1 **Classification:** Physical Sciences. Earth, Atmospheric and Planetary Sciences.

2

3 THE RECENT DISTRIBUTION OF LEAD IN THE INDIAN OCEAN REFLECTS THE

4 IMPACT OF REGIONAL EMISSIONS

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- 15
- 16 **Keywords:** Indian Ocean, Pb content, anthropogenic emissions.
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- 18

19 Abstract

20 Humans have injected lead (Pb) massively into the earth surface 21 environment in a temporally and spatially evolving pattern. A significant fraction is 22 transported by the atmosphere into the surface ocean where we can observe its 23 transport by ocean currents and sinking particles. This study of the Indian Ocean 24 documents high Pb concentrations in the northern and tropical surface waters, and 25 extremely low Pb levels in the deep water. North of 20°S, dissolved Pb 26 concentrations decrease from 42-82 pmol/Kg in surface waters to 1.5-3.3 pmol/Kg 27 in deep waters. South of 20°S, surface water Pb concentrations decrease from 21 pmol/Kg at 31°S to 7 pmol/Kg at 62°S. This surface Pb concentration gradient 28 29 reflects a southward decrease in anthropogenic Pb emissions. The upper waters of 30 the north and central Indian Ocean have high Pb concentrations resulting from 31 recent regional rapid industrialization and a late phase-out of leaded gasoline, and 32 these concentrations are now higher than currently seen in the central North Pacific 33 and North Atlantic Oceans. The Antarctic sector of the Indian Ocean shows very low 34 concentrations due to limited regional anthropogenic Pb emissions, high scavenging 35 rates, and rapid vertical mixing, but Pb still occurs at higher levels than would have 36 existed centuries ago. Penetration of Pb into the northern and central Indian Ocean 37 thermocline waters is minimized by limited ventilation. Pb concentrations in the 38 deep Indian Ocean are comparable to the other oceans at the same latitude, and 39 deep waters of the central Indian Ocean match the lowest observed oceanic Pb 40 concentrations.

41 **Significance Statement:**

42 Humans have altered the earth surface environment by massive injection of certain 43 chemicals into our air and water. Although in some cases these injections are 44 detrimental to environmental health and must be monitored to limit the damage 45 (e.g. freons and the ozone layer), in other cases (e.g. freon dissolving into the ocean) 46 there are no harmful consequences, but the chemicals are actually useful as tracers 47 of ocean circulation patterns. Although lead remains a major hazard when it is 48 proximate to humans (e.g. plumbing, housepaint, contaminated soils), in the open 49 ocean lead serves as an inadvertent experiment demonstrating how metals move 50 through the marine environment. This study is the first to examine the fate of 51 human lead in the Indian Ocean.

53 **body**

54 Introduction

55 Although naturally occurring at trace levels throughout the environment, 56 lead (Pb) in the modern ocean is dominated by anthropogenic sources (from leaded 57 gasoline and high temperature industrial activities). As such, it represents one of the 58 great "global geophysical experiments" performed on the earth, comparable to fossil fuel CO₂, nuclear bomb fallout, and atmospheric trace gases: thus we have massively 59 injected Pb into the surface ocean in a time-and-space dependent pattern, and we 60 61 can now observe where and when it has gone. Although Pb concentrations in the 62 open ocean are low and are not hazardous to marine life or humans, oceanic Pb 63 illustrates the evolving extent of the human footprint upon the natural environment. The first valid oceanic Pb data was obtained for samples collected in 1976-77¹, and 64 65 subsequently there have been numerous studies documenting concentrations and isotope ratios of Pb in seawater²⁻⁷. Most of these studies report data from the North 66 67 Atlantic and North Pacific Oceans. There has only been very limited information 68 from elsewhere in the ocean, with almost no useful data from the entire Indian 69 Ocean. Indian Ocean Pb data are crucial to understanding the impact of 70 anthropogenic Pb on the global scale environment because: (1) large scale economic 71 development in recent decades and limited environmental regulation results in very high fluxes of Pb from the south Asian subcontinent into the Indian Ocean 8 , and (2) 72 73 Southern Asia and Oceania eliminated leaded gasoline later than other industrial 74 countries. Hence the flux of lead to the northern and central Indian Ocean during the past 20 years is likely to have been higher than elsewhere in the world. Here we
present Pb concentration data from a Japanese GEOTRACES cruise in a North-South
transect throughout the Indian Ocean that demonstrates that the surface of the
Indian Ocean has higher Pb concentrations than representative sites in the North
Atlantic and North Pacific Oceans, despite very low concentrations in old deep
waters with Southern Ocean sources.

