# CO5-1 Size Distribution and Concentration Change of Antimony in the Atmospheric Aerosols at Sakai, Osaka

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Antimony is a metal element, of which symbol Sb; atomic number 51; atomic weight 121.76; melting point 630.5°C. It is used in alloys with lead in battery plates and flame-proofing compound. Antimony(Sb) is toxic by small amount (100mg) into body. As it exist naturally in the soil we get it as food contamination from which Sb is absorbed into the body by 0.002-1.mg per day. Also the airborne particulate matter, aerosol in the atmosphere contain antimony as the fume from burning of the waste matter. This suggest that the daily inhaling of Sb from ambient atmosphere could influence our health. To estimate the effect of Sb to our health ,here we report the size distribution and the concentration (mean and variation) and monthly concentration change.

The analyzed samples are size-separated aerosols and non size-separated aerosol. The size-separated samples have been collected with 9-size particle size ranges in 1 weak sampling period. The non size-separated samples have been collected daily on membrane filter in 24h sampling period. The sampler of both samples are located at Osaka Prefecture University, at Sakai.

The element of the samples were analyzed by neutron activation analysis using KUR. The Analysis condition for Sb are , Irradiation : 2 hours in Pn2 1MW, detection of gamma ray : <sup>124</sup>Sb 1661kev.

From the analyzed results the size distribution of Sb in atmospheric aerosol can been seen as mostly concentration distribute in very fine range ,  $<\!0.43$   $\mu m(Fig.1)$ . The central diameter and concentrations of samples collected in 2012 are described on table 1.

Table 1 Central diameter and total concentration of Sb collected by 9 particle size separation at Sakai in 2012.

Period(2012)	D50(μm)	TC(ng/m3)
Feb 14-21	0.94	1.7
Apr 3-10	0.68	1.7
Jun 5-12	0.64	1.3
Aug 7-14	0.69	1.2
Oct 9-16	0.67	2.3
Dec 11-18	0.57	3.3

#### **D50:**Central diameter TC:Total Concentration

From the result of concentration change in daily collected(in 2007) samples we have obtained concentration frequency distribution(Fig.2), showing mostly frequent in range, 1-3ng/m3. And we have got the

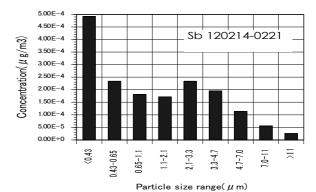


Fig.1 Size distribution of Sb in aerosols collected at Sakai,0214-0221 in 2012.

average and standard deviation,  $2.8 \pm 1.9 \text{ ng/m}^3$ , min=0.5 ng/m³, max=24 ng/m³. This value indicate that mean inhalation of Sb per day is about 0.06 mg.

Monthly changes(mean, sdev., min.,max.) of daily concentration are shown Fig.3. Months in high concentration are Jan.(3.4ng/m³), Oct.(3.5ng/m³), Nov. (3.6ng/m³) and lowest on May(2.0ng/m³). Maximum concentration of daily change in 2007 were observed on September 30th. On this day we observed high concentration in bromine(Br), suggesting the source of fume in high concentration might be waste burning of plastic that contain flame-proofing compound.

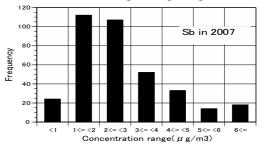


Fig.2 Frequency distribution for daily concentrations of Sb in arosols collected at Sakai, 2007.

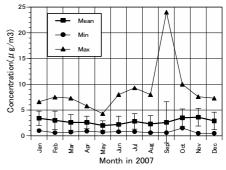


Fig.3 Monthly changeof Sb in aerosols collected at Sakai, 2007.

# CO5-2 Characteristics of Synthetic Calcite Thermoluminescence Studied for Paleoenvironmental Reconstruction of East Asia

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**INTRODUCTION:** Luminescence dating observes the natural accumulated radiation damage caused by radioisotopes such as U and Th as the form of glow after stimulation by heating or lightening. Thermally stimulated luminescence from calcite shows strong red emission. However, thermoluminescence dating of calcite is less popular because of some unknown problems; e.g., sensitivity change of calcite occurred through repeated heating of samples, possible anomalous fading, difference in characteristics of luminescence response against different kinds of radiation (e.g., gamma-ray, beta-ray, alpha-ray, and X-ray). When thermoluminescence characteristics of calcites are examined using natural occurring calcite, their response to the various radiations depends on minor chemistry (Fe, Mg, Mn and Sr). In this study, calcites with controlled impurity concentrations were synthesized and analyzed to evaluate relationship between multiple impurity concentration and thermoluminescence properties quantitatively, together with natural calcites.

