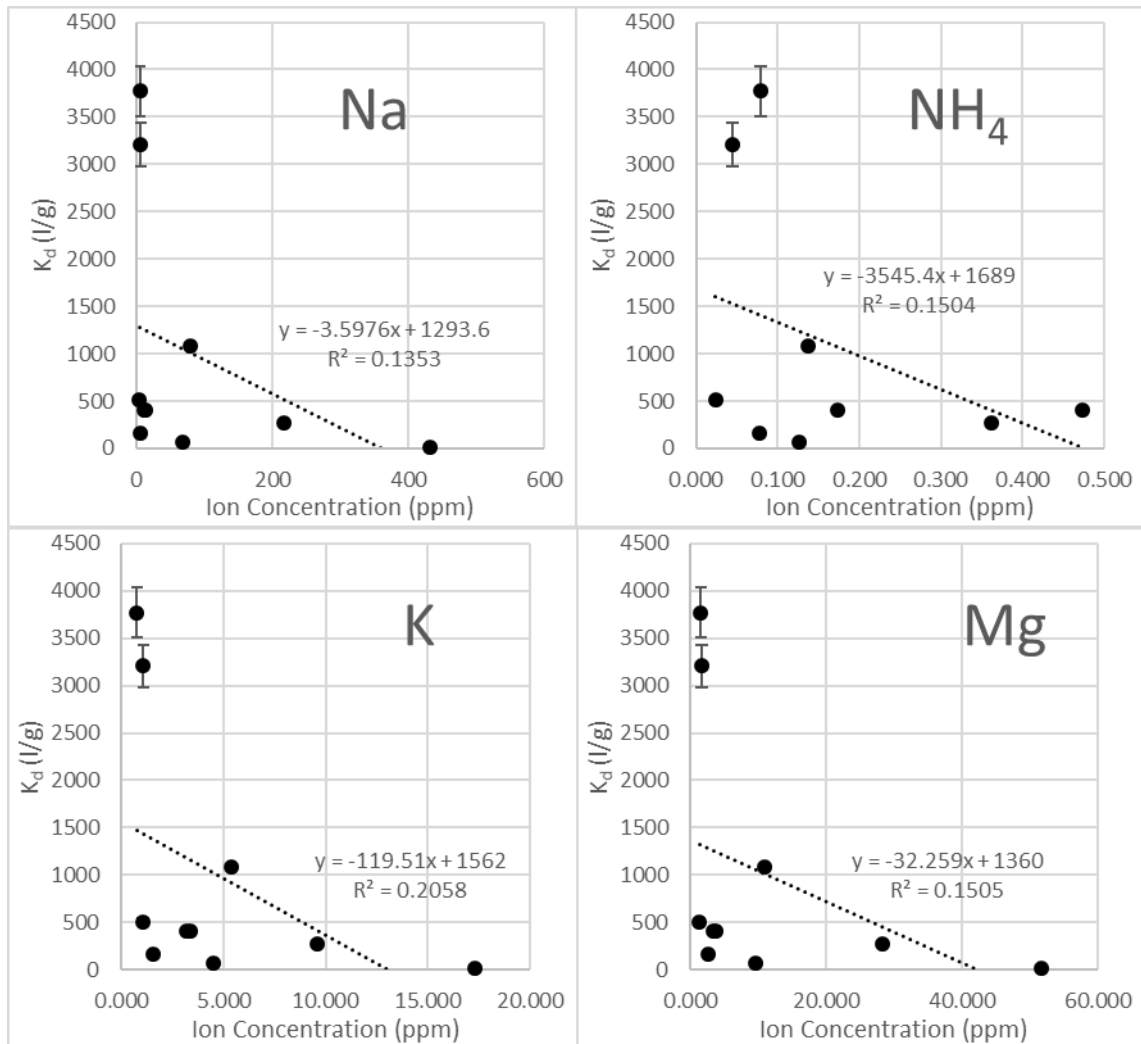


Figure 21. Pond ion concentration compared to associated  $K_d$  values.