GEOTRACES is an international program directed at understanding the
global-scale distribution of trace elements and their isotopes in the marine
environment⁹. Here we show data from 11 stations from the 2009-2010 Japanese
GEOTRACES transect in the Indian Ocean between 18°N and 65°S (Figure 1). This is
the first study of Pb in the Indian Ocean with a detection limit capable of
documenting the extremely low levels in the deep water of this basin that defines
and refines the significance of the global Pb distribution.

88 **Results and discussion**

Surface water Pb concentrations in the northern and central Indian Ocean
(53-82 pmol/Kg north of 20°S, Figure 2) are now higher than in the central North
Atlantic and North Pacific oceans near Bermuda and Hawaii^{10,11}. Given no northern
deep water formation and limited upper water ventilation, anthropogenic Pb in the
Indian Ocean has not penetrated as deeply as in the Atlantic Ocean, where those
waters influenced by anthropogenic emissions are carried to the north, cool and
become dense in the winter, then circulate to the south at depth moving into the

South Atlantic and Antarctic. In contrast, in the Indian Ocean there is insignificant
cooling and limited transport of northern waters to the south. In the northern
Indian Ocean, both thermocline and intermediate water CFC (ChloroFluoroCarbon)
concentrations are relatively lower and CFC ages are older in the Bay of Bengal than
in the Arabian Sea¹². In the upper waters, at a given density, CFC-derived ages
increase and concentrations decrease from the south to north, with lowest
concentrations and oldest ages in Bay of Bengal¹³.

103 Dissolved Pb concentrations along the north-south transect exhibit strong 104 vertical and horizontal gradients (Figure 2). North of 20°S, dissolved Pb 105 concentrations decrease from 42-82 pmol/Kg in surface waters (with the highest 106 concentration seen at 10°N in the Arabian Sea) to extremely low concentrations of 107 1.5-3.3 pmol/Kg in deep waters. South of 20°S, surface water Pb concentrations 108 further decrease from 21 pmol/Kg at 31°S to 7 pmol/Kg at 62°S. This surface Pb 109 concentration gradient reflects a southward decrease in anthropogenic Pb 110 emissions that are carried by atmospheric aerosols and deposited into the sea 111 surface.

112 The southwest and the northeast monsoons over the northern Indian Ocean 113 drive strongly directional winds with a dominant period in wind stress variability of 114 one year¹⁴. For oligotrophic open-ocean upper waters the residence time of lead is 115 around 2 years¹⁵⁻²⁰. Hence Pb concentrations that we observe for surface waters of 116 the Arabian Sea and the Bay of Bengal derive from sources sampled by the mixture 117 of the different atmospheric flows during the year. During winter (northeast) 118 monsoon (November-February), the weak (~ 5 m/s) northeast trade wind brings 119 cool, dry continental air, but during the summer (southwest) monsoon (June-120 September) the strong (\sim 15 m/s) southwest wind brings humid maritime air into the Arabian Sea²¹. The Arabian Sea has high salinity (usually in the range 35 to 37 121 122 pss) due to excess of evaporation over rainfall. In contrast, the Bay of Bengal has 123 much lower salinity due to the large influx of fresh water from river discharge and 124 high amount of rainfall. Wind directions over the Indian Ocean, which is the main forcing function, reverse twice during the year²². Gaseous and particulate pollutants 125 126 emitted by the Indian sub-continent and the south Asian region are transported 127 over the entire north Indian Ocean by the persistent northeastern low-level monsoonal flow reaching as far south as 5° to 10°S^{23,24}. Although southwest 128 129 monsoon winds are stronger, atmospheric aerosol transport into the Arabian Sea 130 during this time comes mostly from marine sources and from less industrialized 131 areas in Africa. In the Arabian Sea atmospheric trace element fluxes are one to two orders of magnitude larger than in the tropical and southern Indian Ocean²⁵. Our 132 133 data show Pb surface concentrations for the Arabian Sea (stations 5-7) (60-82 134 pmol/Kg) and in the Bay of Bengal (76 pmol/Kg) that are significantly higher than in 135 the Western Equatorial Indian Ocean, reflecting higher inputs by atmospheric 136 deposition. Arabian Sea station 7 (10°N) has the highest surface lead concentration 137 (82 pmol/Kg) in the Indian Ocean.