**EXPERIMENTS:** Calcite were synthesized from sodium carbonate (>99.8wt%) and calcium chloride (>95 wt%). Iron (II) chloride (>99.0-102.0 wt%), magnesium chloride (>98.0wt%), and manganese chloride (>99.0wt%) were added at different levels to produce calcites with different amount of impurity. Chemical composition of resultant calcite were measured by LA-ICP-MS, and the results were reported in KURRI progress report 2014. Gamma irradiation was carried out at the 60Co gamma irradiation facility at Kyoto University Research Reactor. Alpha and beta ray irradiations were carried out with disc-type <sup>241</sup>Am and <sup>90</sup>Sr source, respectively. Given doses by Gamma and beta irradiation were estimated by quartz irradiated together with synthesized calcite. A given dose by alpha irradiation was estimated by alpha track counting. Then luminescence emission from calcite was measured by the luminescence reader MOSL-22, and dose was estimated by the external x-ray source, whose dose rate is calibrated as 0.1 Gy/sec with quartz. This measured dose on calcite was compared to the given dose on quartz, and the ratio (calcite/quartz) was calculated for each irradiation (gamma, beta). Results on alpha irradiation will be reported later.

RESULTS: Calcite/quart ratios in dose estimate by luminescence were different among samples. However, ratios by beta-ray and by gamma- rays are more or less concordant (Fig. 1). When Calcite/quartz ratios by gamma dose is plotted against Mn concentration, they show negative correlation (Fig. 2). Thermoluminescence characteristics of calcite is controlled by the Mn impurity. Therefore, measurement of Mn concentration is inevitable for calcite thermoluminescence dating.

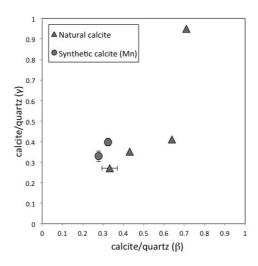


Fig. 1. Relationship between calcite/quartz ratios by beta and gamma irradiation.

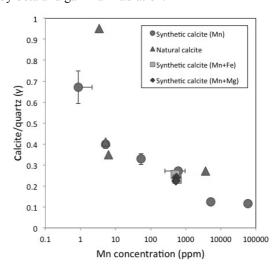


Fig. 2. Calcite/quartz ratios by gamma irradiation are plotted against Mn concentration.

## CO5-3

## Study of Earth and Planetary Matters by Thermoluminescence

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**INTRODUCTION:** Induced TL (thermoluminescence), the response of a luminescent phosphor to a laboratory dose of radiation, reflects the mineralogy and structure of the phosphor, and provides valuable information on the metamorphic and thermal history of meteorites. Thermal metamorphism causes the production of feldspar, the major TL phosphor in ordinary chondrites, by the devitrification of feldspathic mesostasis. TL sensitivity normalized by Dhajala chondrite was measured at around 150 °C or 80 °C in the induced TL intensity. Primitive chondrites of petrologic subtype  $\leq 3.4$  with coefficient of varia-tions ( $\sigma$  as a percentage of the mean) over 50%, of fayalite in the olivine have low TL sensitivities under 0.1. The TL sensitivity was usually used to determine petrologic subtype of unequilibrated ordinary chondrites [1]. On the other hand, the TL sensitivity decreases 10-fold after shock-loading to 25-32 GPa [2], and terrestrial weathering makes the TL sensitivity decrease 16-fold at maximum in Antarctic chondrites [3]. Then these secondary altered chondrites were reported to have low TL sensitivities, equivalent to primitive chondrites [4]. Until now, the induced TL of ordinary chondrites has been measured above room temperature. Last fiscal year, we preliminarily measured low temperature TL from liquid nitrogen temperature of twenty-one Asuka ordinary chondrites under 0.1 in TL sensitivities. This time we measured low temperature TL of additive two Asuka ordinary chondrites, A881397 (LL3.3) and A881236 (remarkably shocked). Also this time, we also measured induced TL of unshocked and artificially shocked terrestrial feldspars, Minas Genas albite, Shiga albite and Niigata plagioclase, to investigate the shock effects. This year, there is no request from NIPR to determine subtype because samples, judged to be unequilibrated, became a little. Then we didn't try to measure new chondrites.

