

SGC053-01

Room:304

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## Effect of tectonic environment on chemical composition of OIBs in the Pacific: implication for a deep mantle origin

Gen Shimoda<sup>1\*</sup>

<sup>1</sup>Geological Survey of Japan, AIST

The Pacific Ocean is known to contain numerous ocean islands, seamounts and several oceanic plateaus and rises those may have formed during the large-scale magmatic event in the South Pacific. Proposed origins for this magmatism can be divided into two categories. One is mantle plume from the Earth's deep interior that is governed by mantle dynamics and the plume sometimes referred to as "superplume". The other is upper mantle process that is controlled by lithospheric dynamics, such as surface lithospheric fractures, melt focusing under broad upwellings condition and small-scale sublithospheric convection. In either case, documenting the large-scale magmatic event over both space and time may provide clues into understanding material recycling throughout the silicate Earth.

The South Pacific region is referred to as a "superswell" or the "South Pacific Isotopic and Thermal Anomaly" (SOPITA) due to its unusually shallow ocean floor, evidence of active intraplate volcanism, and the occurrence of isotopically anomalous magmas. These geophysical and geochemical features are usually attributed to a plume activity or lithospheric dynamics. Although it is unclear whether the process that is now operate in the South Pacific has been active since the Cretaceous, distinct isotopic signatures of the Cretaceous seamounts can be traced back to the magmas of SOPIAT. It might follow that single process has been active since the Cretaceous.

The notable geochemical character of this magmatism is quadratic correlation between elemental ratios ( $La/Yb$ ,  $Sr/Y$ ,  $Nb/Zr$ ,  $Ta/Zr$  and  $Th/Ta$ ) of the seamounts/island and "relative age" that can be an index of the thickness of lithosphere beneath hot spots. As the thickness of oceanic lithosphere is a function of square root of its age, these correlations suggest importance of tectonic environment to determine the chemical composition of the magmas. In addition, these correlations suggest the genetic relationship between oceanic plateaus and island/seamount chain because the elemental ratios of oceanic plateaus are on the trends. Other important geochemical feature is that the elemental ratios of mid-ocean ridge basalts (MORBs) from the East Pacific Rise (EPR) are not on these trends. This observation suggests that source material of the EPR MORBs is different from that of the seamounts and islands. Since the source material of EPR MORBs can have a representative composition of upper mantle beneath the South Pacific, this difference could imply the material flow from the deep mantle. Therefore melting of mantle plume from the lower mantle, which melting condition depends on thickness of lithosphere, can be a plausible origin of the magmatism in the South Pacific.

Keywords: Pacific, hot spots, ocean islands, Pb-Nd-Sr isotopes, superplume

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## K and U budget in the mantle

Takeshi Hanyu<sup>1\*</sup>

<sup>1</sup>IFREE, JAMSTEC

K, U and Th concentrations in bulk silicate Earth (BSE) are significant for isotope geochemistry, including noble gases and Pb isotopes, as well as for geothermal study as internal heat sources. While the concentration of U in BSE has been estimated from the chondritic value, K concentration of BSE is poorly constrained due to its volatility during formation of the Earth. Instead, K concentration in BSE has been calculated using U concentration in BSE multiplied by the canonical K/U ratio (13000) deduced from the relatively uniform K/U of crustal and mantle-derived rocks (e.g., O'Nions and Oxburgh, 1983). However, if subducted and dehydrated slab with low K/U occupied a considerable volume in the mantle, the presence of this "hidden" reservoir would result in a significant reduction in K/U, and hence in the total amount of K, in BSE compared to the previous estimates (Lassiter, 2004). This hypothesis has been debated (Arevalo et al., 2009). However, He-Ne-Ar isotope systematics of the HIMU reservoir, which was produced from subducted old oceanic crust, demonstrate that K/U of this reservoir should be much lower (3000) than the canonical K/U ratio (Hanyu et al., in submission), supporting Lassiter (2004)'s hypothesis.

