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Os isotope systematics of mesoarchean chromitite-PGE deposits in the Singhbhum Craton (India): Implications for the evolution of lithospheric mantle

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Abstract

Os isotope studies of Mesoarchean (3.2 Ga) chromitite and associated gabbro–breccia hosted platinum-group element (PGE) deposits of the Singhbhum Craton in eastern India provide evidence for a complex history involving source mantle heterogeneity and hydrothermal processes. The average initial $^{187}Os/^{188}Os$ compositions of unaltered chromites of massive chromitite layers from two mining districts are 0.1031 +/− 0.0007 (Nuasahi) and 0.1029 +/− 0.0005 (Sukinda), with negative γOs values of − 1.78 +/− 0.65 and −2.04 +/− 0.43, respectively. Mantle extraction ages (T_{MA}) for these chromites indicate depletion from primitive mantle beginning as early as ~3.7 Ga, some 400 Ma before the crystallization of the respective chromites. The subchondritic signatures of Os in the studied chromites are comparable with results of age-constrained chromites from ultramafic intrusions in the Zimbabwe Craton [Nägler, T.F., Kramers, J.D., Kamber, B.S., Frei, R., Prendergast, M.D.A., 1997. Growth of subcontinental lithospheric mantle beneath Zimbabwe started at or before 3.8 Ga: Re–Os study on chromites. Geology 25, 983–986.], interpreted to reflect sequential extraction from evolving Archean subcontinental lithospheric mantle (SCLM). The similarity of depleted Os isotopic signatures of chromites from the Singhbhum Craton with those from the Zimbabwean occurrences, and their compatibility with the proposed Kaapvaal Craton SCLM model, leads us to propose that SCLM beneath the Singhbhum Craton also started to form in the early Archean. Up to now, early Archean continental crustal fragments, expected to have formed with the SCLM in the Singhbhum Craton have not been found, and early Archean detrital zircons [Mishra, S., Deomurari, M.P., Wiedenbeck, M., Goswami, J.N., Ray, S., Saha, A.K., 1999. $^{207}Pb/^{206}Pb$ zircon ages and the evolution of the Singhbhum Craton, eastern India: an ion microprobe study. Precambrian Res. 93, 139–151.] remain the only evidence for their existence. An alternative possibility is that the early Archean depleted mantle region could have remained within the convecting mantle for a few hundred million years before being added to the SCLM beneath the Singhbhum Craton. Re–Os isotopic data of chromites of massive chromitites from PGE and base metal sulfide (BMS) mineralized breccia zones are extremely heterogeneous. Perturbations of the Re–Os isotope systematics in these chromites are best explained as having resulted from interaction with hydrothermal fluids percolating in the shear zones,

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possibly genetically related to the emplacement of 3.1 Ga gabbroic suites. These fluids played a major role for the genesis and modification of the PGE mineralization.

Keywords: Archean chromite; Chromite; Re–Os isotope; Sm–Nd isotope; SCLM; Singhbhum Craton; India

1. Introduction

The study of volcanic and plutonic rocks of greenstone belts provides great insight into understanding the formation and stabilization of cratons and the evolution of Earth's crust and mantle. Sill-like ultramafic bodies related to komatiitic parental magmas within the Archean greenstone belts are significant sources for chromite resources such as those found in the Archean Shurugwi ultramafic complex of the Zimbabwe Craton in southern Africa (e.g. Stowe, 1987, 1994; Rollinson, 1997). Similar chromite deposits are also found within Archean greenstone belts in the Indian shield. Examples include those of the Singhbhum Craton in eastern India, and in the Dharwar Craton in southern India (Fig. 1A) (e.g. Srinivasan and Naqvi, 1990; Mondal et al., 2006a). In the Singhbhum Craton the layered ultramafic bodies occur within low-grade metamorphic rocks of the Iron Ore Group (IOG) mainly in the Nuasahi and Sukinda areas of Orissa state and in the Jojohatu area of Bihar state (Fig. 1B). The chromite deposits of the Nuasahi and Sukinda areas are thought to have formed from a bonitic magma (Fig. 3), which is similar to the compositions of spatially associated chromite-bearing high-Mg metaigneous rocks of the IOG (Mondal et al., 2001, 2006a). It has been suggested that a combination of magma-mixing involving a boninitic magma, coupled with relatively high water contents of the mixed magma in a supra-subduction zone setting were responsible for the chromites in the Nuasahi and Sukinda massifs (Mondal et al., 2006a). In many places these suites of rocks are closely associated with gabbroic intrusions (Figs. 1 and 2). In the Nuasahi and Sukinda areas, in addition to massive chromitite bodies, the deformed ultramafic rocks are associated with breccia-hosted 'discordant-type' PGE-deposits, which are developed in shear zones (Fig. 2) (Mondal and Baidya, 1997; Mondal, 2000). Platinum group element mineralization of this type is broadly described as 'magmatic-hydrothermal' (Mondal et al., 2001; Augé et al., 2002) and the breccia-hosted ores are similar to that of the Robie Zone of the Lac des Iles complex in Canada (e.g. Watkinson et al., 2002) or the Mayville Intrusion of the Bird River Belt (e.g. Peck et al., 2002).

Here we present a study of Re–Os isotopic systematics of a suite of 19 chromite separates from the massive chromitites of the Nuasahi and Sukinda massifs in the Singhbhum Craton. The main aim of this study is to assess the source characters of the chromite and PGE deposits, and to relate the Re–Os characteristics to other worldwide chromite and PGE bearing ultramafic suites. In conjunction with previous petrological studies (Mondal et al., 2001, 2006a) we are able to correlate primary magmatic signatures with perturbations related to later open system process in the PGE deposits. The ^{187}Re/^{188}Os isotope system (where the decay constant, α, for $^{187}\mathrm{Re}=1.666\times10^{-11}$ year^{-1} Smoliar et al., 1996) is particularly useful for discriminating between long-term melt-depleted reservoirs, including subcontinental lithospheric mantle (Walker et al., 1989). The observations from the mantle xenoliths confirm that the SCLM is geochemically distinct from the convective mantle and has remained stable as residue of extensive partial melt extraction in the Archean (e.g. Boyd, 1989; Bernstein et al., 1998; Herzberg, 2004). In this regard we discuss the evolution of the early Earth's mantle regions through time with respect to Re–Os isotopic systematics. Because of the potential power of combining the siderophile Re–Os system with the lithophile Sm–Nd system to characterize mantle depletion/enrichment, we have conducted Sm–Nd isotopic analyses of four whole rock samples of chromite-bearing high-Mg metaigneous rocks of the IOG supracrustal suite from the Nuasahi area.

2. Geological background

2.1. The Singhbhum Craton

The eastern part of the Indian Shield is composed of a high-grade metamorphic terrain known as the Chhotanagpur Craton to the north and a granite–greenstone terrain known as the Singhbhum Craton to the south (Saha, 1994). The latter is principally composed of several granitoid batholiths, which are largely surrounded as well as intervened by supracrustal rocks (Fig. 1B). The Older Metamorphic Group (OMG) is the