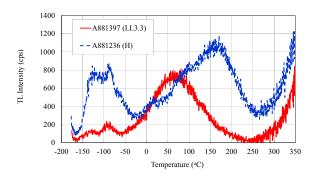


Fig.1. induced TL glow curves of A881397 (LL3.3) and a remarkable shocked chondrite of A881236 (H).

**RESULTS:** Fig.1 shows TL glow curves of a primitive ordinary chondrite of A881397 (LL3.3) and remarkable shocked chondrite of A881236 (H). There were also two peaks around -120 and -80°C lower than room temperature. However, we could not find the qualitative tendency in induced TL glow curves in all twenty-three chondrites.

Induced TL of terrestrial feldspars, Minas Genas albite, Shiga albite and Niigata plagioclase, showed very high intensity. Shocked samples showed relatively less intensity than unshocked samples. However, we unfortunately couldn't find clear difference in the induced TL glow curves between the unshocked and shocked terrestrial feldspars. Fig.2 shows induced TL glow curves of unshocked and artificially shocked Shiga albite.

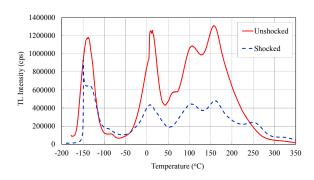


Fig.2. Induced TL of unshocked and artificially shocked Shiga albite.

**ACKNOWLEGEMENTS:** Over twenty years, we have measured induced TL of 407 Antarctic unequilibrated ordinary chondrites, Yamato 242 chondrites (LL3: 30, L3: 113, H3: 99), Asuka 163 chondrites (LL3: 39, L3: 48, H3: 76) and ALH 2 chondrites (L2:, 2), which TL analysis has been requested from NIPR, under the Visiting Researcher's Program of the Research Reactor Institute, Kyoto University We have found 50 samples to be primitive chondrite under  $\leq$  3.4 in them. This time we finished to apply to this program. We would like to thank to not only staffs in the Research Reactor Institute, who has supported us, but also all staffs in the Research Reactor Institute, Kyoto University.

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## CO5-4 Possible Removal of Tritium by Reaction of Magnesia and Water with Tritium

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INTRODUCTION: Characteristics of tritium water (HTO) are almost the same as water (H<sub>2</sub>O) and deuterium water (D<sub>2</sub>O or HDO). The slight difference of the characteristics (e.g. the boiling temperature and electrolysis etc) have been used to fraction three waters. It was recently reported that manganese oxide adsorbed T<sup>+</sup> [1]. However, it is still difficult to fraction the three waters. Magnesium is one of the main elements composed of the earth. Magnesium oxide (MgO) easily reacts with water molecules and magnesium hydroxide (Mg(OH)<sub>2</sub>) is produced, immediately. Additionally, if MgO is placed in the air, MgO will react with water molecules and become  $Mg(OH)_2$  [2]. The mineral name of  $Mg(OH)_2$  is brucite. Brucite is produced by weathering of a mafic rock with other clay minerals on the earth's surface. Mg(OH)<sub>2</sub> was dehydrated at about 300 °C [3]. Recently, there are some reports about the dehydration of Mg(OD)<sub>2</sub> by the X-ray or the neutron diffraction with the heating system. The temperature of the dehydration was about 330 °C [4, 5]. Although the temperature is not certain, the dehydration temperature of Mg(OD)<sub>2</sub> was higher than that of Mg(OH)<sub>2</sub>. When MgO reacts with HTO, the reaction rate and dehydration temperature of Mg(OT)2 might be different from those of Mg(OH)<sub>2</sub>.

We investigated that the reaction rate of Mg(OT)<sub>2</sub> is different from that of Mg(OH)<sub>2</sub>, as a first step.

**EXPERIMENTS:** We used MgO (light and heavy) to be on the market. MgO (light) and (heavy) means a fine particle and a coarse particle, respectively. We prepared six glass bottles with stopper. In every three bottles, 1 g of MgO (light) and 2g of MgO (heavy) were put, respectively and 10 cm<sup>-3</sup> of water with HTO was added in six bottles. These bottles are left for about 1, 4 or 24 h. After the purpose reaction time, suspension was filtered by 0.2 μm of a disposable filter. We diluted 0.1 cm<sup>-3</sup> of supernatant by 20 cm<sup>-3</sup> of the liquid scintillator. The tritium concentration of each sample was measured by a scintillation detector (Packard, Liquid Scintillation Analyzer).