In this presentation, I discuss the impact of subducted oceanic crust on the K and U(+Th) budget in the mantle. Since the subducted oceanic crust is enriched in U, but not in K by dehydration, the preservation of the subducted oceanic crust in the mantle, that is HIMU reservoir, affects the K and U budget in the silicate Earth. Mass balance calculations of K and U were conducted, assuming four major reservoirs, continental crust, depleted mantle reservoir, primitive mantle reservoir and subducted oceanic crust, after Lassiter (2004). The differences between the model presented here and that presented by Lassiter (2004) are twofold. (1) We assume variable U concentrations in the subducted oceanic crust, from U-poor dehydrated N-MORB (0.05 ppm) to U-rich bulk crust observed in natural eclogite (0.12 ppm). (2) We assume some depletion in U and K for primitive mantle reservoir, because it is not necessarily unmodified BSE, but may be moderately depleted due to it making a small contribution to the formation of continental crust (Class and Goldstein, 2005).

The estimated K/U of BSE is modified from canonical value of 13000 down to 8000-10000 by taking the presence of subducted slab into consideration, irrespective of the values chosen for the compositions of the abovementioned reservoirs, if all the oceanic crust ever subducted has accumulated, occupying 6-9 % of the mantle. In this case, K concentration in BSE is also lower than the previous estimate of 250 ppm. The fraction of primitive mantle reservoir in the silicate mantle is also estimated from the mass balance calculations. It is highly dependent on the concentration of U in the subducted crust. In particular, if U concentration in the subducted oceanic crust is relatively low (0.05 ppm), the primitive mantle reservoir should occupy at least 30% in the mantle to balance the K and U budget in the silicate Earth. This suggests that considerable amount of primitive mantle reservoir has been unmixed and isolated in the convecting mantle.

### References -

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Keywords: potassium, uranium, mantle, subducted slab

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## Noble gases in olivines in Udachnaya kimberlite, Siberia

Fumihiko Kitamura<sup>1\*</sup>, Hirochika Sumino<sup>1</sup>, Kyosuke Matsufuji<sup>1</sup>, Keisuke Nagao<sup>1</sup>, Ichiro Kaneoka<sup>2</sup>, Vladimir S. Kamenetsky<sup>3</sup>, Maya B. Kamenetsky<sup>3</sup>

<sup>1</sup>GCRC, Grad. School Sci., Univ. Tokyo, <sup>2</sup>ERI, Univ. Tokyo, <sup>3</sup>University of Tasmania, Australia

Kimberlite is known as host rock of diamonds. Although the origin of kimberlite magma is considered to be deeper than 150 km in the mantle where diamonds formed (e.g., Dawson, 1980; Haggerty, 1994), it has not been well constrained yet (e.g., Price et al., 2000; Chalapathi Rao et al., 2004; Smith, 1983).

Isotopic ratios of noble gases in various components in the Earth are significantly different resulting from that they are sensitive to addition of radiogenic/nucleogenic isotopes to primordial ones due to their low concentrations. Because noble gases can be measured with a high sensitive mass spectrometer, they are useful tracers to constrain origins of samples derived from deep interior of the Earth. Sumino et al. (2006) analyzed olivine phenocrysts in the Udachnaya kimberlite from Siberia, and clarified that they contain plume-derived noble gases. In this work, we analyzed other new sets of samples from the same kimberlite pipe to further investigate noble gas characteristics of the Udachnaya kimberlite magma.

It is important to separate several noble gas components within a sample, because there is not only intrinsic component of magma source but also secondary components acquired from surrounding materials or accumulated radiogenic isotopes produced from the decay of radioactive elements such as U, Th, and <sup>40</sup>K. We separated olivine phenocrysts that would have been less affected from the latter two components than other minerals. Since magmatic noble gases concentrated into fluid inclusions in the phenocrysts, we employed crushing method to extract noble gases selectively from them. We also applied heating method that extracts all noble gases in the phenocrysts. In this work we developed new crushing apparatus to reduce the contaminating atmospheric noble gases, which were adsorbed on the inner surface of the apparatus.

