Subduction factory and its impact on global mantle heterogeneity

H. IWAMORI[1,2], A. IKEMOTO[3], H. NAKAMURA[1,2], M. YOSHIDA[4], R. YANAGI[2]

[1]Dept. Solid Earth Geochemistry, JAMSTEC, Japan
[2]Dept. Earth Planet. Sci., TITECH, Japan
[3]Shinsei Bank, Limited, Japan
[4]Dept. Deep Earth Struct. and Dyn. Res., JAMSTEC, Japan

"Subduction factory" produces arc magmas as a main output, which has been extensively studied to probe the sub-arc structure and processes. Another equally important product is the residual material that subducts deeper into the mantle; e.g., dehydrated or melted slab and mantle materials. Importance of such processed materials has been repeatedly argued for geochemical heterogeneity and global material cycling in the mantle [e.g., 1-3]. In spite of its importance, compared to the arc magmas, the residual materials have been less constrained partly due to its inaccessibility. Of several approaches for quantitatively identifying such processes and materials, based on the fluid dynamical model for trace element transport in subduction zones [4], as well as 3-D mantle convection model that incorporates water transport [5,6], we discuss what comes out and what goes down to the deep mantle, and how the subducted materials may contribute to global geochemical structures, including east-west mantle geochemical hemispheres [7].

Within the subduction factory, melting and melt extraction create compositionally zoned mantle, which is dragged down by corner flow to encounter the fluid from the slab [4]. It has been found that this combined process creates highly complicated 2-D distribution of elements, as well as large variability in parent/daughter ratio of radionuclides, which are comparable to the global variability. Outside the subduction factory, the heterogeneous materials are transported and redistributed by both solid convective flow and percolating fluid that is released upon dehydration of hydrous minerals (including nominally anhydrous minerals) at various depths over the whole mantle, resulting in effective delivery of water (and associated elements) to a broad but a certain region of the mantle.

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Recycled and primordial noble gas components in the upper mantle

RITA PARAI[1] AND COLIN R. M. JACKSON[2]

[1] Department of Terrestrial Magnetism, Carnegie Institution of Washington, Washington, DC; USA
rparai@carnegiescience.edu
[2] Geophysical Laboratory, Carnegie Institution of Washington, Washington, DC, USA

Exchange of volatiles between the deep Earth and surface reservoirs occurs in association with plate tectonics: mantle volatiles are outgassed via magmatism, and ingassed via the downgoing plate at subduction zones. Accordingly, noble gases in the mid-ocean ridge basalt (MORB) mantle source reflect mixing between primordial volatiles from Earth's accretion, radiogenic and fissiogenic components, and recycled surface volatiles.

Kr and Xe isotopes in continental well gases and basalts constrain the proportion of each gas derived from recycling in the MORB source, given a chondritic primordial endmember composition [1,2]. Similar determinations for Ne and Ar, however, are inhibited by uncertainties regarding the primordial Ne and Ar endmember compositions: the MORB source ^{20}Ne/^{22}Ne ratio is ~12.5 [3], which indicates either extensive or negligible recycling of atmospheric Ne (^{20}Ne/^{22}Ne of 9.8), depending on whether MORB source primordial Ne is related the solar nebula (^{20}Ne/^{22}Ne of 13.4; [4]) or later input (Ne-B, which is thought to reflect solar wind implantation and sputtering effects in late-accreting planetary material, ^{20}Ne/^{22}Ne of 12.5-12.8; [5]).