The river samples with very high  $K_d$  values probably contribute to the consistency of the negative trends of  $K_d$  with increasing ion concentration. Generally, in rivers the high ion concentrations are associated with the lowest  $K_d$  values. Unlike the ponds  $K_d$  decreases with higher concentrations of Mg and Ca suggesting that either the greater diversity of sediment composition results in less FES availability and more competition for regular exchange sites,  $K_d$  is lower in general with higher ion concentrations, or some combination of the two.



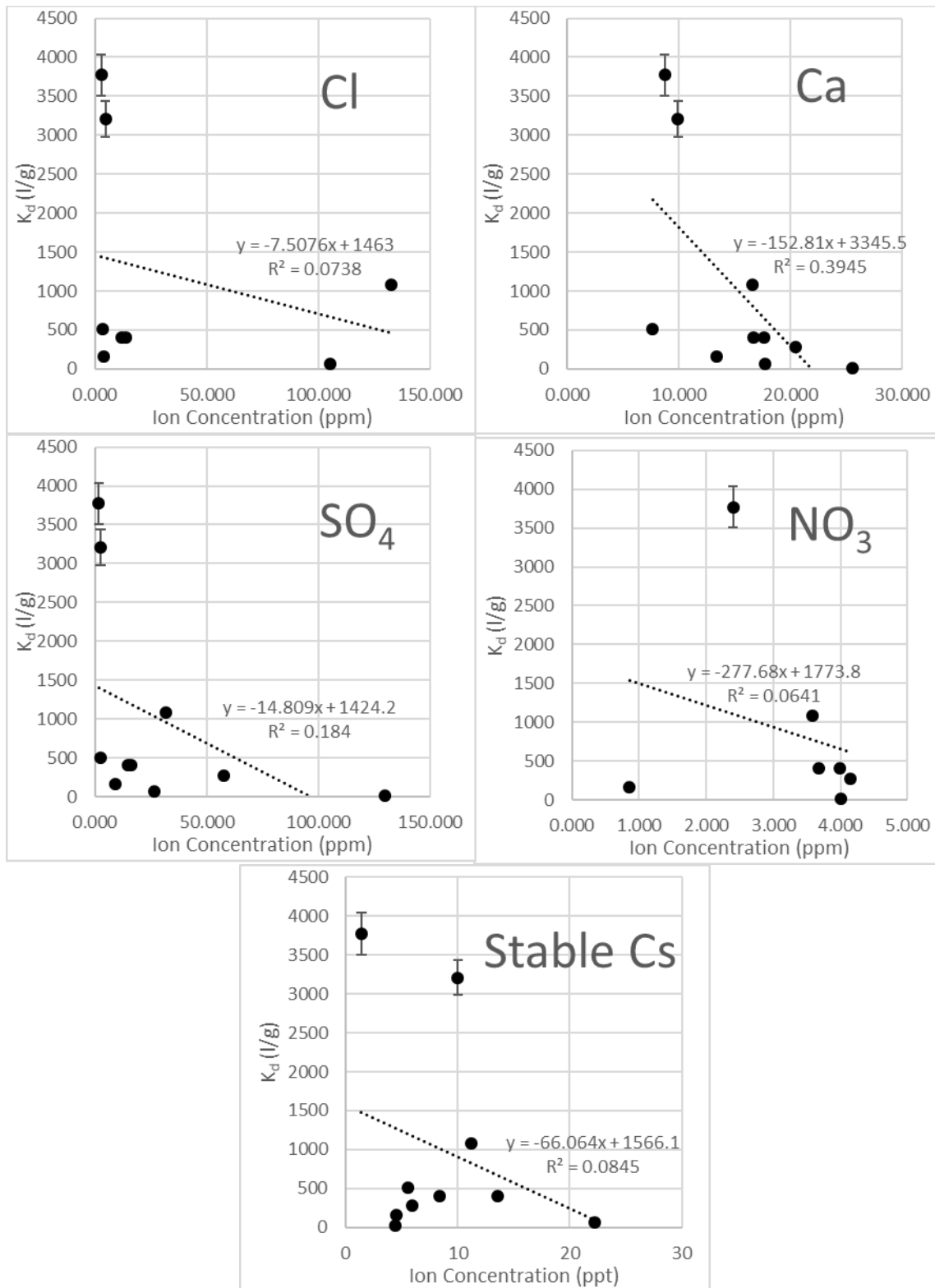
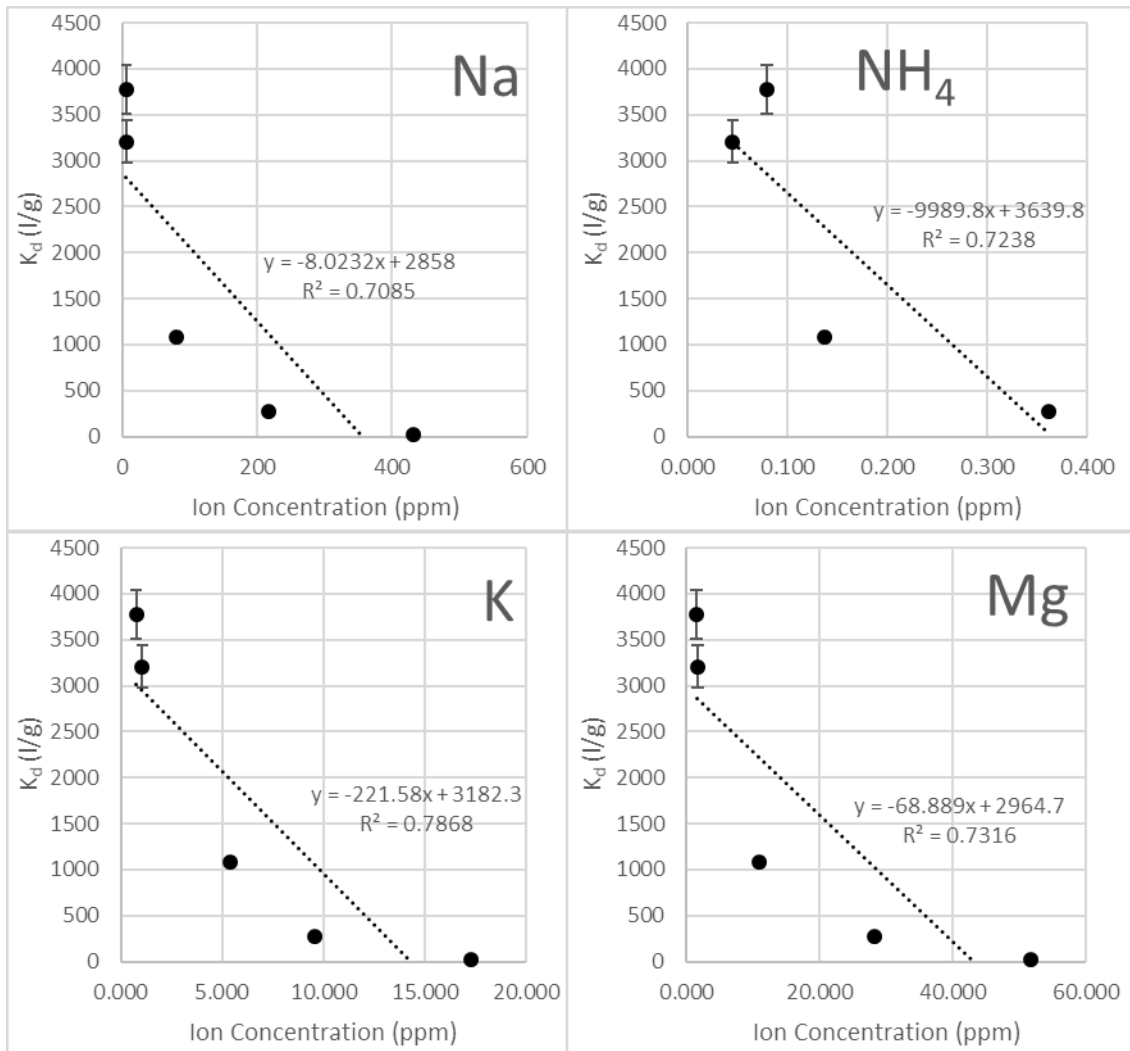


Figure 22. River ion concentration compared to  $K_d$  values.