Crushing the aliquot of olivine separates reported in Sumino et al. (2006) yielded larger contributions of radiogenic <sup>4</sup>He and nucleogenic <sup>21</sup>Ne than the previous results. Since they exist within olivine crystal lattice, the new crusher shows higher crushing efficiency than the old one, resulting in a larger contribution of the lattice-hosted components relative to the inclusion-hosted ones. However, the total abundances of radiogenic <sup>4</sup>He and nucleogenic <sup>21</sup>Ne extracted from both crushing and heating indicate that the olivines studied in this work contained more radiogenic/nucleogenic components than those in the previous ones. The results suggest different noble gas isotopic compositions among the olivine crystals from the same kimberlite.

Additionally we measured olivine samples separated from another Udachnaya kimberlite. The results showed distinct behavior during stepwise crushing. Magmatic He isotope ratio is similar to that of subcontinental lithospheric mantle, and radiogenic <sup>4</sup>He and <sup>40</sup>Ar contributions increase with the progress of crushing. The results are similar to those of the previous work. Meanwhile, Ne isotope ratios deviate from typical mantle trends toward larger contribution of nucleogenic <sup>21</sup>Ne in a Ne three-isotope plot. This is clear contrast to the previous results, in which Ne isotopic feature is explained by a two-component mixing between air and less-nucleogenic mantle component than the MORB source (Sumino et al., 2006). Since the grain sizes of the new olivine separates are relatively larger than the proper phenocrystic olivines which should be smaller than 200 microns in size (Kamenetsky et al., 2008), there might be a larger number of xenocrysts in the new separates. Inclusions in the xenocrysts may be enriched in radiogenic/nucleogenic isotopes compared with phenocrysts containing noble gases in the source magma. This work revealed that contribution of radiogenic/nucleogenic components accumulated in crystals with an old age (350 Ma of the Udachnaya kimberlite, Maas et al., 2005) becomes significant even if the crushing extraction method is applied.

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## Evaluation of parent body processes on presolar components in chondrites

Tetsuya Yokoyama<sup>1</sup>, Conel Alexander<sup>2\*</sup>, Richard Walker<sup>3</sup>

<sup>1</sup>Tokyo Institute of Technology, <sup>2</sup>Carnegie Institution, DTM, <sup>3</sup>University of Maryland

Chondrites and differentiated meteorites at the bulk meteorite scale commonly show small but resolvable mass-independent isotopic deviations from the terrestrial values in refractory elements (e.g., Cr, Ti, Mo, Ru, Ba, Sm). At least some of these anomalies are nucleosynthetic in origin, and have been interpreted to reflect incomplete mixing of isotopically diverse presolar materials in the protosolar nebula, although uniform, terrestrial isotopic compositions in bulk chondrites have been reported for some elements (e.g. Sr, Os). Processes that occur on parent bodies such as thermal metamorphism and aqueous alteration can also potentially affect the isotopic compositions of bulk chondrites. Isotopic investigation of acid residues from primitive chondrites may shed light on this problem. The residues are rich in a variety of isotopically anomalous presolar grains which can control the isotopic composition of bulk chondrites. In this study, we analyzed Os isotope compositions in acid residues from four CMs (MET 01070, ALH 83100, Murchison, QUE 97990) and three CRs (GRO 95577, EET 92042, GRA 95229).