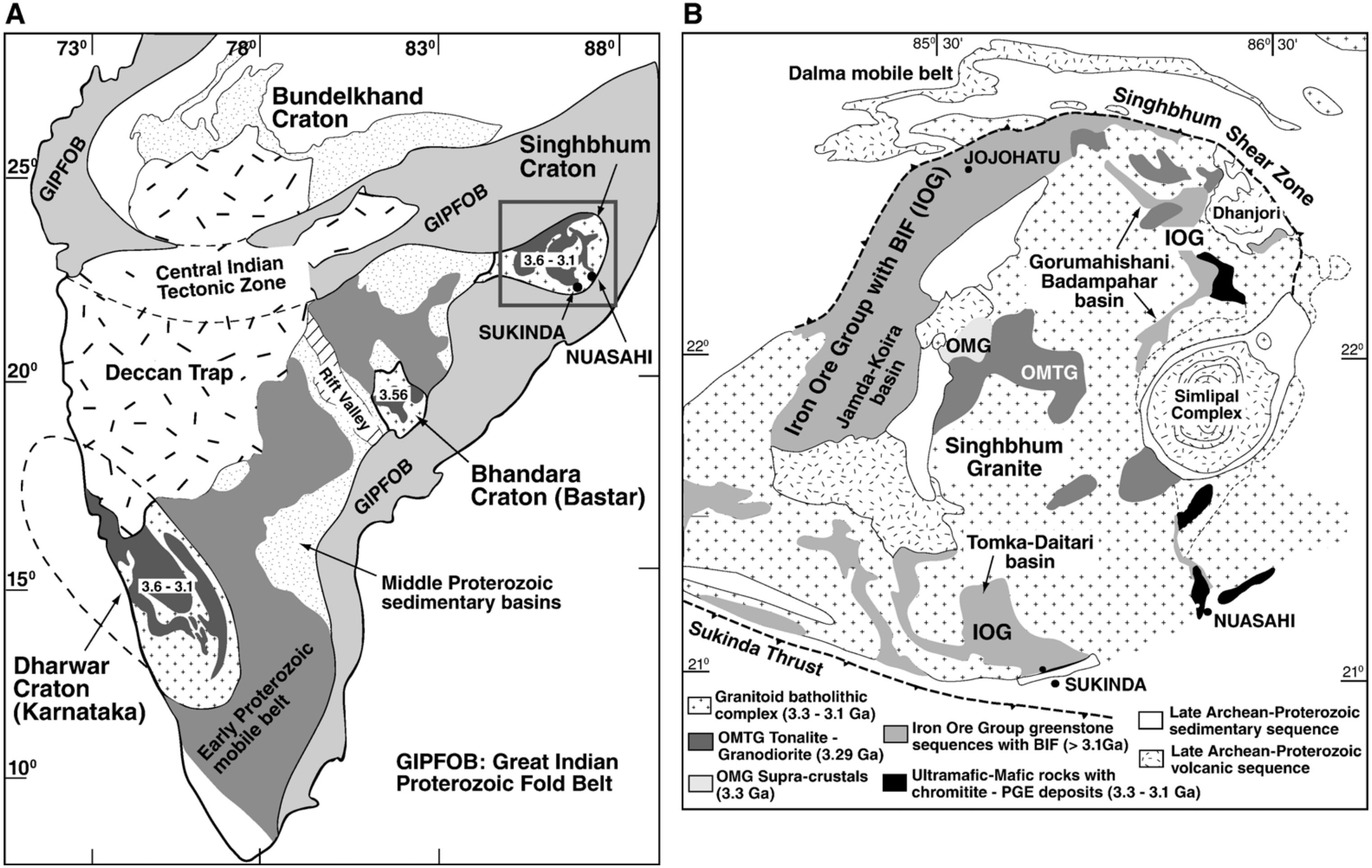


Fig. 1. A: Generalized geology of India showing major tectonic elements, including the four Archean Cratons (after Radhakrishna and Naqvi, 1986; Leelananadam et al., 2006). Geochronological data are from Mishra et al. (1999) and Ghosh (2004). B: Geology of the Singhbhum Craton, India (after Saha, 1994; Mondal et al., 2006a).

oldest unit of the supracrustal suite with zircon U–Pb dates clustering at 3.5, 3.4, and 3.2 Ga (Mishra et al., 1999). A Sm–Nd whole rock isochron for the amphibolites of the OMG yielded an age of 3305 ± 60 Ma (Sharma et al., 1994), which was interpreted to reflect the crystallization age of the primary igneous suite. This suite occurs as remnants along with the Older Metamorphic Tonalitic Gneiss (OMTG) (zircon U–Pb dates clustering at 3.4 and 3.2 Ga Mishra et al., 1999) within the granite–batholithic complex. The Singhbhum

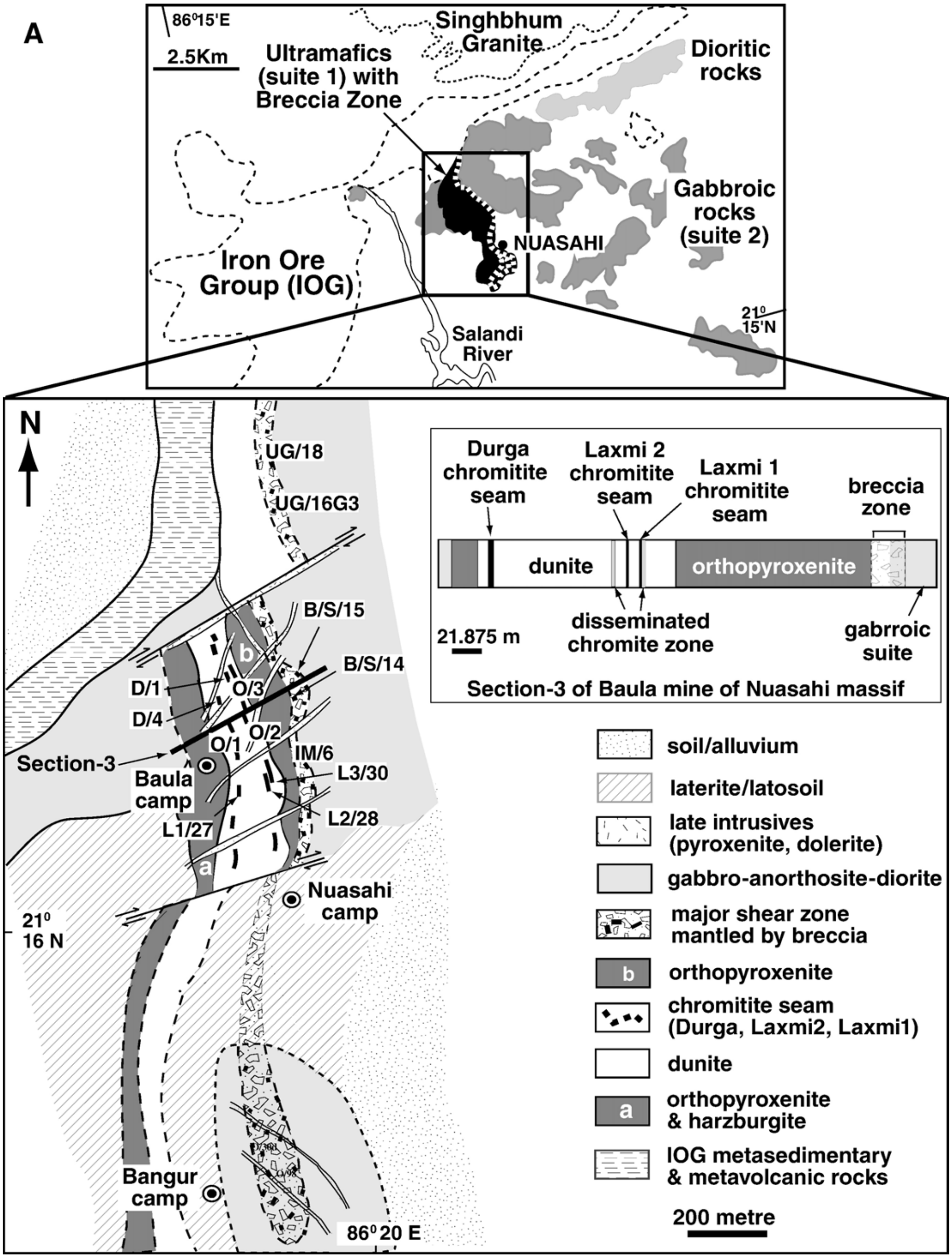


Fig. 2. (A) Geology of the Nuasahi massif (after Mondal, 2000); (B) Geology of the Sukinda massif (after Basu et al., 1997).

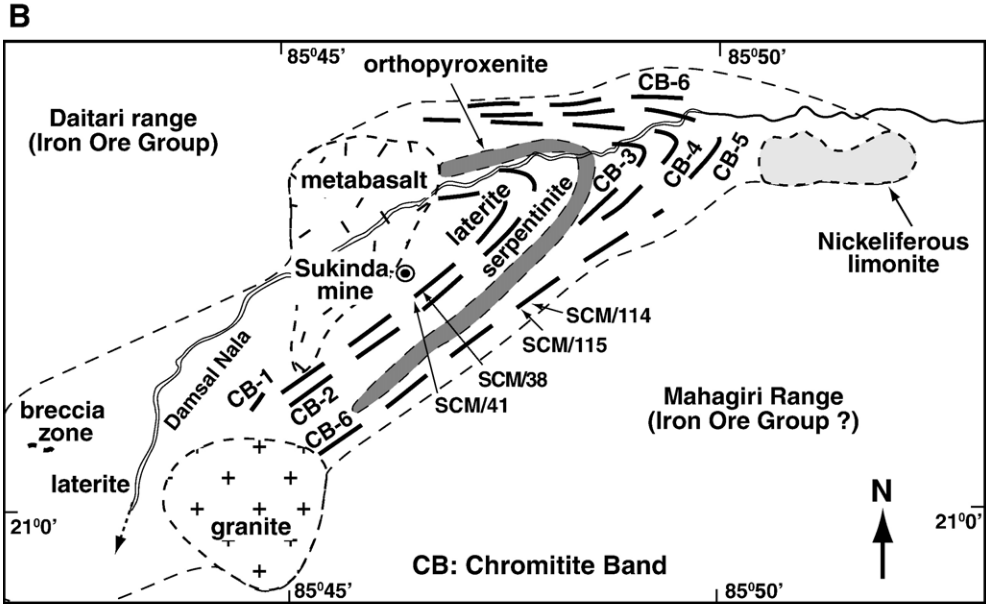


Fig. 2 (*continued*).

granite is the largest granite batholith in the craton (Fig. 1B) and has been divided into two phases: granite of "Type A" (characterized by gently sloping REE pattern with slightly depleted HREE [Saha, 1994]), which has produced a zircon U–Pb date of 3328 ± 7 Ma (Mishra et al., 1999), and granite of "Type B" (characterized by LREE fractionated pattern with flat HREE [Saha, 1994]), which has been dated at ~3.1 Ma using the Pb-Pb whole-rock method (Saha, 1994). The IOG greenstone sequence with Banded Iron Formation (BIF) comprises another supracrustal suite and occurs in three folded belts (Jamda–Koira, Gorumahishani–Badampahar and Tomka–Daitari) (Fig. 1B) surrounding the Singhbhum granite–batholith.