Here we examine noble gas mixing systematics between subducting lithologies and primordial components. We find that in order to explain MORB source Ne given solar primordial Ne, recycled material must have high Ne/(Ar,Xe) and must experience minimal elemental fractionation during subduction. A high Ne/(Ar,Xe) component is observed in altered oceanic crust [6,7], but preservation of this component is at odds with evidence for light noble gas enrichment in arc and back-arc lavas [8] (and complementary light noble gas depletion in the downgoing slab). Thus, mixing systematics favor Ne-B as the primordial MORB source Ne component, supporting delivery of primordial Ne by accreting planetary material rather than solar nebular ingassing. Given the Ne-rich nature of the solar nebula, its contribution to other MORB source volatiles should also be small. [1] Holland et al., 2008, *Science* [2] Parai and Mukhopadhyay, 2015, *G³* [3] Holland and Ballentine, 2005, Nature [4] Heber et al., 2012, ApJ [5] Moreira, 2013, *Geochem Pers*) [6] Kumagai et al., 2004, *G³* [7] Kendrick et al., 2015, *CMP* [8] Hopp and Ionov, 2011, *EPSL*.

U-Th-Pb cycling from ocean to mantle and the HIMU endmember source

T. PETTKE[1], J. KODOLÁNYI[1,2] AND B. S. KAMBER[3]

[1] University of Bern, Institute of Geological Sciences, CH-3012 Bern, Switzerland. pettke@geo.unibe.ch
[2] Max Planck Institute for Chemistry, 55128 Mainz, Germany. janos.kodolanyi@gmail.com
[3] Department of Geology, Trinity College Dublin, Dublin 2, Ireland. kamberbs@tcd.ie

Of the isotopically distinctive mantle domains, the so-called HIMU source is the most extreme and its genesis continues to be debated. Here we report extreme U enrichment at unchanged Th concentrations in oceanic serpentinites, representing altered depleted peridotite. The U-enrichment from seawater has resulted in $^{206}Pb/^{204}Pb$ as high as 56, spectacular for samples drilled from a modern passive margin. By contrast, $^{208}Pb/^{204}Pb$ has remained unchanged. Similar but less extreme features are found in 1.9 billion years old altered oceanic crust (AOC).

These U/Th/Pb systematics are relevant to models of the HIMU source. Forward modelling demonstrates that convecting mantle, metasomatised by deep fluids derived from subducted AOC and serpentinites evolves to the HIMU Pb isotope signature, while satisfying experimental and empirical constraints on subduction zone element processing. Over time, the net U addition from subducted serpentinite to convecting mantle has become expressed as the second terrestrial Pb paradox. By contrast, the traditional genetic model – recycled oceanic crust residues as the HIMU source - can only satisfy uranogenic Pb isotope systematics but is irreconcilable with observed $^{208}Pb/^{204}Pb$.

Arc fluxes and the composition of the recycled ocean crust reservoir

CHARLES H LANGMUIR[1*] AND STEPHEN J TURNER[2],

[1]Harvard University, 20 Oxford Street Cambridge, MA, USA 02138 (*correspondence: langmuir@eps.harvard.edu)
[3]Oxford Universiry S Parks Rd, Oxford, UK OX1 3AN

Recycled ocean crust is often proposed as an enriched reservoir in the mantle. Here we propose that it is instead the fertile, depleted reservoir that is commonly sampled at ocean ridges, not as "enriched pyroxenite veins" but as a major depleted component. Recent experimental data for fluxes from subducting ocean crust [1,2], as well as models of global arc volcanism [3], require that contributions from slab melts are ubiquitous at convergent margins. Slabs always melt. Since this is true in the present Earth, it would also have been true in the past when Earth's thermal environment was hotter. The extent of melting and mass of melt can be estimated by what is required to account for the chemical compositions of arc volcanics. Sr is particularly valuable in this regard because it is not particularly enriched in sediments, but nonetheless strongly enriched in convergent margin magmas, requiring a large flux from the subducting ocean crust. Using experimental data it is then possible to make estimates of the amounts of more incompatible elements that are being released from the ocean crust portion of the slab. The quantities are substantial, leading to a recycled ocean crust reservoir that is even more depleted than the average MORB entering subduction zones. This recycled reservoir is then entirely inappropriate as an enriched reservoir, because it is strongly depleted in highly incompatible elements. It nonetheless remains fertile in mineralogy and major elements. These are the essential requirements for the depleted MORB mantle. Enriched reservoirs then can never be the direct result of recycled ocean crust. Only small degree melts from such recycled crust, or other forms of near surface metasomatism by low degree melts, are the exclusive source apart from recycled sediments of enriched reservoirs.