138The increase in lead concentrations at ~1000 meters for stations 11 and 12139(20-25 pmol/Kg) with respect to the northern stations (10-15 pmol/Kg) is in

140	proximity to a salinity minimum attributable to the presence of the Antarctic
141	Intermediate Water (AAIW). This is a low salinity water mass that in the Indian
142	Ocean sinks at around 45-55°S that spreads northwards at a depth of 1000-1800
143	meters. This feature disappears near a hydrographic front at $10^\circ S^{26}$, which is
144	consistent with lower lead concentrations at 1000 meters (9 pmol/Kg) for station 9
145	(5°S). A weak Pb maximum also corresponds to the salinity minimum at a depth of
146	800 m at station 10 (Fig. 2 and Fig. 3). These Pb maxima corresponding to mid-
147	depth AAIW imply that the Pb supply to the surface water of AAIW formation region
148	\sim 20 years ago was higher than it is now.
149	Dissolved lead concentrations (1.5 pmol/Kg) in the bottom water at 4000m
150	at station 8 (4°N) are similar to that obtained in deep water at 7°S in the Pacific
151	Ocean ⁶ . Here, both sites are 2-3 fold lower than the lowest dissolved Pb
152	concentration reported elsewhere for the world ocean. At station 14 (62°S), the
153	closest station to Antarctica, there is scant vertical gradient with lead
154	concentrations between 4-7 pmol/Kg at all depths except for a slight maximum of
155	13 pmol/Kg at ~200m.

Lead concentrations in old deep waters in the stations around and above the
equatorial zone (1.5-3.3 pmol/Kg) are lower than seen in contemporary Antarctic
source waters. Two processes account for this observation: (1) although
anthropogenic Pb fluxes to the Antarctic are low, they are not zero, and hence
modern Pb concentrations are bound to be higher in the modern Antarctic than in
the waters that sank centuries ago to form the present-day Northern Indian deep

162	waters, and (2) 210 Pb- 226 Ra studies have established that Pb is removed from the
163	deep ocean on a time scale of decades to a century by "scavenging" onto sinking
164	particulate matter and "boundary scavenging" onto bottom sediments ^{15-19, 27} . So it is
165	expected that much of the Pb that may have been present in the north and central
166	Indian Ocean deep waters at the time they sank from the surface will have been
167	removed by scavenging by the time the water reaches the deep northern Indian
168	Ocean. Abyssal waters of the Indian Ocean are occupied by Antarctic Bottom water
169	(AABW) with two significant flows. One flow originates from the Weddell Sea, filling
170	the western basins of the Indian Ocean. The other flow originates from the Adelie
171	Land coast/Ross Sea and filling the eastern basins ^{28,29} . Both waters flow northward,
172	and in the northern basin, they gradually upwell to form the overlying Indian Deep
173	Water (IDW), which occupies the depths between AAIW and AABW. Unlike AAIW
174	and AABW, IDW is not formed in the Southern Ocean. Rather, it is a mixture of
175	NADW carried from the Atlantic sector with Antarctic Bottom Water, forming
176	Circumpolar Deep Water along the path into the Indian Ocean. The details of these
177	two flow paths and mixing could create differences in the source Pb between
178	eastern and western Indian Ocean, which are not apparent in Pb concentrations but
179	may influence isotopic compositions which will be reported elsewhere.
180	In contrast, comparing the CFC concentrations from the subtropical bottom
181	Indian Ocean ¹² with those in the South Pacific and South Atlantic oceans at
182	comparable latitudes, Indian Ocean bottom water CFC concentrations are lower,

183 consistent with its high dissipation rates from tidal mixing and current fluctuations.

184 The generally high dilutions and low CFC concentrations in bottom water of the 185 Indian Ocean result from their distance to the water mass source regions and the 186 relative effectiveness of mixing. In contrast, for Pb in the Southern Indian Ocean, 187 near-bottom Pb concentrations are comparable to those observed in the South 188 Pacific and South Atlantic at similar latitudes. At 23°S and 4000m we observe 5.2 189 pmol/Kg in the Indian Ocean and 4.6 pmol/Kg in the Pacific Ocean (28°S, 88°W) and 190 at 20°S and 4000m we observe 7.3 pmol/Kg in the Indian Ocean and the same 191 concentration in the Western Atlantic Ocean (19°S, 34°W).

192 In conclusion, at this time, the upper waters of the north and central Indian 193 Ocean show extremely high Pb concentrations resulting from anthropogenic 194 emissions from recent regional rapid industrialization and a late phase-out of 195 leaded gasoline. The contemporary Antarctic sector of the Indian Ocean shows very 196 low concentrations due to limited regional anthropogenic Pb emissions, high 197 scavenging rates, and rapid vertical mixing, although these concentrations are 198 probably somewhat higher than they would have been in preindustrial times. There 199 is only limited penetration of anthropogenic Pb into the northern and central Indian 200 Ocean upper waters because of limited ventilation dominated by water derived 201 from the low-Pb Southern Hemisphere, although there is a small Pb maximum 202 associated with the Antarctic Intermediate water. Pb concentrations in the deep 203 Indian Ocean are comparable to those in the South Atlantic and South Pacific at the 204 same latitude, and the deep waters of the central Indian Ocean match the lowest Pb 205 concentrations observed anywhere in the ocean, due to low initial Pb concentrations 206 in the source waters and scavenging of Pb along the flow path. It should be expected

207	that the continued	evolution of humar	activities around th	e Indian Ocean will lead

to a continuing evolution of the anthropogenic footprint of Pb on the ocean.