No dates for the chromitite-bearing ultramafic intrusions of the Nuasahi and Sukinda massifs are available, however field relations and available ages for the associated rocks suggest possible age limits between 3.3 and 3.1 Ga. The gabbroic matrix and pegmatitic gabbro of the breccia zone and the closely associated gabbroic suite from the Nuasahi area yielded U–Pb zircon ages of 3123 ± 7 and 3119 ± 6 Ma and have generated a whole rock Sm–Nd isochron age of 3205 ± 280 Ma (Augé et al., 2003). During late Archean to early Proterozoic the Singhbhum Craton experienced extensive mafic–ultramafic magmatism now present within the encircling younger mobile belts (Fig. 1B). An extensive dolerite dyke swarm, known as the Newer Dolerite Suite, intruded the mid-Archaean terrains during this period.

2.2. Chromite deposits

The general geological description and details of petrological characteristics of the ultramafic suites, including the chromite and PGE deposits of the Nuasahi and Sukinda massifs, can be found in Mondal et al. (Mondal et al., 2001, 2006a) and in Augé et al. (2003).

The Sukinda chromite deposits (Fig. 2B) comprise the largest chromite resources in India and consist of a package of extrusive metabasalts and intrusive ultramafics (Basu et al., 1997). The ultramafic component consists of serpentinized dunite, orthopyroxenite, and chromitite. The subvertical chromitite bodies are now present in the serpentinite and occur as disconnected and irregular lenses or bands. The Nuasahi chromite deposits (Fig. 2A) consist of an interlayered sequence of serpentinized dunite, orthopyroxenite, harzburgite, and chromitite. Three major dismembered, subvertical to steeply dipping chromitite seams occur in dunite. All the massive chromitite bands in the Nuasahi massif contain high-Mg chromite (Fig. 3), the Mg# and Cr# of which ranges between 62–77 and 75–87 (Mondal et al., 2001, 2006a). In the Sukinda chromite deposits, the chromite in massive chromitites are also compositionally characterized by high Mg# (62–73) and high Cr# (75–81) (Mondal et al., 2006a). The co-existing olivine and orthopyroxene in host rocks are characterized by high Fo content ($Fo_{92–95}$) and high En content ($En_{89–94}$), respectively (Mondal et al., 2006a). The disseminated chromites in high-Mg metaigneous rocks of the IOG

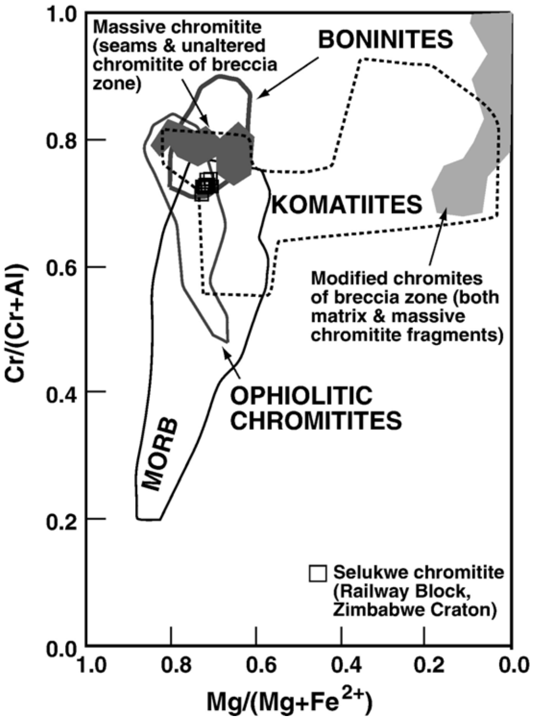


Fig. 3. Cr#–Mg# variations in chromites compared with chromites produced from primitive magma. Chromite data are from (Mondal et al. 2006a) and Mondal and Stumpfl (unpublished data). Chromite data fields are after (Barnes and Roeder, 2001; Mondal et al., 2006a).

exposed in the Nuasahi area are extensively altered and are compositionally more similar to altered accessory chromites in the serpentinized rocks of the ultramafic intrusions.

2.3. PGE deposits

The largest PGE deposits occur in a breccia-filled shear zone in the Nuasahi massif (Fig. 2A). This shear zone is present discordantly at or near the interface between orthopyroxenite and gabbro. The shear zone is filled with breccia composed of blocks (cm to tens of meters across) of chromitites and ultramafic rocks of ultramafic suite in a hybridized pegmatitic gabbroic matrix with a variable mineralogy of primary and secondary assemblages. The breccia assemblages, including the PGE and sulfide mineralization, were primarily due to magmatic metasomatism of preexisting ultramafic rocks by volatile-rich gabbroic melt within the shear zone (Mondal et al., 2001). This evolved gabbroic melt impregnated the shear zone during its waning stages and after the intrusion of the main gabbroic suite.

Both sulfide-free and sulfide-bearing zones of PGE-enrichments are present in the breccia zone (Mondal et al., 2001; Augé et al., 2002). Platinum group element concentrations are highly variable with values up to 26 ppm is reported for the altered chromite-sulfide assemblages (Mondal et al., 2001). The PGE minerals are dominated by Pd, Pt, Bi, Sb, and Te bearing phases (e.g. sudburyite, michenerite–testibiopalladite, stibiopalladinite, sperrylite, merenskyite along with Pd-bearing melonite) and are commonly found in the matrix assemblages together with base metal sulfides (BMS) and disseminated chromite grains. Similar PGE and BMS assemblages are also present in more altered portions of massive chromitite fragments. Irarsite–hollingworthite is commonly present as tiny inclusions within cobaltite–gersdorffite crystals in the sulfide-rich assemblage (Mondal et al., 2001). Sulfide-

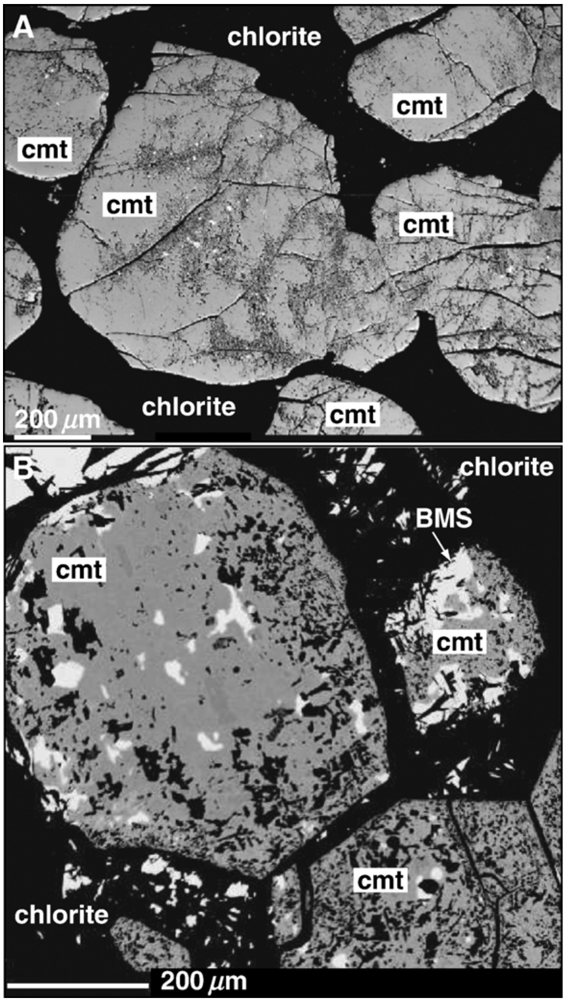


Fig. 4. Back-scattered electron images of chromitite from the breccia showing (A) chlorite (dark) and base metal sulfide (BMS, white) inclusions within chromite, (B) chlorite (dark), chalcopyrite (white) and ilmenite (dark grey) inclusions within chromite. Base metal sulfide, chlorite, and chromite show extensive reaction-replacement texture (right).

free PGE mineralization is Pt-dominated and has been reported from the southern part of the breccia zone (Fig. 2A) (Augé et al., 2002). Isoferroplatinum along with braggite and malanite are commonly found to occur in association with unaltered pyroxene and plagioclase grains in matrix assemblages from the southern part of the breccia zone. Laurite and Os–Ir alloys are present in – chromite as inclusions as documented from the breccia by Augé et al. (2002).

The breccia assemblages have been variably affected by hydrothermal alteration (Fig. 4). Extensive reaction-replacement textures between co-existing chromite, chlorite, and BMS minerals are commonly developed in the matrix as well as in the oxidized portions of the chromitite fragments. Ferrianchromite of the oxidized samples contain abundant inclusions of sulfides, chlorite, ilmenite, magnetite, rutile, as well as platinum-group minerals (Fig. 4).