**RESULTS:** When the tritium concentration of supernatant is smaller than that of the initial solution, more amount of HTO reacts with MgO than that of H<sub>2</sub>O. If the tritium concentration of supernatant is larger than that of the initial solution, H<sub>2</sub>O will be preferred to HTO in the reaction with MgO. In the case of the similar tritium concentration of the initial solution and supernatant, almost the same amounts of HTO and H<sub>2</sub>O react with

MgO.

The tritium concentration of supernatant is shown in Table 1. For MgO (light), the tritium concentration decreased for 1 and 4 h after MgO contacted with water with HTO. The removal ratio was about 5.1%. After 24 h, the tritium concentration of supernatant was almost the same as that of the initial solution. The removal ratio was about 1.5 %. If MgO contacts with water with HTO for a few hours, the reaction of HTO with MgO might be quicker than that of H<sub>2</sub>O. MgO might react with slightly more HTO than H<sub>2</sub>O in water for 4 h. After the synthesis of brucite, the exchange reaction would occur between T<sup>+</sup> or H<sup>+</sup> in brucite and H<sup>+</sup> or T<sup>+</sup> in water. The exchange reaction might achieve the equilibrium for 24 h. The tritium concentration of supernatant after 24 h might be similar to that of the initial solution

For MgO (heavy), we used 2g of MgO, because it was expected that the reaction of MgO and water with HTO would be slow. After 4 h contact, the tritium concentration of supernatant was a smaller than that of the initial solution. The ratio of 2.4 % of T<sup>+</sup> decreased from the solution. This removal ratio was smaller than that of MgO (light). Since MgO (heavy) is a coarse particle, MgO (heavy) would not sufficiently react with water with HTO. The removal ratio of MgO (heavy) would be smaller than that of MgO (light). In 24 h, the removal ratio was about 1.9 %. The tritium concentration of supernatant in MgO (heavy) and the initial solution is almost the same in the case of MgO (light). Probably, T<sup>+</sup> or H<sup>+</sup> in brucite will be exchanged for H<sup>+</sup> or T<sup>+</sup> in water.

Table 1 Change of the tritium concentration in supernatant by the contact time of MgO and water with tritium.

N	IgO(light)	MgO(light)	MgO(light)
time (h)	1	4	24
concentration (Bq)	522066	522440	541768
removal ratio (%)	5.12	5.05	1.54
	MgO (heavy	) MgO (hea	avy) initial
time (h)	4	24	
concentration (Bq)	533224	539834	550232
removal ratio (%)	3.09	1.89	

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## CO5-5 Application of Neutron Activation Analysis to Micro Gram Scale of Solid Samples

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INTRODUCTION: Instrumental neutron activation analysis (INAA) is often used in cosmochemistry, where meteorites are the objects for INAA. Chondritic meteorites (chondrites) and iron meteorites contain relatively high contents of Co and Ir compared with those in the earth crust. As Co and Ir have high sensitivity in INAA, they can be good markers for the identification of such extraterrestrial materials. In NAA of chondrites, a few tens mg of specimen is commonly used. For such a case, a few hundred µg kg<sup>-1</sup> of Ir and a few hundred mg kg<sup>-1</sup> of Co can be reliably determined. When an extremely small size (e.g., micro gram) of samples such as micrometeor-ites recovered on the Earth surface and tiny particles re-turned from extraterrestrial asteroids are to be analyzed by INAA, the conventional INAA procedure used for a few tens mg is not suitable. For such tiny samples, neu-tron irradiation with high neutron flux and long irradia-tion time (namely, high neutron dose) is required. Re-cently we have presented the INAA procedure for micro gram scale of solid samples [1]. In this report, the degree of increase in isotopic abundance induced by neutron irradiation is described in detail.