209 Material and Methods

210 Sample collection information: samples were obtained during the Japanese Indian

211 Ocean GEOTRACES cruise (KH09-5, November to December 2010), which collected

Pb samples from 11 stations from the Bay of Bengal, Arabian Sea, to the Antarctic

213 Circumpolar water (62°S, Figure 1). Seawater samples for Pb analysis were collected

from 11 stations using a CTD carousel with an Epoxy-coated Al frame and Teflon-

215 coated Niskin-X bottles (General Oceanics, 12 L-type), which were thoroughly

cleaned by soaking in 1.5% Extran MA01 (EMD Millipore), 0.1M HCl, and high-purity

217 water. Upon retrieval, Niskin-X bottles were transferred into a HEPA-filtered air

218 "bubble", and seawater samples were pressure-filtered through 0.2 μm-size capsule

219 filters (Pall Scientific, AcroPak 200). Filtered samples were acidified to pH 2.0 on

board with clean HCl and shipped to MIT for analysis.

221 Pb analysis: Total Pb concentration was analyzed by the NTA ID-ICPMS method

described by Lee et al.¹¹. Acid-leached 1.5 mL vials were used. The acidified

seawater sample was poured into the vial and 1.3 mL of the sample was pipetted out

using a clean pipette tip. The 1.3 mL sample in the pipette tip was pipetted back into

the same vial after quickly emptying the vial. Then, the sample was spiked with 25

226 μ L of stable ²⁰⁴Pb isotope spike and left for a few minutes to establish equilibrium

227 between added isotope spikes with their natural isotopes in the samples. Then, the

228 pH of the sample was adjusted to pH=5.3 using ammonium acetate buffer solution 229 (pH=7.9-8.0). Finally, 150 µL of a NTA resin suspension (2400 beads) were added 230 to the vial and it was shaken on an orbital table at 2000 rpm for 4 days to allow the 231 Pb and resin to bind. After 4 days, the sample vial was centrifuged (for 45 seconds at 232 14,500 rpm) and the overlying seawater was siphoned off carefully, leaving the 233 resin beads at the bottom. The resin beads were then rinsed three times with high 234 purity H₂O to remove seasalt. In each rinse, the vial was filled with high purity 235 water, centrifuged down, and the supernatant was siphoned off. After rinsing, 150 236 μ L of 0.5 M HNO₃ were added to the vial, and then two days allowed for Pb to be 237 released into solution. The final solution was then brought to the ICPMS for measurement of the isotope ratio $(^{208}Pb/^{204}Pb)$ of this solution (after on-peak zero 238 239 acid blank correction) which is used to calculate the Pb concentration. Exactly 0.3 240 mL of low-Pb seawater was used to determine the procedure blank. At least three 241 replicates of each sample were measured. Above the detection limit, the precision of 242 the data is about 3% based on a long-term standard with 32 pmol/Kg. The detection 243 limit (3σ of the std. dev. of low concentration samples) is about 1.5 pmol/Kg. By 244 comparison, of the only other publications reporting Pb data from the Indian Ocean, one only reported data as low as 0.01 nM³⁰, a concentration that exceeds that found 245 246 in 40% of the samples in this data set, and the other publication only plotted 247 overlain profiles from some stations in a supplementary figure with no discussion of the data 31 . 248

250 Acknowledgements

- 251 Y. Echegoyen thanks the Spanish Ministry of Science and Innovation for a
- 252 postdoctoral MEC-Fulbright grant. MIT laboratory expenses were supported by a
- 253 grant from the Singapore National Research Foundation to the SMART-CENSAM
- 254 project. Sample collection was supported by grants from the Steel Foundation for
- 255 Environmental Protection Technology and from Grant-in-Aid of Scientific Research,
- the Ministry of Education, Culture, Sports, Science, and Technology of Japan. We
- 257 thank the crew, officers, and onboard scientists of R/V Hakuho Maru during the KH-
- 258 09-5 cruise for their help with sampling and routine analyses.

260 **References**

261	1. Schaule BK, Patterson CC	(1981)	Lead concentrations in the northeast Pacify	ic:
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262 evidence for global anthropogenic perturbations. *Earth Planet Sci Lett*263 32(1):304-312.