The chromite compositions in different assemblages in the breccia zone are highly variable (Fig. 3) (Mondal, 2000; Mondal et al., 2001). In sulfide-rich assemblages (Fig. 4B), in the matrix, and in highly oxidized portions of the massive chromitite blocks (Fig. 4A) chromite is Fe-rich ferrianchromite (Fig. 3). The compositions of some of these chromites are more similar to ferritchromit (very high Cr/Al ratios due to Al loss with a concomitantly high Fe^{2+}# and Fe^{3+}#) (e.g. Barnes and Roeder, 2001), which formed due to hydrothermal alteration of the original assemblages.

3. Samples and analytical techniques

The samples used for this isotopic study belong to the Nuasahi and Sukinda massifs (Fig. 2, Tables 1 and 2). They were collected systematically across the ultramafic belt in different mine sections. The chromitite samples used for this study were previously characterized by electron microprobe (Mondal et al., 2001, 2006a). 14 of 19 samples are purified chromite separates from unaltered massive chromitite of seams (Fig. 2, Table 1). These include the chromite separates of sample D/1, which is from a massive-type spotted (rounded olivine) chromitite. The remaining five samples were separated from large chromitite blocks from within the PGE and BMS mineralized breccia zone of the Nuasahi massif. Among these, the composition of unaltered chromites in sample B/S/15 is similar to those of chromites of massive chromitites from seams (similar Cr# and Mg#; Mondal et al., 2006a). Four samples of chromites of the breccia-hosted chromitites have highly variable compositions (Fig. 5). Among these, sample IM/6 is less altered (relatively less Fe^{3+}# and high Cr#) than the other three.

Four whole rock powders of high-Mg metaigneous rocks from the Nuasahi area of the IOG were prepared for Sm–Nd isotopic analyses in the mineral separation laboratories at the University of Copenhagen. The chromite separates used for the Re–Os isotopic study were prepared from hand-crushed samples of massive chromitites in the geochemistry laboratory at Indiana University by handpicking and by using a hand magnet to separate the finer chromite fraction.

The chemical separation techniques for Re and Os used in this study have been previously published (Nägler and Frei, 1997). Samples were dissolved and equilibrated with an ^{185}Re and ^{190}Os spike in “inversed” *aqua regia* digestion in sealed Carius tubes. 200–300 mg chromite (50–200 micron grain size), together with 6 ml of inversed *aqua regia* (1/3 concentrated HCl, 2/3 concentrated HNO_3) were frozen into the tubes, sealed and heated to 230 °C for 2 days. Os was extracted from the inversed *aqua regia* by distillation directly into 3 ml 8 N HBr and purified by a microdistillation procedure adopted by Birck et al. (1997). Rhenium was purified by extraction into a ~1% solution of tribenzylamine (TBA) in $CHCL_3$, following the method of Walker (1988). Total analytical blanks averaged about 15+/−6 pg for Re (with a natural composition of Re) and 9+/−3 pg for Os (with ^{187}Os/^{188}Os ratios of 0.16+/−0.02). All data are blank corrected and respective blank uncertainties were propagated to the final isotopic and concentration data. The blank corrections were negligible for the Os analyses, and minor for most Re analyses.

The isotopic composition of Re was measured separately via solution ICP-MS, performed on a Perkin Elmer Elan 5000 quadrupole instrument at the Geological Survey of Denmark and Greenland, whereby the solutions were doped with natural Ir for online mass bias control of the ^{185}Re/^{187}Re ratio via the ratio ^{191}Ir/^{193}Ir. The external reproducibility for repeated analyses of standard solutions of comparable amounts are approximately +/−0.3% for Re.

Os isotopic analyses were performed on a VG Sector 54 IT solid-source negative thermal ionization mass spectrometer at the University of Copenhagen, using a multi-collector static routine. Analyses were accomplished using Faraday cups. ^{189}Os/^{188}Os = 1.21978 was used for in-run fractionation corrections. Samples were loaded in 1.5 ml 8 N HBr onto high purity Pt filaments and 0.5 ml of a saturated $Ba(OH)_2$ solution were added as an ionization activator. Samples were analyzed at temperatures >800 °C. Repeated analyses of 1 ng loads of the University of Maryland Johnson Matthey reference solution yielded an external reproducibility

Table 1
Re and Os concentrations and Os isotopic data of chromite separates from massive chromitites, Singhbhum Craton, eastern India

Sample	Locality	$^{187}Os/^{188}Os$	$2\sigma_{pop}$	Os (ppb)	Re (ppt)	$^{187}Re/^{188}Os$	T_{Ma} (Ma)	T_{RD} (Ma)	$^{187}Os/^{188}Os_i$ at 3200 Ma	γ_{Os}
Nuasahi massif (massive chromitite seam)										
D/1	Baula mine	0.10602	0.00004	145.1	1377	0.0456	3400	3023	0.10352	−1.42
D/4	Baula mine	0.10371	0.00001	815.4	1631	0.0096	3432	3351	0.10318	−1.74
L1/27	Nuasahi mine	0.11769	0.00014	11.1	577	0.2489	3467	1338	0.10406	−0.90
L2/28	Nuasahi mine	0.10551	0.00001	72.4	550	0.0365	3396	3095	0.10352	−1.42
L3/30	Nuasahi mine	0.10530	0.00002	82.1	648	0.0379	3443	3126	0.10322	−1.70
O/1	Baula mine	0.10410	0.00000	102.6	646	0.0303	3558	3296	0.10244	−2.45
O/2	Baula mine	0.10541	0.00006	91.8	1001	0.0524	3564	3110	0.10254	−2.35
O/3	Baula mine	0.10485	0.00001	137.3	1202	0.0421	3553	3190	0.10254	−2.35
O/30d	Bangur mine	0.10606	0.00002	157.7	1128	0.0344	3293	3017	0.10418	−0.79
O/98	Bangur mine	0.10739	0.00018	25.0	490	0.0942	3670	2828	0.10223	−2.64
Average							3478	2937	0.10314	−1.78
1σ							109	581	0.00069	0.65
Sukinda massif (massive chromitite seam)										
SCM/114	Sukinda mine	0.10430	0.00001	108.1	860	0.0382	3602	3268	0.10221	−2.67
SCM/115	Sukinda mine	0.10532	0.00016	233.6	1891	0.0389	3449	3123	0.10319	−1.74
SCM/38	Sukinda mine	0.10536	0.00003	109.7	994	0.0436	3486	3117	0.10298	−1.94
SCM/41	Sukinda mine	0.10377	0.00005	424.3	1073	0.0122	3444	3343	0.10310	−1.81
Average							3495	3212	0.10287	−2.04
1σ							73	111	0.00045	0.43
Breccia Zone, Nuasahi massif (massive chromitite boulder)										
B/S/14	Baula mine	0.10964	0.00004	73.0	1379	0.0908	3220	2505	0.10467	−0.33
B/S/15	Baula mine	0.10669	0.00002	35.3	1066	0.1454	4531	2927	0.09873	−5.98
IM/6	Nuasahi mine	0.10566	0.00024	93.5	380	0.0195	3227	3074	0.10459	−0.40
UG/16-G3	Ganga mine	0.10982	0.00001	12.7	792	0.3005	9350	2479	0.09337	−11.09
UG/18	Ganga mine	0.12729	0.00010	17.5	1608	0.4412	836	−85	0.10313	−1.79

D/1, D/4, O/1:Durga seam; L1/27, L2/28, L3/30:Laxmi seam; O/2: Laxmi1 seam; O/3:Laxmi2 seam; O/30d, O/98:boulder; SCM/38, SCM/41:OB2 pit (CB1); SCM/114, SCM/115:OB10 pit (CB6) B/S/14, UG/16-G3, UG/18: altered chromitite of breccia zone; B/S/15: unaltered chromitite of breccia zone; IM/6: chromitite pebble of breccia zone, relatively less altered.

Samples D/1 to SCM/41: $Fe^{3+}/(Cr+Al+Fe^{3+})$ ranges between 0.9 and 2.6; B/S/14: $Fe^{3+}/R \approx 43.5$; B/S/15: 2.2; IM/6: 12.4; UG/16-G3: 26.7; UG/18: 40.3 γOs (at 3200 Ma) is calculated using the parameters from Shirey and Walker (1998) and $\lambda = 1 \times 10^{-11}$ year^{-1} (Smoliar et al., 1996). Uncertainties are ±5% or better for Re and $^{187}Re/^{188}Os$, better than ±0.1% for Os and $^{187}Os/^{188}Os$, and better than 0.5% for calculated $^{187}Os/^{188}Os$ and γOs units.

of ±0.07% (2σ; $n=15$) of the ratio $^{187}Os/^{188}Os=0.11377$. Repeated multi-dynamic analyses of 50 ng loads of the University of Maryland Johnson Matthey reference solution using $^{189}Os/^{188}Os=1.21978$ for in-run fractionation corrections yielded an external long-term precision of +/−50 ppm ($2s_m$) of the ratio $^{187}Os/^{188}Os=0.113789$; $n=29$).