All the CM/CR-residues are characterized by positive epsilon-Os (eOs) values that are resolvable from the solar (= terrestrial) component, suggesting the enrichment of Os isotopes produced by the s-process. There are significant variations in eOs values across CM-residues. The magnitude of the positive Os isotope anomalies in the residue of the ALH 83100 (CM1/2) (e188Os = +3.10) is nearly twice as large as those present in residues from Murchison (CM2) (e188Os = +1.66). The residue of MET 01070 (CM1) has eOs values (e188Os = +2.31) larger than those of Murchison, whereas that from QUE 97990 (CM2.6) has the smallest deviation in eOs values from zero among all CM chondrites (e188Os = +0.95). Variations in the magnitude of the eOs values are also observed in residues from CR chondrites, where the residue from GRO 95577 (CR1) showed the largest positive Os isotope anomalies (e188Os +2.64). We propose that the observed isotopic variation was caused by the destruction of presolar phases via progressive aqueous alteration on the CM/CR parent bodies, rather than any nebular processes creating heterogeneous distributions of presolar grains that predate the planetesimal formation. Some petrologic observations of CM chondrites point to the occurrence of aqueous alteration prior to parent body accretion. However, destruction of presolar phases during preaccretionary alteration would result in the modification of Os isotopic compositions at the bulk meteorite scale, which is evidently not the case. The enrichment of s-process Os isotopes in the residues from highly altered CM/CR chondrites implies that aqueous alteration on the parent body preferentially destroyed the r-process-Os carrier(s) and modified it into an acid leachable phase in the chondrite matrices, while acid resistant s-process-rich grains (SiC, graphite) survived. This process, however, did not disturb the Os isotopic composition at bulk meteorite scale. Even though bulk chondrites are isotopically homogeneous in Os, our new results suggest that parent body aqueous/metamorphic processes have acted on the distribution of Os among presolar phases. This suggests that isotopic anomalies reported for some other elements that have been interpreted as reflecting nebular heterogeneities, may partly or wholly be the result of parent body processing. Presolar phases that are enriched in certain nucleosynthetic components could release these components upon destruction of the hosting phases. If the element is more highly soluble than Os, fluid transport could lead to the formation of isotopically modified bulk samples whose isotopic compositions are not representative of the whole parent body. Thus, isotopic heterogeneities among bulk chondrites should not automatically be interpreted as evidence for nebular heterogeneity.

Keywords: chondrite, presolar grain, aqueous alteration, osmium isotope

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## Fractal distribution of mercury inclusions in quartz by in situ X-ray computed tomography (CT) system

Tomo Shibata<sup>1\*</sup>, Teruyuki Maruoka<sup>2</sup>, Takuya Echigo<sup>3</sup>

<sup>1</sup>Geological Survey of Hokkaido, HRO, <sup>2</sup>Univ. Tsukuba, <sup>3</sup>JIRCAS

Large amounts of data are obtained by contemporary analytical instruments and observation systems. Mathematical and statistical analysis of the obtained data serves for the states or origin of objective matter. As X-ray computed tomography (CT) analysis has been developed in medical imaging method, the CT analysis for terrestrial materials yields a new approach in resolving their internal structure and origin (e.g., Tsuchiyama et al, 2009). We here investigate in situ X-ray CT analysis of mercury inclusions in quartz, and discuss relationship between the inclusions and the quartz using mathematical and statistical analysis.

We used quartz crystal (2 cm x 5 cm x 5 cm) from San Benito, California. This crystal contains visible mercury inclusions of their size, 1-2 mm, and this crystal is a suitable sample for the CT analysis because great density difference between the inclusions and the crystal. We used a microfocus X-ray CT system (Shimazu Corp., SMX-225CT), and obtained 260 CT images of 2-D slices over rotating a sample. The obtained images were processed using an image processing software.

These 2-D binary images were utilized for fractal and multifractal analysis using box-counting method. Obtained fractal dimension ( $D$ ) is 1.76, suggesting that the distribution of mercury inclusions in quartz indicate planate form ( $D=1$ , linear form;  $D=2$ , planate form). Based on multifractal analysis, fractal structure could not be uniform because its singularity is widely. As the fractal structure of mercury inclusion are formed by random walk of mercury particles in Euclid spatial, the mercury inclusion would coincide with quartz growth or flow without constrained situation. Preliminary results from in situ X-ray computed tomography (CT) analysis also yield to recognize quantitative and spatial information of the inclusions in the quartz using fractal and multifractal analysis.