- 264 2. Boyle EA, Chapnick SD, Shen GT (1986) Temporal variability of lead in the
 265 western North Atlantic. *J Geophys Res* 91(C7):8573-8593.
- 266 3. Flegal AR, Itoh K, Patterson CC, Wong CS (1986) Vertical profile of lead isotopic
- 267 compositions in the north-east Atlantic. *Nature* 321:689-690.
- 268 4. Helmers E, Rutgers van der Loeff MM (1993) Lead and Aluminum in Atlantic

Surface Waters (50°N to 50°S) Reflecting Anthropogenic and Natural Sources in
the Eolian Transport. *J Geophys Res* 98(C11):20261-20263.

- 5. Alleman LY, Veron AJ, Church TM, Flegal AR, Hamelin B (1999) Invasion of the
- abyssal North Atlantic by modern anthropogenic lead. *Geophysical Res Lett*26(10):1477-1480.
- 6. Wu J, Rember R, Jin M, Boyle EA, Flegal AR (2010) Isotopic evidence for the source
 of lead in the North Pacific abyssal water. *Geochim Cosmochim Acta*
- 276 74(16):4629-4638.
- 277 7. Gallon C et al. (2011) Asian Industrial Lead Inputs to the North Pacific Evidenced

by Lead Concentrations and Isotopic Compositions in Surface Waters and

Aerosols. *Env Sci Tech* 45(23):9874-9882.

8. Flegal AR, Gallon C, Ganguli PM, Conaway CH (2013) All the Lead in China. *Crit Rev Environ Sci Technol* 43(17):1869-1944.

9. SCOR Working group (2007) GEOTRACES – An international study of the global
 marine biogeochemical cycles of trace elements and their isotopes. *Chemie der Erde* 67(2):85-131.

10. Boyle EA, Bergquist BA, Kayser RA, Mahowald N (2005) Iron, Manganese, and
lead at Hawaii Ocean Time-series station ALOHA: Temporal variability and an
intermediate water hydrothermal plume. *Geochim Cosmochim Acta* 69(4):933952.

289 11. Lee JM et al. (2011) Analysis of trace metals (Cu, Cd, Pb, and Fe) in seawater

using single batch nitrilotriacetate resin extraction and isotope dilution

inductively coupled plasma mass spectrometry. *Anal Chim Acta* 686(1-2):93-

292 101.

293 12. Fine RA et al. (2008) Decadal ventilation and mixing of Indian Ocean waters.
294 *Deep Sea Res Part 1 Oceanogr Res Pap* 55(1):20–37.

295 13. Haine TWN, Watson AJ, Liddicoat MI, Dickson RR (1998) The flow of Antarctic

bottom water to the southwest Indian Ocean estimated using CFCs. *J Geophys Res*

297 103(C12):27637-27653.

298 14. Shetye SR, Gouveia AD, Shenoi SSC (1994) Circulation and water masses of the
299 Arabian Sea. *Proc Indian Acad Sci (Earth Planet Sci)* 103(2):107-123.

300 15. Bacon MP, Spencer DW, Brewer PG (1976) ²¹⁰Pb/²²⁶Ra and ²¹⁰Po/2¹⁰Pb

301 disequilibria in seawater and suspended particulate matter. *Earth Planet Sci Lett*302 32(2):277-296.

303 16. Nozaki Y, Thomson J, Turekian KK (1976) The distribution of ²¹⁰Pb and ²¹⁰Po in

the surface waters of the Pacific Ocean. *Earth Planet Sci Lett* 32(2):304-312.

305 17. Cochran JK, Bacon MP, Krishnaswami S, Turekian KK (1983) Po-210 and Pb- 210

306 distributions in the central and eastern Indian Ocean. *Earth Planet. Sci. Lett.*

307 65(2):433-452.

308 18. Chung, Y, Finkel, R (1987) Pb-210 in the western Indian Ocean: distribution,

309 disequiibrium, and partitioning between dissolved and particulate phases. *Earth*310 *Planet Sci Lett* 85:28-40.

311 19. Turekian KK, Graustein WC, Cochran JK (1989) in *Chemical Oceanography*, vol.

312 10, eds Riley JP, Chester R (Academic Press, London), pp. 51-81.

20. Kim G, Hussain N, Scudlark JR, Church TM (2000) Factors influencing the

314 atmospheric depositional fluxes of stable Pb, ²¹⁰Pb and ⁷Be into Chesapeake Bay.

315 *J Atmospheric Chem* 36(1):65-79.

316 21. Rengarajan R, Sarin MM (2004) Atmospheric Deposition Fluxes of ⁷Be, ²¹⁰Pb and

317 Chemical Species to the Arabian Sea and Bay of Bengal. *Indian J Mar Sci* 33(1):56-

318 64.