RESULTS: As INAA is a nondestructive method of elemental analysis, the same sample once subjected to INAA can be reused for different analytical purposes. For such a case, effects caused by neutron irradiation are of concern. Major concerns are the induced radioactivity and the increase in isotopic abundance including the production of long-lived radioactive nuclides. Scientifically, the latter case is important by far and, therefore, is considered here. In order to make the evaluation of degree of the increase in isotopic abundance meaningful, used are the data from the experiment [2], where 1.66 mg of a tiny grain from the asteroid Itokawa was irradiated by neutrons under the condition (19 h-irradiation,  $\phi_{th}$ : 8.2 × 10<sup>13</sup> n cm<sup>-2</sup> s<sup>-1</sup>,  $f\phi$ :  $3.9 \times 10^{13}$  n cm<sup>-2</sup> s<sup>-1</sup>, and 5MW-operation). Assuming the elemental composition of bulk LL chondrite [3] for this grain, the number of produced nuclides with neutron irradiation was calculated based on the Monte-Calro simulation code (MVP 2.0) [4] and the reaction cross section data (JENDL-4.0) [5]. Both stable and unstable (radioactive) nuclides could be produced and the result is summarized in Table 1. In calculation, the position of control rod, temperature of the reactor core, and the combustion rate of nuclear fuel were all taken into consideration, because the neutron irradiation of the experiment [2] was performed near the reactor core and, therefore, these factors

affect the neutron energy spectrum. The produced nuclides in Table 1 are grouped into stable nuclides (<sup>21</sup>Ne, <sup>22</sup>Ne and <sup>38</sup>Ar) and long-lived radioactive nuclides (<sup>36</sup>Cl, <sup>26</sup>Al and <sup>10</sup>Be). These nuclides are typical cosmogenic nuclides and radionuclides produced by nuclear reactions triggered by cosmic rays and commonly detected in extraterrestrial materials like meteorites.

In Table 1, the calculated values are compared with numbers of corresponding nuclides observed in extraterrestrial samples. For stable nuclides (of noble gases), measured values for a different Itokawa grain are given for comparison [6]. It is well acknowledged that noble gases are extremely sensitive in mass spectrometry. As seen in Table 1, calculated values and measured values in the Itokawa grain are mostly comparable for all three nuclides. For long-lived radionuclides, concentrations in the Gold Basin L4 chondrite (a shower sample, UA-1188) [7] are given for comparison in Table 1. These data were obtained by accelerator mass spectrometry. The measured values are smaller or much smaller than the calculated values, with the difference varying by an order to six orders of magnitude. Evidently the reuse of neutron-irradiated samples should not be allowed for noble gas mass spectrometry and accelerator mass spectrometry for the study on noble gas nuclides and long-lived radioactive nuclides, respectively.

Table 1 Production of some cosmogenic nuclides from 1.66  $\mu g$  of LL chondrite by neutron-induced reaction in INAA $^a$ 

Target nuclides	Reaction	Produced nuclides	Number of Produced nuclides	Number of nuclides (original)		
Stable nuclides (noble gas)-production						
$^{24}Mg$	(n,α)	<sup>21</sup> Ne	$1.58 \times 10^{8}$	$1.15 \times 10^{7}$		
<sup>25</sup> Mg	(n,α)	<sup>22</sup> Ne	$5.21 \times 10^{7}$	$3.49 \times 10^{8}$		
<sup>37</sup> Cl	(n,γ), β <sup>-</sup>	<sup>38</sup> Ar	$7.97 \times 10^{6}$	$1.78 \times 10^{7}$		
Radioactive nuclides-production						
35Cl	(n,γ)	<sup>36</sup> Cl	$2.53 \times 10^{9}$	$9.06 \times 10^{2}$		
<sup>39</sup> K	(n,a)	<sup>36</sup> Cl	$3.24 \times 10^{7}$	$9.06 \times 10^{2}$		
<sup>27</sup> Al	(n,2n)	<sup>26</sup> Al	$1.30 \times 10^{5}$	$3.73 \times 10^{3}$		
<sup>13</sup> C	(n,a)	<sup>10</sup> Be	$3.03 \times 10^4$	$2.07 \times 10^3$		

<sup>&</sup>lt;sup>a</sup>See text for detail.