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Intermediate arc magmatism and crustal evolution: The view from big(ger) data.

ADAM J.R. KENT[1]

[1]CEOAS, Oregon State University, Corvallis, OR, 97330, USA, adam.kent@geo.oregonstate.edu

Convergent margin systems are characterized by uniquely high production rates of intermediate and silicic magmas. Compositional characteristics of these magmas are shared by bulk average continental crust, and thus there has been a longstanding interest in the role of convergent magma systems in crustal production and evolution. Despite this little clear consensus exists about the dominant processes that control formation of intermediate magmas in arc settings. Key questions include the roles of fractionation vs. mixing; the relationships between magmatic and liquid compositions; and the interplay between plutonic and volcanic processes.

This presentation will summarize recent results regarding the origin of intermediate magmas within subduction zones, and the relationship between magmatism and crustal growth, focussing on large global petrologic data sets as a source of information. Extensive data for rock, melt inclusion, and mineral compositions are widely available. Coupled with increasing sophistication of approaches to interpretation, these data sets provide important insights into the origin of intermediate magmas in convergent margins, and the role of magmatism in crustal evolution. However differences of opinion still persist; for example, some data sets indicate the importance of high level mixing processes in the generation of intermediate magmas, whereas others are interpreted to reflect the importance of fractionation. Despite these on going issues the application of big(ger) data provides important means for identifying important processes and testing models of convergent margin processes and crustal growth.

Water, *f*O_2, and the creation of continental crust signatures in Aleutian arc magmas

K. A. KELLEY[1]*, E. COTTRELL[2], M. COOMBS[3], AND E. R. GRANT[1,2]

[1]GSO, Univ. of Rhode Island, Narragansett, RI 02882 USA (*correspondence: kelley@uri.edu)
[2]NMNH, Smithsonian Inst., Washington, DC 20560, USA
[3]AVO, US Geological Survey, Achorage, AK 99508, USA

Early Fe depletion of arc magmas (i.e., calc-alkaline affinity) drives them towards the composition of bulk continental crust. Arc magmas may develop early Fe depletion due to high magmatic H_2O, which suppresses silicates, high oxygen fugacity (*f*O_2), which promotes early Fe-oxide crystallization, or primary Fe-poor magmas may emerge via slab melting and mix with Fe-rich basaltic magmas, causing an apparent Fe-depletion trend that is unrelated to differentiation of a single parent magma. Yet, the relative importance of these key factors in generating calc-alkaline trends in natural arc magmas remains under-constrained.

In the oceanic Aleutian island arc, magmas become strongly calc-alkaline towards the west, where convergence is highly oblique. This unique setting provides an ideal locale to test the effects of H_2O, *f*O_2, and parental magma composition on the development of Fe-depletion in arc magmas. Here, we present new measurements of western Aleutian whole-rock lavas paired with $Fe^{3+}/\sum Fe$ ratios (a proxy for magmatic *f*O_2; acquired by XANES), dissolved volatile contents, and major elements of olivine-hosted melt inclusions from western and central Alaska-Aleutian arc volcanoes. The Tholeiitic Index (THI; [1]) quantifies Fe depletion in the magma series at each volcano (<1 is Fe-depleted, >1 is Fe-enriched). Over a range of THI, from 0.9-0.65, the $Fe^{3+}/\sum Fe$ ratios of Aleutian basalts, referenced to 6 wt.% MgO (i.e., $Fe^{3+}/\sum Fe_{6.0}$) range from 0.22-0.31 and correlate strongly with THI, such that more Fe-depleted magmas contain proportionally more oxidized Fe. Maximum dissolved H_2O contents of these volcanoes also negatively correlate with THI and with $Fe^{3+}/\sum Fe_{6.0}$ ratios, suggesting Fe-depletion originates by differentiation rather than by mixing. The effects of H_2O and *f*O_2 on arc magmatic differentiation are challenging to isolate in nature, but experimental data suggest that *f*O_2 exhibits stronger control than H_2O on the relative appearance of magnetite vs. silicates on the liquidus, thus exerting greater leverage on the THI.