For whole rock Sm–Nd isotopic analyses, 300 mg of powdered rock samples were dissolved in a mixture of concentrated HF and 14 N HNO_3 in Teflon beakers on a

Table 2
Sm–Nd concentrations and isotopic compositions for meta-igneous rocks of the IOG, Nuasahi area, Singhbhum Craton

Sample	Sm (ppm)	Nd (ppm)	$^{147}Sm/^{144}Nd$	$^{143}Nd/^{144}Nd$	±2σ mean (%)	T_{DM}	T_{CHUR}	$\varepsilon(0)$	$\varepsilon_{Nd}(T)$ at 3200 Ma	$^{143}Nd/^{144}Nd$ at 3200 Ma
IOG high-Mg metaigneous rocks in the Nuasahi area										
H49C	1.27	5.46	0.14087	0.51164	0.00090	3.18	2.71	−19.51	3.56	0.50866
H54A	4.20	19.73	0.12869	0.51135	0.00090	3.25	2.88	−25.21	2.87	0.50862
H54B	4.32	19.19	0.13632	0.51143	0.00100	3.39	3.02	−23.47	1.45	0.50855
H54C	3.72	17.38	0.12956	0.51135	0.00090	3.27	2.90	−25.08	2.64	0.50861
Average						3.27	2.88		2.63	0.50861
1σ						0.09	0.12		0.88	0.00004

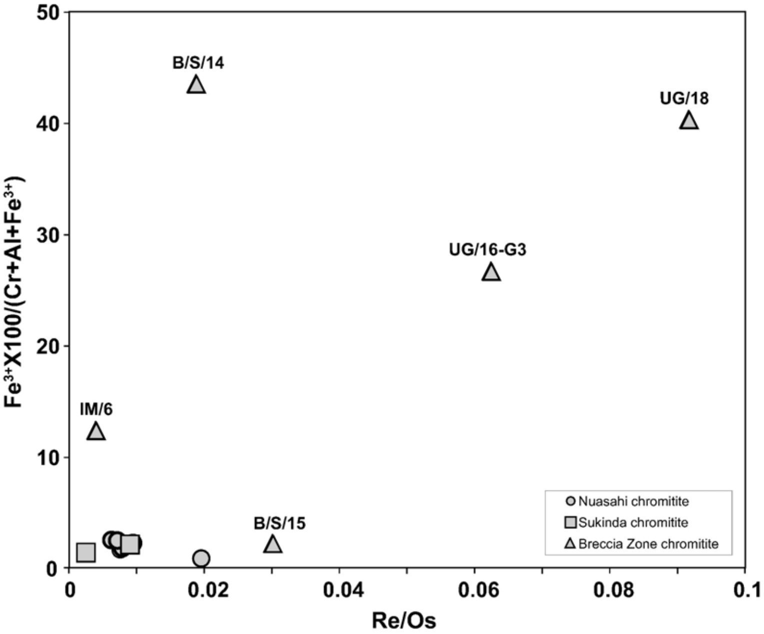


Fig. 5. Re/Os–Fe^{3+}X100/(Cr+Al+Fe^{3+}) variations in chromites from massive chromitites of the Nuasahi and Sukinda massifs. Chromites with high Fe^{3+}X100/(Cr+Al+Fe^{3+}) ratios are more altered representing breccia-hosted chromitites.

hot plate for 72 h. Samples were spiked with a mixed ^{147}Sm–^{150}Nd spike beforehand. Chemical separation of REEs from whole rocks was then carried out on conventional 12 ml glass stem cation exchange columns, followed by a separation using HDEHP-coated beads (BIO-RAD) charged in 6 ml quartz glass columns. Isotope analyses were carried out on the same VG Sector 54-IT instrument. Nd ratios were normalized to ^{146}Nd/^{144}Nd = 0.7219. The mean value of our long-term internal JM Nd reference solution analyses (referenced against the Geological Survey of Japan Shin Etsu Nd standard) is 0.51109 for ^{143}Nd/^{144}Nd, with a 2σ external reproducibility of ±0.000015 (fifty five measurements). The Nd isotopic data presented here are normalized to ^{143}Nd/^{144}Nd=0.512102 for the Shin Etsu standard, which is equivalent to 0.511850 for the La Jolla standard (Tanaka et al., 2000). Our own Shin Etsu standard runs during the period of this project yielded an average ^{143}Nd/^{144}Nd value of 0.512095±0.000011 (five measurements).

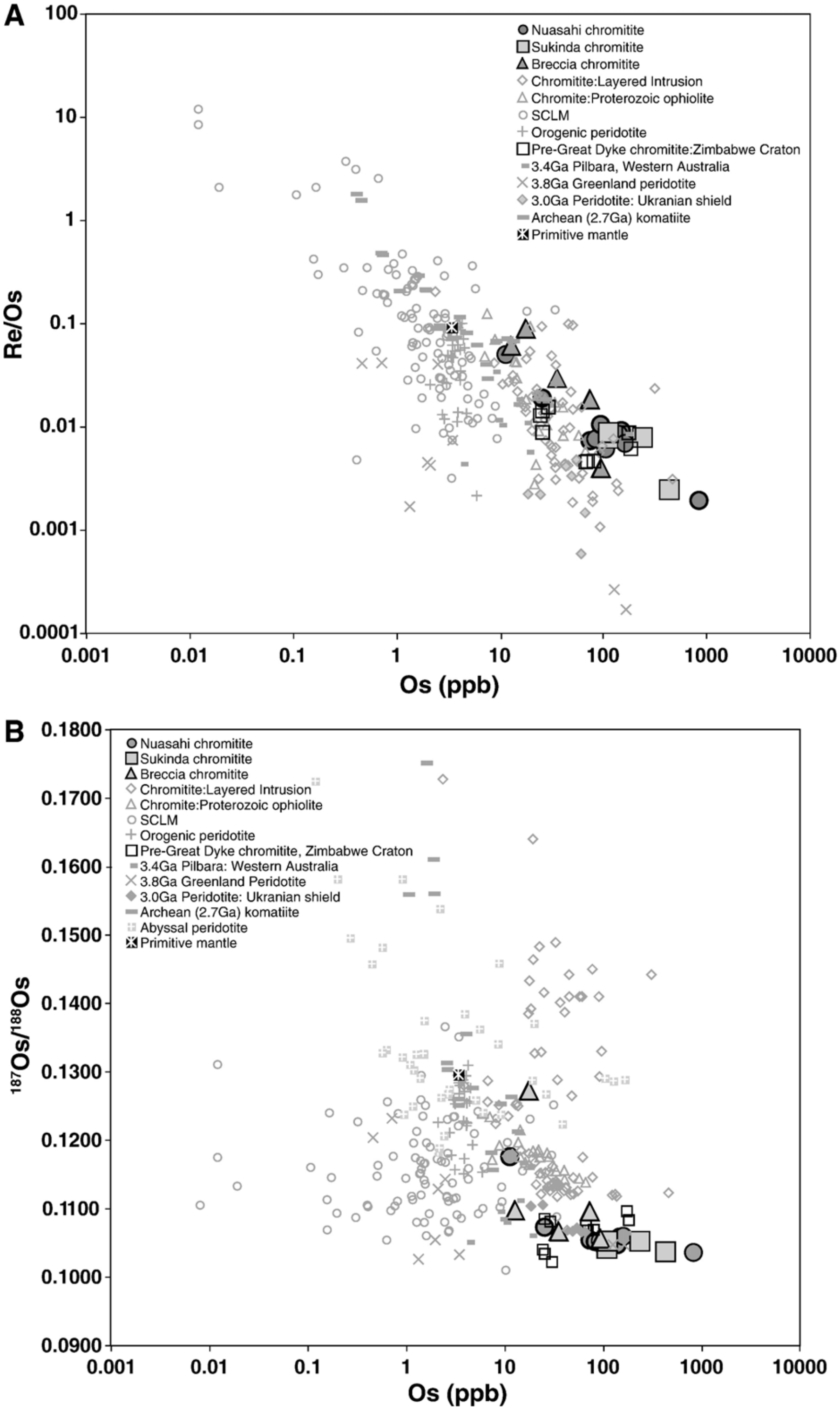
4. Results

4.1. Os isotopes

Re and Os concentrations and Os isotopic compositions of chromite separates from the Nuasahi and Sukinda massive chromitites, as well as chromites from the breccia-zone, are listed in Table 1. Re and Os concentrations vary considerably within each data subset, with Os concentrations ranging from 11.1 ppb to 815 ppb, and Re concentrations ranging from 380 ppt to 1631 ppt. The Os concentrations of these chromites are high and typical of massive chromitite from layered intrusions or from ophiolitic complexes worldwide (Fig. 6) (e.g. Walker et al., 2002; Frei et al., 2006; Ahmed et al., 2006 and references therein), and our values are in agreement with generally high concentrations of IPGE (Os, Ir, Ru) of bulk chromitites from the area (Mondal et al., 2004). In contrast, Os concentrations of chromite from chromitite fragments within the breccia zone are lower (range from 13 to 93 ppb, Fig. 6).