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# CO5-6 Chemical Variations of Miller Range 07710 (L4) Collected in the Ice at Miller Range, Antarctica

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INTRODUCTION: Most of meteorites are currently collected from cold and hot deserts. As these meteorites may have resided on the Earth's surface, they have experienced variable degree of terrestrial weathering. Terrestrial weathering obscures petrographic and chemical features, leading to misclassification and misunderstanding of geochemistry and cosmochemistry of these meteorites. In addition, misinterpretations probably result from sampling effect [e.g., 1] A large data set for chondrites were obtained by using 250-300 mg of sample [e.g., 2]. This amount of sample is not enough to obtain representative chemical compositions [1]. Therefore, we examine how terrestrial weathering and sampling problem affect chemical classification by using Miller Range (MIL) 07710 (L4) which was collected in the ice at Miller Range, Antarctica.

**EXPERIMENTS:** Eight different aliquots were taken from MIL 07710 and carefully ground in an agate mortar. Eight powder samples were analyzed by using INAA and IPAA at the Kyoto University Research Reactor Institute (KURRI). Neutron irradiations were successively performed two times (10 s and 4 hrs irradiations). For IPAA, we used a linear electron accelerator at KURRI. Electrons were accelerated by the linear accelerator to about 20 MeV.

**RESULTS:** Twenty seven elements could be determined by using both INAA and IPAA. MIL 07710 was grouped into L4 based on petrological and mineralogical studies [3]. The homogeneity of chemical compositions was found to be less than 10% ( $1\sigma$ ; n=8) for most lithophile elements analyzed, In contrast, the heterogeneity exceeding 10% was found in siderophile (Co, Ni, Os, Ir and Au) and chalcophile elements (Se). This observed heterogeneity is considered to be due to sampling problem and/or terrestrail weathering. We investigated how this heterogeneity affect chemical classification of this meteorites. Fe/Mn and Fe/(Fe+Mg) ratios of eight different aliquots of this meteorite are plotted in Fig. 1. As seen in Fig. 1, two samples weighing about 1 g fall within the field of L, while Fe/Mn and Fe/(Fe+Mg) ratios for other samples with having less than 0.6 g fall in the ranges of L and LL. Eight samples display a positive correlation between Fe/Mn and Fe/(Fe+Mg) ratios. Such a trend can be explained by different modal abundances of metal and/or terrestrial weathering of metal. Jarosewich [4] compared major element abundances of fall meteorites with those of find meteorites from Antarctica and found that total Fe contents of Antarctic H chondrites are lower than those of fall H chondrites, but that total Fe contents of Antarctic L

and LL chondrites cannot be distinguished from those of fall L and LL chondrites. Therefore, terrestrial weathering could not explain such a positive correlation seen in Fig. 1. Fe/Mn and Fe/(Fe+Mg) ratios for mixtures of metal and olivine, and metal and low-Ca pyroxene were calculated, and the calculated results are shown in Fig.1. Different modal abundances of metal can explaine observed positive correlation among eight samples. It is concluded that metal is heterogenous distributed in sample weight less than 0.6 g.

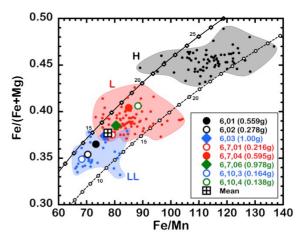


Figure 1. Fe/Mn vs. Fe/(Fe+Mg) diagram for ordinary chondrites. Solid and dash lines represent mixing between metal [7] and low Ca-pyroxene [8], and metal [7] and olivine [8]. The proportional contributions of metal are shown as percentages among the mixing lines. Parentheses in sample position represent homogenized mass.

Siderophile element abundances are also useful for classification of ordinary chondrites [2]. Siderophile element abundances for most MIL 07710 samples analyzed in this study do not fall within the any range of ordinary chondrites. It is well known that sulfide and metal are more susceptible to terrestrial weathering than silicate [5] and that some Antarctic chondrites show fractionated abundance patterns for chalcophile and siderophile elements [6]. Therefore, observed scattering of siderophile element are attributed to terrestrial weathering.

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## CO5-7 A study on Evaluation of Radioactive Cesium Behavior in Fukushima

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#### INTRODUCTION:

After the Fukushima Nuclear Power Plant (FNPP) accident in March of 2011, radioactive cesium 137 (137Cs) was deposited over a large area in eastern Japan. Most of the <sup>137</sup>Cs remained in environmental soils and sediments, because of its 30-year physical half-life and strong adsorptive properties. A portion of the <sup>137</sup>Cs within these soils and sediments was transported downstream with suspended solids (SS). Additional sites affected by <sup>137</sup>Cs appeared downstream of the contaminated watershed (Sato et al., 2013), and most <sup>137</sup>Cs had been accumulated in Ogaki dam, along with suspended matter which originated from the watershed soils (Kitamura et al., 2014). The purpose of this research was to quantitatively evaluate the form and distribution of <sup>137</sup>Cs in watershed soils and river/lake waters. Based on the measurement results, the factors facilitating the determination of the solid-phase <sup>137</sup>Cs concentration were discussed.