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Molybdenum isotope evidence for melting of subducted sediments beneath the Lesser Antilles arc

HEYE FREYMUTH[1,2]*, MATTHIJS VAN SOEST[3], SUSANNE SKORA[4], TIM ELLIOTT[1]

[1]Bristol Isotope Group, University of Bristol, Queen's Road, Bristol BS8 1RJ, UK (*correspondence: heye.freymuth@manchester.ac.uk)
[2]School of Earth, Atmospheric and Environmental *Science,* University of Manchester, Oxford Road, Manchester, M13 9PL, UK
[3]School of Earth and Space Exploration, Arizona State University, 781 E. Terrace Mall, Tempe, AZ 85287-6004, USA
[4]Institute of Geochemistry and Petrology, Earth Sciences, ETH Zurich, Clausiusstrasse 25NW, 8092 Zurich, Switzerland

Molybdenum isotope ratios in arc magmas have recently been shown to trace fluid and melt components derived from the subducted slab. [1]. Here we present Mo isotope data for Lesser Antilles arc lavas and sediments subducting at the Lesser Antilles trench. Radiogenic isotope characteristics in the Lesser Antilles lavas indicate the addition of enriched, continental material to the magmas. It is hotly debated whether this continental material is a sediment derived from the subducted Atlantic plate or if the magmas assimilated material from the overlying plate while ascending to the surface [e.g. 2,3].

Sediments subducting at the Lesser Antilles trench contain sequences of black shales that have unusually high $\delta^{98/95}$Mo (the per mil deviation in ^{98}Mo/^{95}Mo relative to the NIST 3134 standard) of ~0.6. The black shales are highly enriched in Mo and despite being a volumetrically minor component in the sediment column they dominate the Mo isotope composition of the bulk sediment subducting at the Lesser Antilles which has $\delta^{98/95}$Mo = 0.55, about 0.8 ‰ higher than the sediment subducing at the Mariana trench in the Pacific.

Lavas from the depleted, northern Lesser Antilles islands have $\delta^{98/95}$Mo = -0.1 – 0.05 and are within the range of previously published data from the Mariana arc [1] whereas lavas from the more enriched, southern Lesser Antilles islands have substantially higher higher $\delta^{98/95}$Mo = 0.15-0.35. We attribute the high $\delta^{98/95}$Mo in the southern Lesser Antilles islands to black shale-derived Mo and show that these lavas require the addition of sediment-dominated slab melts to their mantle sources.

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Constraints from Arc Magmas on the Pb Isotope Composition of the Subducted Igneous Oceanic Crust

SUSANNE M. STRAUB[1]*

[1]Lamont Doherty Earth Observatory, Palisades NY, U.S.A. (*correspondence: smstraub@ldeo.columbia.edu)

Arc magmas have typically low ratios of Ce/Pb<10 that are considered to reflect the addition of Pb-rich components from subducted slab. Quantification shows that the slab flux contributes ca. 80-90% of arc Pb while the mantle wedge contributrion are negligible [1]. Consequently, the slab flux must control the Pb isotope composition of arc magmas, implying that the typical linear arc arrays in Pb isotope space reflect binary mixing between ‘continental’ (sediment/eroded continental crust) and ‘mantle-type’ (crust formed at midocean ridges and intraplate volcanoes) components from the subducted slab. Comparative studies of Pb isotope compositions of arc magmas and sediments and/or eroded crust from the conjugate trenches confirmed strong links to recycled continental material, but the connection between the arc chemistry and the subducted igneous crust is less well explored. However, given its large volume, the depleted subducted igneous crust should provide a Pb flux that rivals the Pb flux of the enriched continental components, and that should transmit the Pb isotope heterogeneity of the oceanic crust (e.g. Pacific vs Indian domains, or MORB vs. intraplate domains) to arc magmas.