319	22. Vinayachandran PN, Kurian J (2008) Modeling Indian Ocean Circulation: Bay of
320	Bengal fresh plume and Arabian Sea mini warm pool. Proceedings of the 12th
321	Asian Congress of Fluid Mechanics 18-21 August 2008, Daejeon, Korea*.
322	23. Satheesh SK, Moorthy KK, Murthy BVK (1998) Spatial gradients in aerosol
323	characteristics over the Arabian Sea and Indian Ocean. J Geophys Res
324	103(D20):26183-26192.
325	24. N.W. Tindale, N.W., P.P. Pease (1999) Aerosols over the Arabian Sea:
326	Atmospheric transport pathways and concentrations of dust and sea salt Deep-
327	Sea Research II 46: 1577-1595
328	25. Saager PM (1994) The biogeochemical distribution of trace elements in the
329	Indian Ocean. Proc Indian Acad Sc. (Earth Planet Sci) 103(2):237-278.
330	26. Tchernia P (1980) Descriptive regional oceanography (Oxford, Pergamon Press)
331	253 pp.
332	27. Spencer DW, Bacon MP, Brewer PG (1981) Models of the distribution of Pb-210
333	in a section across the North Equatorial Atlantic Ocean. J. Mar. Res. 39(1):119-
334	138.
335	28. Gordon AL, Tchernia PL (1972) in Antarctic Oceanography II: The Australian-
336	New Zealand Sector, ed Hayes E (AGU, Antarctic Reseach Series, Washington,
337	D.C.), pp. 59-69.
338	29. Mantyla AW, Reid JL (1995) On the Origins of Deep and Bottom Waters of the

339 Indian-Ocean, *J Geophys Res-Oceans*, 100(C2):2417-2439.

340 30. Morley NH, Statham PJ, Burton JD (1993) Dissolved trace metals in the

- 341 southwestern Indian Ocean. *Deep Sea Res Part 1 Oceanogr Res Pap* 40(5):1043-
- 342 1062.
- 343 31. Vu HTD, Sohrin Y (2013) Diverse stoichiometry of dissolved trace metals in the
- 344 Indian Ocean. *Nature Sci Repts* 3:1-5.
- 345

FIGURE LEGENDS

347

348 Figure 1: Map showing station positions and indicating section segments.

349

350 Figure 2: Seawater dissolved Pb section (north-south transects) in the Indian Ocean.

351

352 Figure 3: Vertical Pb profile data from stations in the Indian Ocean.

354 Figures



355 Figure 1: Map showing station positions and indicating section segments

356

- 358 Figure 2: Seawater dissolved Pb section (north-south transect) in the Indian
- 359 Ocean.
- 360



361





366	Station	"Latitude, L	ongitude"	"pressure, dbar"	"Pb, pmol/kg"
367	s.d.	n			
368	2 "08°3	8 1.10'N, 86°0 2	1.55'E" 11	75.9 1.2 3	
369	2	26 72.2	1.4 3		
370	2	50 56.1	1.2 3		
371	2	100 31.2	0.6 2		
372	2	151 38	0.7 3		
373	2	201 29.4	0.2 2		
374	2	401 21.2	0.1 2		
375	2	601 16.8	0.3 3		
376	2	802 15.1	1.2 3		
377	2	999 15.6	4.1 2		
378	2	1250 10.9	0.6 3		
379	2	1500 10.5	0.3 2		
380	2	2000 7.2	1.2 3		
381	2	2501 7.7	0.3 3		
382	2	3001 6.1	0.2 3		
383	2	3500 5.8	0.4 2		
384	2	3706 8.1	0.4 2		
385					
386	Station	"Latitude, L	ongitude"	"pressure, dbar"	"Pb, pmol/kg"
387	s.d.	n			
388	3 "00°()0.34'S, 80°00).38'E" 11	53 1.3 3	

389	3	26	52.8	0.3	2				
390	3	51	47.4	1.1	3				
391	3	101	51.9	0.3	3				
392	3	150	30.7	1.6	3				
393	3	203	29.3	1	3				
394	3	403	20.3	0.4	2				
395	3	601	17.6	1.6	3				
396	3	808	14.6	0.2	3				
397	3	1002	10	0.3	3				
398	3	1251	8.1	0.3	2				
399	3	1504	7.6	0.5	3				
400	3	2001	7.7	0.6	2				
401	3	2501	5.1	1.6	2				
402	3	3002	5.6	0.2	2				
403	3	3501	4.7	0.1	2				
404	3	4000	3.7	0.8	3				
405	3	4500	3.8	0.9	3				
406	3	4730	7	1.4	3				
407									
408	Station	"Latit	ude, Lo	ongitud	le"	"pres	sure, d	lbar"	"Pb, pmol/kg"
409	S.(d. n							
410	5 "1	.6°44.58'N,	, 68°59	.72'E"	10	61.5	2.6	2	
411	5	25	64.2	2.9	2				