As is typical for chromites, the ^{187}Re/^{188}Os ratios are generally low, and with the exception of one sample (sample UG/18) are less than the chondritic average of approximately 0.4. Despite this, the age correction of Os isotopic ratios was nevertheless significant over the long time span considered (Table 1). Initial ratios are provided as calculated ^{187}Os/^{188}Os ratios, and in the γOs notation, which is the percent deviation of the isotopic composition of a sample from that of the chondritic reference at 3200 Ma, the presumed age of emplacement of the ultramafic magma and associated gabbro (Augé et al., 2003).

When compared with data from other worldwide occurrence in various geotectonic settings (f.e. layered intrusions, ophiolitic complexes, orogenic settings, SCLM), the Singhbhum chromites plot on a similar Re/Os versus Os concentration trend (Fig. 6A) with a slight offset to the right of the trend, which is due to relatively higher Re contents of these chromites. The same is true when comparing our data with bulk rock



and chromite analyses of Archean komatiites and chromite from massive chromitites from Greenland (Bennet et al., 2002), from the Pilbara Craton (Bennet et al., 2002), and from the Ukrainian shield (Gornostayev et al., 2004) (Fig. 6B).

Chromite from massive chromitite seams in the Nuasahi and Sukinda massifs have (with the exception of sample L1/27, Table 1) very unradiogenic $^{187}Os/^{188}Os$ ratios (Nuasahi = 0.10410 to 0.10739 and Sukinda = 0.10377 to 0.10536, Table 1), and these ratios are lower than those generally reported for chromites from large layered intrusions (Fig. 6B), where elevated radiogenic signatures have been interpreted to result from crustal contamination of the parental magmas (e.g. Schoenberg et al., 1999, 2003; McCandless et al., 1999). Instead, the low Os isotopic ratios are comparable to those of ultramafic rocks and xenoliths that are thought to have been derived from SCLM domains (e.g. Walker et al., 1989; Carlson and Irving, 1994; Pearson et al., 1995a,b; Chesley et al., 1999; HanghØj et al., 2001; Lee et al., 2001; Carlson and Moore, 2004) and of chromites and peridotites from Archean cratonic shields (e.g., Nägler et al., 1997; Bennet et al., 2002; Gornostayev et al., 2004), in particular those of pre-Great Dyke occurrences in Archean (2.7 Ga to 3.5 Ga; Nägler et al., 1997) greenstone belts within the Zimbabwe Craton (Fig. 6B).

Initial γOs values of unaltered chromites from massive chromitite layers are consistently negative (range from −0.79 to −2.67) and average $^{187}Os/^{188}Os_i$ values for both mining districts are statistically indistinguishable (Nuasahi = 0.10314 +/− 00069; Sukinda 0.10287 +/− 0.00045, Table 1). In contrast, the initial values of chromites from chromitite fragments within the breccia zone are not meaningful. The breccia initial ratios are highly variable ($^{187}Os/^{188}Os_i = 0.09873$ to 0.10467) and show a huge spread of γOs values ranging from −0.33 to −11.09. As mentioned before, the chromite samples from the breccia zone are variably altered (Figs. 4, 5) and characterized by the presence of abundant inclusions of chlorite, ilmenite, magnetite and Pd, Bi, Te, Sb bearing PGE and BMS minerals (Fig. 4). The presence of such secondary altered assemblages as inclusions within altered chromites with elevated Re/Os ratios (Fig. 6A), and the tendency of these phases to be more easily affected by partial open system behavior during later hydrothermal alteration and weathering might explain the variably radiogenic signature of these chromites. However, the measured Os isotopic compositions of samples B/S/15 and IM/6 are relatively unradiogenic (B/S/15 $^{187}Os/^{188}Os = 0.10669$ and IM/6 $^{187}Os/^{188}Os = 0.10566$) and similar to those of unaltered chromites from seam (Table 1). Both of these samples are relatively less altered (Fe^{3+}# is lower than other samples within the Breccia, Fig. 5) and relatively free of secondary mineral inclusions than the other three samples. Therefore, all that can be said about these strongly heterogeneous Os isotopic signatures of breccia-hosted chromites is that they are likely controlled by inhomogeneous distribution of entrained assemblages of secondary silicates and Pd, Bi, Te, Sb bearing PGE plus BMS minerals within the chromite grains. These may have formed in response to early (i.e. post-magmatic), hydrothermal alteration and/or remobilization of primary PGE-BMS by fluids that might have interacted with crustal rocks, and their Re–Os system was likely affected by open system behavior during subsequent alteration stages.

4.2. Nd isotopes

Sm–Nd concentrations and Nd isotopic data of four high-Mg metaigneous rocks of the IOG from Nuasahi area are presented in Table 2. Our data are compared with existing Sm–Nd isotopic results of amphibolites from the OMG, tonalitic rocks from the OMTG and with data from the Nuasahi gabbroic suite (Sharma et al., 1994; Augé et al., 2003) and plotted on an isochron diagram in Fig. 7. The four whole rock samples show a narrow variation in Sm/Nd ratio with values similar to those of the gabbroic suite in the Nuasahi massif as well as the OMG amphibolites. Our data plot close to the ~3.3 Ga reference isochron reported by Sharma et al. (1994) for the OMG amphibolites and suggests the more or less contemporaneous formation of the IOG high-Mg igneous rocks with the rocks from the OMG. In addition,

Fig. 6. Comparison of Re/Os vs. Os concentrations (B) and Re/Os vs. measured Os isotopic compositions (B) from chromites of the Nuasahi and Sukinda massifs, Singhbhum Craton compared to those of other occurrences worldwide. Chromitite from layered intrusions representing Bushveld, Stillwater, and Great Dyke Complex; Chromitite from Proterozoic ophiolites representing Jormua and Outokumpu ophiolite; SCLM data representing Wyoming, Kaapvaal, Siberia, Zimbabwe, Tanzania and Greenland Craton; Orogenic peridotite representing Pyrenean peridotites; Archean komatiites representing Pyke Hill and Alexo, Ontario, Canada; Abyssal peridotites representing samples of the Southwest Indian Ridge and the Mid-Atlantic Ridge. Data sources: Walker et al. (1989); Carlson and Irving, (1994); Pearson et al. (1995a,b); Reisberg and Lorand, (1995); Walker et al. (1996); Nägler et al. (1997); Burnham et al. (1998); Chesley et al. (1999); McCandless et al. (1999); Tsuru et al. (2000); Horan et al. (2001); HanghØj et al. (2001); Standish et al. (2002); Bennet et al. (2002); Schoenberg et al. (1999, 2003); Gangopadhyay and Walker (2003); Gornostayev et al. (2004); Puchtel et al. (2004).

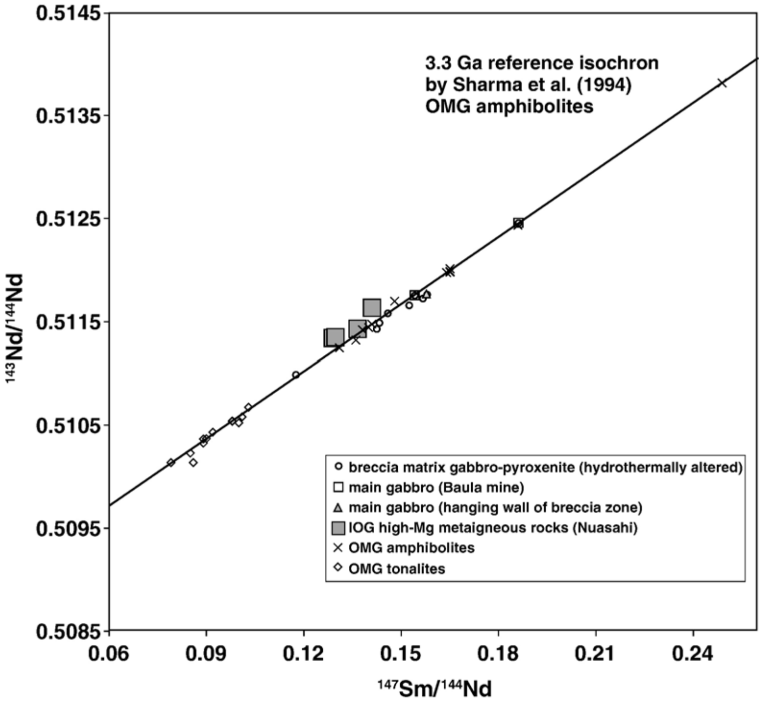


Fig. 7. Comparison of Sm–Nd isotopic characteristics of the chromite bearing high-Mg metaigneous rocks of the Iron Ore Group (IOG) with the Nuasahi gabbroic suite, Older Metamorphic Group (OMG) amphibolites, and rocks of the Older Metamorphic Tonalitic Gneiss (OMTG) in the Singhbhum Craton. Data sources: Sharma et al. (1994); Augé et al. (2003).

the data support the assertion that the presumed emplacement age of the ultramafic suite would be between 3.3 and 3.1 Ga. The average initial Nd ratio, calculated at the presumed emplacement age of 3.2 Ga, is 0.50861 +/− 0.00004 with a corresponding average ε_{Nd} value of 2.63 +/− 0.88. For these high-Mg metaigneous rocks this is consistent with an origin from a long-term depleted source region (Mondal et al., 2006b).