This hypothesis is being tested by a systematic evaluation of arc magmas composition using combined Pb isotope and trace element systematics. This allows for isolating the Pb isotope signals of the igneous oceanic crust from the signals of the recycled continental crust and the subarc mantle. First results confirm that Pb isotope diversity of the igenous oceanic crust can be traced (e.g Indian vs. Pacific MORB, intraplate crust vs. MORB-type crust). Remarkably, the arc Pb isotope systematics seem to require a normal MORB-type crust that is on average less radiogenic in Pb (^{206}Pb/^{204}Pb ~18.1) than the average global MORB based on surface samples (^{206}Pb/^{204}Pb ~18.4, [2]). This discrepancy may either reflect temporal variations in the Pb isotope composition of MORB, or possibly a systematic difference in the MORB average obtained from the top of the extrusive layer, and the overall Pb flux to arcs that should integrate Pb from the entire subducted MORB-type crust.

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Noble gas and halogen variation of volcanic rocks from the Izu-Mariana subduction zone

H. SUMINO[1]*, M. KOBAYASHI[2], R. BURGESS[3], L. JEPSON[3], S. MACHIDA[4], A. SHIMIZU[5], D. HAHM[6,7],
D.R. HILTON[6] AND C.J. BALLENTINE[8]

[1] Dept. Basic Sci., Univ. Tokyo, Tokyo 153-0041, Japan (*correspondence: sumino@igcl.c.u-tokyo.ac.jp)
[2] GCRC, Univ. Tokyo, Tokyo 113-0033, Japan
[3] SEAES, Univ. Manchester, Manchester M13 9PL, UK
[4] JAMSTEC, Kanagawa 237-0061, Japan
[5] TIRI, Tokyo 135-0064, Japan
[6] Scripps Inst. Ocean., UCSD, California 92093-0244, USA
[7] Dept. Ocean., Pusan National Univ., Busan 46241, Korea
[8] Dept. Earth Sci., Univ. Oxford, Oxford OX1 3AN, UK

The subduction of sedimentary-pore-fluid-like noble gases and halogens is supported by several studies. These include noble gases and halogens with seawater and sedimentary pore-fluid signatures in exhumed mantle wedge peridotites from the Sanbagawa-metamorphic belt, southwest Japan [1], mantle-derived xenoliths from Kamchatka and Luzon arcs [2], and in seafloor and forearc serpentinites [3] along with seawater-like heavy noble gases in the convecting mantle [4]. Here we present noble gas and halogen compositions of olivines in arc lavas of the Izu-Mariana subduction zone to develop a better understanding of the processes that control the return of these volatile and highly incompatible elements into the mantle.

The MORB-like $^{3}He/^{4}He$ of most samples are consistent with those of the subduction zone mantle xenoliths [2], which indicate a considerably low contribution of radiogenic ^{4}He in the subduction fluids observed in the Sanbagawa samples [1]. In contrast, the $^{40}Ar/^{36}Ar$ indicate significant involvement of atmospheric Ar in the magma source. Systematically higher atmospheric contribution in the volcanic front compared to the rear arc in the Izu arc suggests that subduction of seawater-derived Ar has a significant effect on the noble gas composition at the magma-generation region. Although the halogen compositions of most samples are close to that of MORB-source mantle, some rear-arc samples show a significant contribution from pore-fluid-derived halogens. The results suggest that halogen-poor fluid may be dominantly released from the subducting slab beneath the arc, while halogen supply from the slab is limited beneath some volcanoes in the rear arc. This implies the relative persistence of halogens in the subducting slab compared to noble gases.