### Reference

Tsuchiyama et al., *Meteoritics and Planetary Science*, 44, 1203-1224, 2009.

Keywords: fractal, random walk, mathematical and statistical analysis, X-ray computed tomography system, mercury inclusions, in situ observation

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## Development of tin isotopic analysis for cassiterite and its archaeological application

Erika Yamazaki<sup>1\*</sup>, Shun'ichi Nakai<sup>2</sup>

<sup>1</sup>Tokyo Institute of Technology, <sup>2</sup>ERI, Tokyo University

[Introduction] Bronze products are alloys composed of Cu, Sn, and Pb and they were used before iron products had become common. For provenance studies of bronze materials, Pb isotopic composition has been utilized (Brill and Wampler, 1965). However, this method includes a problem that if raw materials from different localities are mixed, Pb isotopic composition of bronze can not indicate their origins. Additionally, some ancient bronze products seem to have experienced recycling, and mixing could occur during recycling. So Pb isotopes are not always helpful to determine provenances of bronze products.

Budd et al., (1995a) proposed that Sn isotopes are useful to detect recycling of bronze products, since metallurgical processes could cause measurable Sn isotope fractionation because Sn is volatile and has the largest number of isotopes among all elements. Although Cu, other main component of bronze could show isotopic fractionation, no significant isotopic fractionation has been detected for Cu (Mathur et al., 2009). Moreover, it was concluded that Pb isotopic fractionation during metallurgical process was smaller compared with isotopic variation among each locality (Barnes et al., 1978; Budd et al., 1995b). So Sn isotope is expected to give us an aid in the provenances studies of bronze.

Tin isotope analysis for ancient bronze products was carried out by Gale (1997). This research showed that there was no significant isotopic difference between the analyzed bronze products and concluded that these samples did not experience recycling with metallurgical processes. However, variation of Sn isotopic composition was detected in preliminary investigation for bronze products discovered from China (Nakai, unpublished).

In order to evaluate recycling by Sn isotopes, we need an assumption that raw materials of tin ore (cassiterite) have uniform Sn isotopic composition regardless of their origins. Haustein et al. (2010) reported a series of Sn isotopic data for cassiterite and observed the largest significant variation of the isotopic composition (about 0.6 permil).

[Objective] Although variations of Sn isotopic composition have been detected in nature, there are few studies for cassiterite. In addition, analytical methods are not standardized. So we have established the method of Sn isotopic analysis for cassiterite and applied to cassiterite from Japan. These cassiterite samples are provided by Dr. Ishihara at National Institute of Advanced Industrial Science and Technology (AIST).

[Experiment] Cassiterite was crushed in a stainless mortar and powdered in an agate mortar. About 1 mg of powdered sample was weighed into PFA vials. 0.3 ml of hydroiodic acid was added and heated overnight by using teflonbomb at 100°C. Subsequently, samples were dried at 80°C and leached in hydrochloric acid. The procedures were repeated until the sample is dissolved. Moreover, hydrofluoric acid and perchloric acid were added if some residue remained. We used an extraction chromatography system to separate Sn from other elements (Yi et al., 1995) and purified tin solutions were diluted in 2% nitric acid including trace of hydrofluoric acid. Sb was added for external fractionation correction. The Sn solutions were analyzed by a MC-ICP-MS (the Micromass Isoprobe). Sn isotopic ratios of samples were evaluated as the deviation from Sn isotopic ratios of standard.

[Result] As a result, we also observed significant variations of Sn isotopic composition for cassiterite samples. We detected up to 0.16 permil per mass for Japanese cassiterite and each sample was followed mass dependent isotopic fractionation. We detected Sn isotopic variations as large as 0.7 permil per mass in ancient bronzes from China, which is larger than the variation in cassiterite samples. So the results of this study suggest a possibility that Sn isotopes could be used to detect recycling.

Keywords: tin, stable isotope, cassiterite, ICP-MS, archaeology