5. Discussion

5.1. Heterogeneity of upper mantle

A steadily increasing amount of data are becoming available which suggest that materials from <1 Ma convecting upper mantle have Os isotopic compositions that are broadly similar to chondritic meteorites (e.g. Walker et al., 2002). However, there is also a growing amount of evidence that points to the fact that mantle heterogeneity in terms of siderophile elements, particularly with respect to Os isotopic signatures, increases with increasingly younger upper convecting mantle domains (e.g. Snow and Reisberg, 1995; Schiano et al., 1997; Frei et al., 2006; Ahmed et al., 2006 and others).

Many peridotites that have recently been removed from the mantle, particularly from old SCLM, are observed to have Os isotopic compositions that are much less radiogenic than chondritic meteorites (e.g. Walker et al., 1989; Pearson et al., 1995a, 1995b and others). The depleted, subchondritic Os isotopic compositions in these rocks have been attributed to long-term Re depletion, which evidently reflects ancient melt removal, coupled with the moderate incompatibility of Re but compatibility of Os, during mantle melting. Similarly unradiogenic Os isotopic ratios are typically encountered in orogenic peridotites such as Ronda and the Pyrenees (Reisberg et al., 1991; Reisberg and Lorand, 1995), which are thought to represent Proterozoic SCLM. Some peridotite xenoliths removed from the SCLM underlying Archean portions of the Siberian, Kaapvaal, Greenland, Wyoming, Tanzania, and Siberia Cratons indicate melt depletion as much as 3.6 Ga ago (e.g. Walker et al., 1989; Carlson and Irving, 1994; Pearson et al., 1995a,b; Chesley et al., 1999; HanghØj et al., 2001; Lee et al., 2001; Rudnick and Lee, 2002; Griffin et al., 2004; Bernstein et al., 2006). However, the heterogeneity of the siderophile elements as seen in SCLM xenoliths may result from the introduction of several generations of metasomatic sulfides as demonstrated by some workers (e.g. Alard et al., 2000, 2002; Griffin et al., 2004).

Recently Walker and Stone (2001) reported significantly subchondritic Os isotopic compositions for an Archean volcanic rock, the Boston Creek flow of the Abitibi Greenstone Belt (Ontario, Canada). This type of Os isotopic signature suggests derivation from ancient SCLM. In addition to SCLM, ^{187}Os depleted materials have also been identified in oceanic rocks (e.g. Hassler and Shimizu, 1998; Parkinson et al., 1998; Harvey et al., 2006). The depleted compositions were interpreted to reflect either derivation from long-term Re-depleted SCLM that had been delaminated from the craton, or that highly Re-depleted reservoirs can survive for more than 1 Ga in the convecting mantle. Negative γOs values of up to −5 have been reported for some portions of the ca. 1.95 Ga Jormua ophiolite in Finland by Tsuru et al. (2000), interpreted to indicate derivation of the melts from a mantle source that was depleted in Re prior to 3.1 Ga ago.

It has been shown that in terms of Os isotopic compositions the Earth's primitive upper mantle (PUM) is chondritic, which implies that Re and Os have occurred in chondritic relative abundances in the bulk upper mantle throughout Earth's history (Meisel et al., 1996). However, the chondritic Os isotopic ratios vary considerably, and systematic differences are observed between the various meteorite classes (e.g. Shirey and Walker, 1998; Meisel et al., 1996, 2001). Since most of our comparison is based on subtle variations of the Os isotopic compositions relative to that of a 'chondritic' mantle evolution curve, the choice of this curve is an important issue. We have used the mantle evolution parameters of Shirey and Walker (1998) to calculate model ages and initial γOs values (average chondrite ^{187}Os/^{188}Os = 0.12672 and ^{187}Re/^{188}Os = 0.40186). However, in Fig. 8 the mantle evolution curve is based on the primitive upper mantle ^{187}Os/^{188}Os ratios (0.1296) deduced by Meisel et al. (2001). If we use this curve for the calculations of model ages and initial γOs values, it can be found that the model ages are more than 100 Ma older and initial γOs values at 3200 Ma nearly 1 unit lower. However, it is noted that the Shirey and Walker (1998) curve has been used historically, which facilitates comparison with other studies and that is the reason of using their mantle evolution parameters for our calculations.

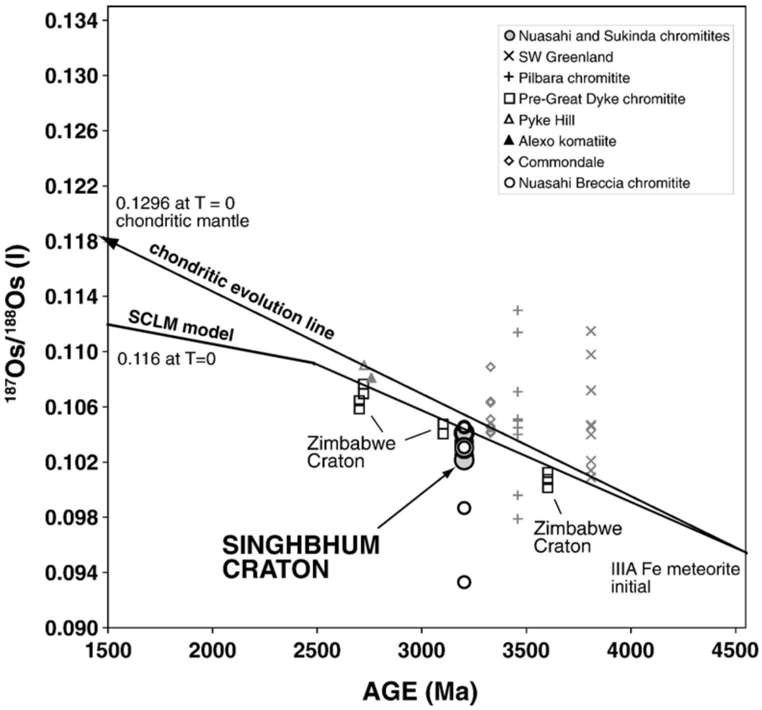


Fig. 8. Comparison of initial Os isotopic compositions of Archean chromites and bulk komatiitic rocks with respect to the chondritic evolution trend of the Re–Os isotopic system. The SCLM model trend is after Nägler et al. (1997). Present day chondritic mantle composition is after Meisel et al. (2001); IIIA meteorite initial at 4558 Ma is after Shirey and Walker (1998). Other data sources: Bennet et al. (2002); Gangopadhyay and Walker, (2003); Wilson et al. (2003); Puchtel et al. (2004).

5.2. Implications for the evolution of the lithospheric mantle beneath the Singhbhum Craton

Earlier studies, based on results from xenoliths from different continents, have revealed that cratonic SCLM is characterized by significant Archean Re depletion. In particular, Pearson et al. (1995a) found kimberlite xenoliths from the Kaapvaal Craton to be almost exclusively less radiogenic than estimates of bulk silicate earth (BSE) and concluded that cratonic lithosphere stabilization occurred by at least 3.5 Ga beneath the Kaapvaal Craton. In a follow up study, Nägler et al. (1997), based on subchondritic Re–Os isotope results on chromites separated from massive chromitites of age-constrained Archean (2.7 to 3.5 Ga) ultramafic intrusions in the Zimbabwe Craton, were able to shed light on the evolution of the Archean SCLM through time. These authors concluded, based on a forward model that utilizes present-day SCLM data, that the Zimbabwean SCLM began to be separated from asthenospheric mantle before 3.8 Ga and grew quasi-continuously through the Archean.

Data presented herein from the Nuasahi and Sukinda massifs have remarkably similar characteristics to the chromite data from Zimbabwe (Figs. 6, 8). The T_{Ma} model ages calculated for the chromite samples are in the range of 3.29–3.67 Ga and average 3.48 +/− 0.11 Ga for the Nuasahi suite and 3.50 +/− 0.07 Ga for the Sukinda suite. Both averages are too old to represent the true extraction ages from contemporaneous primitive chondritic mantle, if the zircon U–Pb age constraints of ca. 3.1 Ga (Augé et al., 2003) and the less precise 3.2 Ga Sm–Nd isochron age (Augé et al., 2003) are considered to be representative of their emplacement. Instead, they indicate the sub-chondritic nature of the source from which they crystallized, and are more compatible with subcontinental lithospheric mantle extraction characteristics. Alternatively, because depleted reservoirs can survive within the convecting mantle for hundreds of Ma as identified in oceanic rocks (e.g. Hassler and Shimizu, 1998; Parkinson et al., 1998; Harvey et al., 2006) the parent melts may have come from the depleted zones within the convecting mantle. It may be envisioned that the ancient melt depleted source reservoirs for the parental melts of the chromite-bearing ultramafic suites in the Singhbhum Craton could have formed due to early subduction of sections of melt depleted subchondritic oceanic lithospheric mantle.