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Hydrolysis of fluorine-rich titanium complex in hydrothermal fluids: implications on mobility and fractionation of the high-field-strength-element in subduction zones

XING DING[1*], JUNJIE HE[2], WEIDONG SUN[2]

[1]State Key Laboratory of Isotope Geochemistry, Chinese Academy of Sciences, Guangzhou, P. R. China, xding@gig.ac.cn (* presenting author)
[2]Key Laboraotry of Mineralogy and Metallogeny, Chinese Academy of Sciences, Guangzhou, P. R. China, jjhe_kid@163.com; weidongsun@gig.ac.cn

It is generally accepted that solubility of rutile, a major host mineral for titanium, governs the activity and cycling of high-field-strength element (HFSE) in subduction zone [1-4]. Because of extremely high solubility of rutile in fluoride-bearing fluids [5], HFSE mobility in fluorine-rich fluids is generally thought to be high [6-7]. However, geochemical behaviors of fluorine-rich HFSE complexes are not clear yet, as metal elements are thought to move in hydrothermal fluids in the form of complex.

A series of hydrolysis experiments under the conditions of 200-800°C and 0.1-2.5 GPa were conduct using synthetic potassium fluotitanate, K_2TiF_6. In the experimental conditions the Ti complex hydrolyze gradually to form different complex species through ion exchange between F^- in the fluorides and OH^- in the H_2O, and to produce TiO_2 deposit in the acid fluids. Our experimental results show that, i) the cumulative hydrolysis ratio of Ti fluoride complex, which represents the amount of deposit, is enhanced with the pressure and especially the temperature increasing and the initial concentration decreasing. Ti complex hydrolysis is not good to HFSE mobility in fluids; and ii) the deposit changes from anatase phase in the low temperature and pressure to rutile phase in the high temperature and pressure. Ti fluoride complex hydrolysis in fluids can also form rutile due to the change of enviromental conditions (eg. increasing P, T or pH).

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Geochemical behaviour of Mo and porphyry Mo mineralization

W SUN[1,2], C LI[1], M LING[1], T IRELAND[2], W FAN[2]

[1] Guangzhou Institute of Geochemistry;
[2] CAS Center for Excellence in Tibetan Plateau Earth Sciences, The Chinese Academy of Sciences, China;
[3] The Australian National University, Bldg 61 Mills Road Acton ACT, Canberra, ACT, 0200

Molybdenum is very rare in the silicate Earth, with abundances of 50 ppb in the primitive mantle, and 0.8 ppm in the continental crust, yet it forms “pure” Mo porphyry deposits. Given that Mo is moderated incompatible, it requires pre-enrichment to form porphyry Mo deposits, e.g., through the oxidation-reduction cycle. Oceanic Anoxic Events are the most efficient and important process that enriches Mo. Large amounts of Mo-enriched sediments formed on the Pacific Ocean floor during at least 9 major OAEs since the Late Jurassic. The Eastern Pacific plate has been mostly subducted, with only Cenozoic crust left. During plate subduction, Mo-rich sediments may be taken down to the mantle wedge through subduction erosion, which were metamorphosed and then transferred to porphyry deposits through partial melting.

Molybdenum deposits can be classified into three types: porphyry-Cu-Mo, high-F porphyry and low-F porphyry-Mo deposits. Porphyry-Cu-Mo deposits are usually formed through partial melting of subducted oceanic crust and metamorphosed Mo-rich sedimentary material due to subduction erosion. The high-F porphyry-Mo deposits were likely formed by partial melting of metamorphosed Mo-enriched sediments during slab rollback. Low-F porphyry-Mo deposits were formed through direct partial melting of metamorphosed Mo-enriched sediments entrained into the mantle wedge through subduction erosion (usually associated with arc granite). Porphyry-Cu (Au) deposits in the southwest Pacific margin are all associated with subduction of backarc basins younger than the last Oceanic Anoxic Event. Therefore, porphyry Cu (Au) deposits in the southwest Pacific margin have no economic levels of Mo. The oxygen fugacity of northwest Pacific is lower than ΔFMQ +2, which inhibits the formation of porphyry deposits.

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