Looking specifically at samples taken from areas with Gray Lowland soil the trends are again negative as seen in Figure 22, but with steeper slopes than when all river samples were combined. The river samples with the highest  $K_d$  values are surrounded by gray lowland soil. The lowest  $K_d$  sample, the morning Niida river mouth sample has gray lowland soil nearby but is also surrounded by lower inorganic peat soil as well as having the highest ion concentrations of any sample. The afternoon Niida river mouth sample has the second lowest  $K_d$  shown and compared to the morning the increased  $K_d$  is likely a result of reduced competition from other ions.



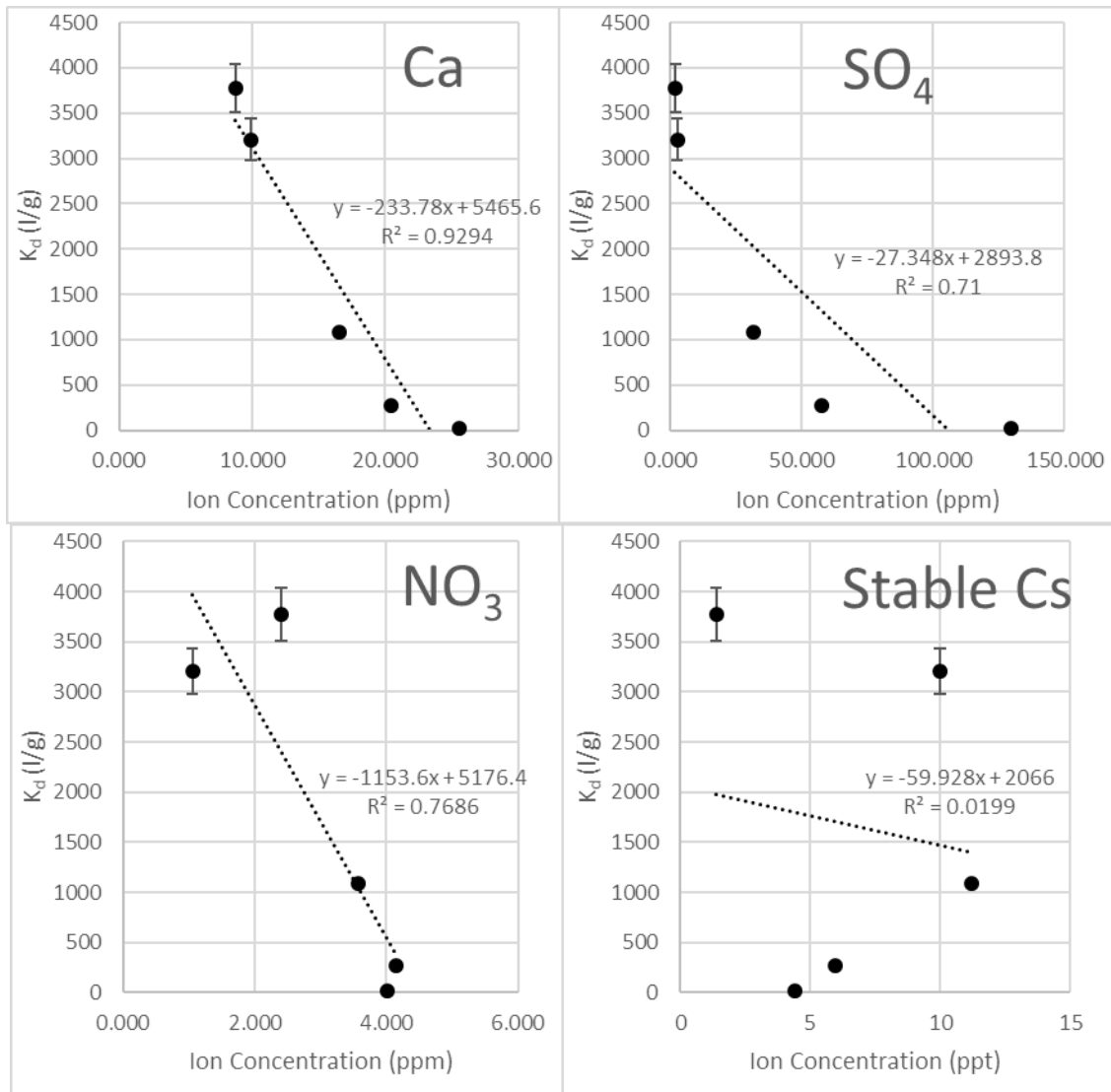


Figure 23. Gray lowland surrounding soil ion concentrations compared to  $K_d$  values.