Fig. 8 is a summary age versus initial $^{187}Os/^{188}Os$ diagram in which the Singhbhum data presented herein are compared with those of the Zimbabwe Craton, and with initial Os isotopic data from Archean komatiites, peridotites, and chromitites. We have plotted the SCLM model evolution line of Nägler et al. (1997) and the chondritic mantle evolution line for reference purposes. Chromites from the Singhbhum Craton plot along the trend indicated by the Zimbabwean chromites and can likewise be explained by the SCLM model. It can also be seen that our data, similar to the chromites from Zimbabwe, show a much more homogeneous Os isotopic signature (excepting the breccia samples) compared with at least some of the data from early to late Archean komatiites and ultramafic rocks, which (perhaps with the exception of some data points from the Pilbara Craton), indicate chondritic to suprachondritic $^{187}Os/^{188}Os$ ratios. The Singhbhum data match the pre-2 Ga part of the model evolution, where this is characterized by faster apparent $^{187}Os/^{188}Os$ increase relative to the low rate of $^{187}Os/^{188}Os$ increase after the end of the Archean. This was explained by Nägler et al. (1997) as the result of continuous addition of new upper convecting mantle material with higher $^{187}Os/^{188}Os$ ratios to the SCLM during the Archean. This is in contrast to the SCLM reservoir that evolved as a more or less closed reservoir since the Archean.

Sm–Nd isotopic results of ~3.3 Ga amphibolites from the OMG and tonalities from the OMTG (Sharma et al., 1994) point to chondritic initial >3.3 Ga) basement reservoir is supported by the chromite Re–Os isotopic data, as there is a significant overlap between T_{Ma} ages of the chromites and T_{DM} ages of these sandstones. In addition, Mishra et al. (1999) reported $^{207}Pb/^{206}Pb$ ages of 3.5–3.6 Ga for single detrital zircons extracted from rocks of the OMG, which were interpreted to indicate that crustal formation had already been initiated by 3.6 Ga in this region. These results are compatible with the inference from Re–Os systematics of chromites from the Nuasahi and Sukinda massifs that the formation of a SCLM beneath the Singhbhum Craton was likely to have started in the early Archean. Up to now, no continental crustal fragments of this age have been found in the Singhbhum Craton. If the mantle lithosphere formed in situ, and if it remained attached to the overlying craton, then continental crustal rocks produced as the result of the depletion of this SCLM were either subsequently destroyed, or have not been identified because of a lack of surface exposure. The only evidence for the potential existence of early Archean continental crust, predicted to have likely formed as a counter-reservoir to an early SCLM, stems from the detrital zircons in mid-Archean OMG rocks (Mishra et al., 1999) mentioned above.

In contrast to the OMG and OMTG rock suites (~3.3 Ga), and also in contrast to ~3.1–3.2 Ga old gabbros that intrude the chromite-bearing breccia zones and have broadly chondritic initial Nd signatures (Augé et al., 2003) Sm–Nd isotopic data presented here for chromite-bearing IOG high-Mg metaigneous rocks suggest an origin of the parental melts in a Nd-depleted reservoir. Therefore, both Os and Nd systematics strongly suggest that the magmas parental to the chromites in the Singhbhum Craton were derived from a depleted reservoir at 3200 Ma. In fact, the Nd and Os systematics are in quite good agreement, considering that the Os mantle evolution curve is not well-constrained.

The close range of Re–Os and Sm–Nd melt depletion ages for all the Archaean rock suites so far studied from the Singhbhum Craton and corresponding available crystallization dates strongly suggest large degree melt extraction events and concomitant major crust building processes during middle-to late-Archean in the Singhbhum Craton. This scenario is comparable with crust-mantle separation histories of other Archean cratonic blocks that formed during intense middle-to late-Archean episodes of continent formation (e.g. Myers, 1995). The high Mg# of olivine in dunite and orthopyroxene in orthopyroxenite and high Cr# and Mg# of chromites from ultramafic suites (Mondal et al., 2006a) studied herein, are supportive of the mechanism whereby magmatic products formed as the result of high degrees of mantle melting. Studies by Mondal et al. (2006a) showed that the chromitites of the Nuasahi and Sukinda massifs in the Singhbhum Craton and their ultramafic host rocks crystallized from a boninitic magma (Fig. 3), the composition of which is similar to the spatially associated high-Mg metaigneous rocks of the IOG. One hypothesis is that the parental magma that produced the ultramafic suite was produced in a suprasubduction zone setting and may have intruded the volcano-sedimentary greenstone belts at shallower crustal levels. If this was the case, crustal contamination of the magma was obviously minimal. This model allows for 'second stage melting' of a previously depleted mantle reservoir (e.g. Sun et al., 1989), in the presence of fluids derived in response to the dehydration of a subducting slab, and the production of a Re-depleted, boninitic magma. The high Os concentrations (and IPGE-rich character in general; Mondal et al., 2004) of the studied Archean chromitites are supportive of a high degree of melting of a previously depleted upper mantle source domain. A suprasubduction zone setting is also suggested for the origin of the OMG and the OMTG rocks in the Singhbhum Craton (Saha et al., 2004), and it has recently been suggested to explain the existence of boninite-like metabasalts in the 3.7 Ga old Isua Greenstone Belt in Western Greenland (Polat et al., 2002; Polat and Frei, 2005).

5.3. Open system behavior in breccia-hosted chromites

As noted in the previous section, one sample of chromitite (L1/27) from the main ultramafic suite is more radiogenic than the other chromitites in terms of the measured Os isotopic composition. The model melt depletion date for this particular sample is also very old (3467 Ma) and similar to the other unaltered samples, whereas the minimum Re depletion date is much younger (1338 Ma) relative to the rest of the samples. This may indicate the effects of possible Re-addition during later magmatic-hydrothermal stages, most likely related to remobilization of sulfides.

Open system effects in breccia-hosted chromites resulted in geologically meaningless model ages and initial Os isotopic ratios, as indicated in Table 1. Two samples still reveal T_{MA} ages of around 3.2 Ga, which are close to the emplacement age of the ultramafic and gabbroic suite. However, the breccia-hosted chromites also show enhanced scatter in T_{RD} (minimum melt depletion) ages. The fact that most (with the exception of one sample; UG/18) T_{RD} ages are still mid-to late-Archean indicates that open system behavior (most likely Re addition) did occur relatively late in the history of these chromites. The exception to this (as mentioned above) is exemplified by sample UG/18, which shows a suprachondritic Os isotopic composition implying radiogenic Os ingrowth during an extended time period. Chromites from this sample most probably experienced Re-addition sometime shortly after their crystallization. This might have happened in the course of hydrothermal alteration associated with the emplacement of evolved gabbroic melts, potentially by remobilization / renewed introduction of Re-rich sulfides into the breccia zone.

6. Conclusions

1) Re–Os isotopic measurements of chromites from the Nuasahi and Sukinda massifs in the Singhbhum Craton indicate the existence of a SCLM domain beneath the Singhbhum Craton that started to form in the early Archean by depletion of a primitive mantle. This has also been shown for the SCLM beneath the Zimbabwe Craton in southern Africa. Alternative possibility is that the parental magmas for the chromitites were derived from the early Archean depleted mantle region and this depleted region could have remained within the convecting mantle for a few hundred million years before being added to the SCLM beneath the Singhbhum Craton.

2) The T_{RD} age constraints of the chromites are supportive of previously published Sm–Nd and U–Pb zircon geochronological results indicating large-scale melt extraction from a depleted source during middle-to late-Archean. The highly primitive character of chromite, olivine, and orthopyroxene, together with the unradiogenic Os isotopic characteristics of the chromites, indicate extensive partial melting in the mantle. These conclusions are also supported by the positive ε_{Nd} values of spatially associated high-Mg metaigneous rocks of the IOG.
3) The average initial Os isotopic ratios of chromites from two different ultramafic massifs (Nuasahi and Sukinda) are statistically indistinguishable and define subchondritic initial γOs values of −1.8+/−0.7 and −2.0+/−0.4, respectively.
4) The strong unradiogenic character of the chromites suggest that the parental boninitic magma was not crustally contaminated and may have been produced due to second-stage melting of a previously depleted mantle source in a supra-subduction setting as suggested in previous studies by Mondal et al. (2001, 2006a).
5) Highly variable and strongly radiogenic Os isotopic compositions for chromites from the massive chromitites within the breccia zone in the Nuasahi massif are related to a combination of primary processes and late stage, open-system fluid-rock interaction within the shear zone.

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