412	5	100	44.2	6.2	3				
413	5	150	34.9	0.7	3				
414	5	100	58.6	1.8	3				
415	5	201	34.7	1.5	3				
416	5	400	24.6	0.2	2				
417	5	602	18.9	0.9	3				
418	5	803	15.9	0.9	3				
419	5	1002	12.9	3.5	2				
420	5	1250	19	0.6	2				
421	5	1500	7.9	0.6	3				
422	5	2001	6.5	0.8	2				
423	5	2501	9.7	0.7	2				
424	5	3000	15.8	1	2				
425	5	3500	4.3	0.4	2				
426	5	3658	5.1	0.4	3				
427									
428	Station	"Latit	ude, Lo	ongitud	le"	"pres	sure, d	lbar"	"Pb, pmol/kg"
429	s.d.	n							
430	6 "14°0	0.12'N,	, 68°59	9.45'E"	1	60.7	0.5	3	
431	6	2	48.2	1.4	2				
432	6	5	45.9	1.3	3				
433	6	100	45.8	0.7	3				
434	6	150	37.2	0.8	2				

435	6		200	31.3	0.8	4					
436	6		401	23.9	0.6	3					
437	6		604	16.1	1.2	2					
438	6		801	18.4	0.5	2					
439	6		1000	12.9	0.3	2					
440	6		1253	11.7	1.1	3					
441	6		1504	8.3	1.3	2					
442	6		2004	6.4	0.7	3					
443	6		2501	4.9	0.7	3					
444	6		3001	5.2	0.6	3					
445	6		3501	3.4	0.4	2					
446	6		4001	5	0.4	3					
447											
448	Statio	n	"Latit	ude, Lo	ongitud	le"	"pres	sure, d	lbar"	"Pb, pmol/kg"	
449		s.d.	n								
450	7	"09°5	9.88'N,	, 68°44	.79'E"	11	82.4	0.1	2		
451	7		26	82.4	2	2					
452	7		51	68.7	3.2	2					
453	7		101	43.5	1.9	3					
454	7		151	32.1		1					
455	7		201	29.4	1.2	3					
456	7		400	23.1	1.4	2					
457	7		601	24.1	1.9	3					

458	7	802 1	13.1	0.1	2				
459	7	1003 1	11.6	1.1	3				
460	7	1251	10.9	0.7	2				
461	7	1504 8	8.9	1.5	2				
462	7	1997 7	7.6	1.8	2				
463	7	2501	10.6	0.7	2				
464	7	3004 2	2.6	0.3	3				
465	7	3501 4	4.2	0.6	2				
466	7	4000 (6	1.5	2				
467	7	4500 2	2.1	0.4	2				
468	7	4570 2	2.5	0.2	2				
469									
469 470	Station	"Latitu	de, Lo	ongitud	le"	"press	sure, d	bar"	"Pb, pmol/kg"
469 470 471	Station s.d	"Latitu	de, Lo	ongitud	le"	"press	sure, d	bar"	"Pb, pmol/kg"
469470471472	Station s.d 8 "0	"Latitu l. n 4°00.83'N, 6	de, Lo 69°00	ongitud .42'"	le" 1	"press 66.9	sure, d 2.5	bar" 3	"Pb, pmol/kg"
469470471472473	Station s.d 8 "0- 8	"Latitu l. n 4°00.83'N, 6 2 5	de, Lo 69°00 53	ongitud .42'" 1.1	le" 1 3	"press 66.9	sure, d 2.5	bar" 3	"Pb, pmol/kg"
 469 470 471 472 473 474 	Station s.d 8 "0- 8 8	"Latitu l. n 4°00.83'N, 6 2 5	de, Lo 69°00 53 49	ongitud .42'" 1.1 0.4	le" 1 3 2	"press 66.9	sure, d 2.5	bar" 3	"Pb, pmol/kg"
 469 470 471 472 473 474 475 	Station s.d 8 "04 8 8 8	"Latitu l. n 4°00.83'N, 6 2 5 5 4 100 4	de, Lo 69°00 53 49 46.1	ongitud .42'" 1.1 0.4 2.8	le" 1 3 2 3	"press	sure, d 2.5	bar" 3	"Pb, pmol/kg"
 469 470 471 472 473 474 475 476 	Station s.d 8 "0 8 8 8 8 8	"Latitu l. n 4°00.83'N, 6 2 5 5 4 100 4 200 3	de, Lo 69°00 53 49 46.1 31	ongitud .42''' 1.1 0.4 2.8 3.7	le" 1 3 2 3 3	"press	sure, d	bar" 3	"Pb, pmol/kg"
 469 470 471 472 473 474 475 476 477 	Station s.d 8 "0 8 8 8 8 8 8	"Latitu l. n 4°00.83'N, 6 2 5 5 4 100 4 200 3 401 2	de, Lo 69°00 53 49 46.1 31 22.6	ongitud .42'" 1.1 0.4 2.8 3.7 2.4	le" 1 3 2 3 3 2	"press	sure, d	bar" 3	"Pb, pmol/kg"
 469 470 471 472 473 474 475 476 477 478 	Station s.d 8 "0 8 8 8 8 8 8 8 8 8 8 5	"Latitu l. n 4°00.83'N, 6 2 5 5 4 100 4 200 3 401 2 602 1	de, Lo 69°00 53 49 46.1 31 22.6 18.4	ongitud .42'" 1.1 0.4 2.8 3.7 2.4 0.4	le" 1 3 2 3 3 2 2 2	"press	sure, d	bar" 3	"Pb, pmol/kg"
 469 470 471 472 473 474 475 476 477 478 479 	Station s.d 8 "04 8 8 8 8 8 8 8 8 8 8 5 8	"Latitu l. n 4°00.83'N, 6 2 5 5 4 100 4 200 3 401 2 602 1 801 1	de, Lo 69°00 53 49 46.1 31 22.6 18.4 15.3	ongitud .42'" 1.1 0.4 2.8 3.7 2.4 0.4 2.1	le" 1 3 2 3 3 2 2 2 2	"press	sure, d	bar" 3	"Pb, pmol/kg"