The variance in mean  $K_d$  of rivers is likely a result of the area the rivers traverse and the different sediments that are accumulated. When comparing different river sample sites surrounding soil seems to have as large of an effect on  $K_d$  as ion concentration. The difference between the river and pond mean  $K_d$  values is likely more driven by the diversity of suspended sediment composition found in rivers than hydrochemistry. In the case of r-Cs binding to FES, competition from K and  $NH_4$  should be considered together if both are present.  $NH_4$  has been shown to have an approximately 5 times higher affinity for FES than K (Konoplev 2020, Uematsu

et al. 2015). Figure 24 shows the comparison of  $K_d$  to the combined  $NH_4$  and K concentrations.

The effects of K and  $NH_4$  are much more prevalent in the rivers than on the ponds. The relatively static ion concentration in ponds is likely why there is little change in  $K_d$

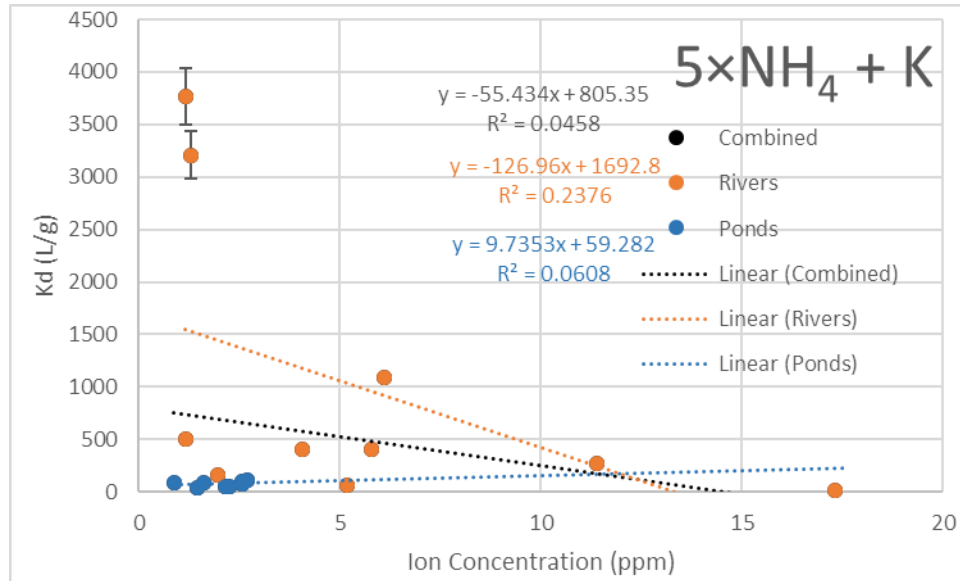


Figure 24.  $K_d$  compared to combined  $NH_4$  and K concentrations.

Using the concentrations found for K and  $NH_4$  the Radiocesium Interception Potential (RIP) can be found according to the following simplified equation:

$$K_d = \frac{RIP(K)}{[K] + 5 \times [NH_4]}$$

Table 13 shows the results of using this equation to calculate the RIP for each sample.

Table 13. Results of RIP calculation (mmol/kg)				
River	Abukuma	Maeda	Niida*	
Upstream	191051	156423	127443	
Middle**	52767	9903	16869	
Mouth	88997	10135	AM-6964 PM-94999	
Pond	Funasawa 1	Funasawa 2	Suzuuchi	Inkyozaka
June 6	3076	3220	2215	1717
July 10	8187	8998	6209	4438
*Niida morning mouth sample had no detectable $NH_4$ .				
**The Hiso River sample is listed as the middle sample for Niida River.				