#### **SURVEY and EXPERIMENT METHODS:**

We collected samples from the Natsui River Basin (750 km²), located 40 km away from FNPP. The Natsui River is the water resource for the 160,000 people living in Iwaki city. River and lake waters, surface soils, and sediments were sampled using buckets, stainless shovels, and Ekman-berge sediment samplers, respectively. Surface soils from a depth range of 0-5 cm were mixed and combined from five point samples into a single sample.

The collected soil samples were desiccated by a freeze dryer(FD-81), and were then divided by particle size using sieves (2000, 850, 425, 250, 106, 75 and 25 µm) after removing particles over 2 mm in diameter. Measurements of <sup>137</sup>Cs and organic carbon content (NC-22F) were carried out for the separated particles. Water samples (20 L) were separated into SS residue and filtrate through filtration, using a 1 µm glass fiber filter (GF/B). The <sup>137</sup>Cs contained in the filtrate was concentrated into a disk (Empore TM Cesium Rad Disk) by forcible filtration. Afterwards, the <sup>137</sup>Cs in SS residues within the filters and in the disks were measured by a Ge semiconductor detector (GC4020/3020, CANBERRA).

### **RESULTS and DISCUSSIONS:**

It was previously reported that <sup>137</sup>Cs in soils were dependent on the specific surface area of the soil particles (He and Walling, 1995). In order to analyze the adsorption characteristics of <sup>137</sup>Cs in soils, the <sup>137</sup>Cs (logarithm value) in each particle size category was plotted against representative particle size (logarithm value). A regression line slope of S=0 indicates that the <sup>137</sup>Cs was independent of particle size, while S =-1 indicates that the soil <sup>137</sup>Cs was dependent on the specific surface area of all

particles. Regression line slopes from the forest soil samples ranged from -0.8 to -0.4. This suggests that the <sup>137</sup>Cs in forest soils tend to be relatively dependent on the specific surface area. Conversely, regression line slopes from the bottom sediment samples in dam and paddy soils were almost zero. It was considered that the <sup>137</sup>Cs was equalized between different particle sizes, because the soil particles were more frequently exposed to water.

<sup>137</sup>Cs in each particle size, plotted against the organic carbon content in the forest soil at upstream site (Fig. 2). The result showed there was a strong correlation between these variables, and that the <sup>137</sup>Cs per organic carbon (*i.e.*, regression slope) was relatively constant (2,500 Bq/g -organic carbon) regardless of the season. This value was strongly related to the <sup>137</sup>Cs deposition, indicating that the organic carbon was an essential qualitative factor influencing <sup>137</sup>Cs.

Ratios of particulate <sup>137</sup>Cs in water were about 90% in upstream site, and about 20% in Kodama dam site, even during the normal river water stage. These results suggest that SS, as a carrier substrate, had a significant impact on the total <sup>137</sup>Cs loading to the sea. Calculated K<sub>d</sub> values (<sup>137</sup>Cs in SS residue divided by the <sup>137</sup>Cs in filtrate) were distributed from 0.066 to 62 ML/kg. Because there was not enough time to dissolve from particulate <sup>137</sup>Cs to dissolved one, the K<sub>d</sub> was significantly higher than those values of previous studies. These results indicate that the behavior of <sup>137</sup>Cs in the natural environment is dominated by the movement of particles during floods, as well their adsorption on organic matter particles.

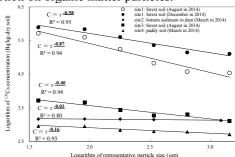


Fig.1 Relationship between <sup>137</sup>Cs and the particle size

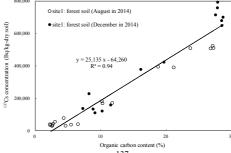


Fig.2 Relationship between <sup>137</sup>Cs and the organic carbon **REFERENCES:** 

[1]He, Q. et al., Journal of Environmental Radioactivity, **30/2**(1995), 117-137.

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