Uematsu et al., 2015 tested 58 different Japanese soils and found RIP values ranging from 80 mmol/kg to 4009 mmol/kg and a mean of 1140. The calculated values from ponds in this study are approximately in that range except the Funasawa 1, 2, and Suzuuchi July 10<sup>th</sup> samples. The rivers are all much higher, by several orders of magnitude in most cases. The large discrepancy between this study and Uematsu et al. is likely a result of comparing the filtered suspended sediment in this study to the soil-based experiments that were performed by Uematsu et al. It is interesting to note that the ponds were near the range of values found by Uematsu et al. which may be a result of the more limited variation of suspended sediment that would be found from more stagnant pond water while the rivers should have a much more diverse makeup of soils, minerals, and organic matter. The larger area covered by rivers also means the possibility for picking up hot glassy particles is higher. What portion of measured r-Cs is bound to the hot glassy particles and the rate that r-Cs is leached from them requires further research. The hot glassy particles are approximately 2  $\mu\text{m}$  (Adachi, 2013) so they are readily filtered from water samples with sediment. Glassy particle contribution to the filtered sediment measurement could be affecting the calculation of  $K_d$  due to increased sediment activities. Raised  $K_d$  values due to glassy particles would be overestimating the capability of soils and sediments to fix r-Cs resulting in faster than expected migration potentially affecting remediation plans and strategies.

## CONCLUSION

Data on activity concentrations of Cs-137 (dissolved and particulate) from rivers, ponds and a reservoir and the distribution between solid and liquid phases was analyzed and found to vary between the different bodies. Maeda and Niida rivers had the highest  $K_d$  values of over 3000 L/g at the upstream sample points. Both Maeda and Niida had  $K_d$  decrease as the river flowed downstream to values below 300 L/g. Total r-Cs activity concentration increased in the Niida River from 0.06 Bq/L at the Hiso sample point to around 0.8 Bq/L at the mouth. Maeda river increased from 0.1 Bq/L to 0.25 Bq/L as it flowed downstream. Abukuma River  $K_d$  was consistent around 400 L/g at the two upstream sample points and increased at the mouth to over 1000 L/g. The total r-Cs activity concentration in Abukuma was highest at the mouth (0.04 Bq/L) and lowest at the middle sample point (0.02 Bq/L). The middle sample point, however, may have had lost sediment during the sampling. If sediment was lost, then Abukuma river total r-Cs would likely increase as the river flows downstream. In most cases the river r-Cs was concentrated in suspended sediment rather than dissolved, except for the Niida river morning mouth sample. Total r-Cs activity concentration in the sampled ponds remained constant at approximately 0.2 Bq/L between the two sample dates, except in Suzuuchi pond which increased to over 10 Bq/L.  $K_d$  in the ponds increased between the sample dates from around 50 L/g to around 90 L/g in all the ponds except Suzuuchi was constant at 85 L/g for both samples. All the ponds except Suzuuchi switched from r-Cs being primarily concentrated in solution in June to being primarily found in the sediment in July. Ponds had an order of magnitude higher total r-Cs levels than what was found in the rivers, but lower mean  $K_d$  levels than found in the

river samples. Sakashita reservoir total activity concentration was around 0.02 Bq/L the same level of the Abukuma river. Mean  $K_d$  for the reservoir was 133 L/g which was slightly above the pond  $K_d$  means which ranged from 67 to 78 L/g, but below the river  $K_d$  means which ranged from 633 to 1333 L/g. The reservoir r-Cs was primarily dissolved in the water. Using the sediment core depth profile, the sedimentation rate in Sakashita reservoir was estimated to be 2.56 cm/yr. The Sakashita sediment core reconstruction lacked the peak that was seen in the monitoring survey data from the Japanese Government. The lack of peak may be due to mixing of bottom sediment since the top 20 cm of the sediment cores of the reservoir had constant r-Cs activity throughout. The difference in soil composition between the different ponds, reservoir, and rivers seemed to contribute to differences in  $K_d$  more than the differences in hydrochemistry. When comparing amongst similar soil types the effects of ion competition on  $K_d$  is seen.

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