481	8		150	8.7	0.9	3				
482	8		200	6.8	0.8	2				
483	8		300	2.3	0.2	3				
484	8		407	1.5	0.3	2				
485										
486	Statio	on	"Latit	ude, Lo	ongitud	le"	"pres	sure, o	dbar"	"Pb, pmol/kg"
487		s.d.	n							
488	9	"05°1	6.01'S,	, 67°54	.25'E"	1	65.4	5.2	2	
489	9		2	60.9	1.4	3				
490	9		5	37.4	0.9	2				
491	9		10	24.9	0.4	3				
492	9		20	23.1	0.9	3				
493	9		40	17.6	2.7	2				
494	9		80	12.9	1.1	2				
495	9		100	9	2.4	2				
496	9		150	6.5	1.4	2				
497	9		200	3.3	0.9	2				
498										
499	Statio	on	"Latit	ude, Lo	ongitud	le"	"pres	sure, o	dbar"	"Pb, pmol/kg"
500		s.d.	n							
501	10	"19°5	9.63'S,	, 72°32	.67'E"	11	42.1	1.1	2	
502	10		25	43.6	2	3				
503	10		51	45.2	1.2	2				

504	10	101	43.1	1.7	2				
505	10	151	34.3	0.7	2				
506	10	200	33.7	0.8	2				
507	10	400	20.5	1.1	4				
508	10	601	16.1	0.2	2				
509	10	800	17	0.2	2				
510	10	1001	11	0.2	3				
511	10	1251	10.8	1.1	2				
512	10	1501	13.3	2	3				
513	10	2000	7.4	0.6	2				
514	10	2502	5.6	1.6	2				
515	10	3000	4	1.6	3				
516	10	3500	12.1		1				
517	10	4001	7.3	1.6	2				
518	10	4367	12	2	2				
519									
520	Station	"Latit	ude, Lo	ongitud	le"	"pres	sure, c	lbar"	"Pb, pmol/kg"
521	s.d.	n							
522	11 "30"	°00.02'S,	64°59	.93'E"	10	28.8	1.6	3	
523	11	25	33.3	1	3				
524	11	50	23.8	1.3	3				
525	11	100	23.7	2.2	2				
526	11	151	22.8	1.3	2				

527	11	199	22.8	1	3				
528	11	400	21.3	0.5	2				
529	11	601	20.2		1				
530	11	800	19.8	0.1	2				
531	11	1001	24.7	6	2				
532	11	1252	21	3.2	2				
533	11	1500	16.7	2	2				
534	11	1999	7.2	0.3	2				
535	11	2501	7.7	2.8	2				
536	11	3001	7.7	3.2	2				
537	11	3501	9.4	0.2	2				
538	11	4001	5.2	1.4	2				
539	11	4500	7.2	0.3	2				
540	11	4782	7.3	2.2	2				
541									
542	Station	"Latit	ude, Lo	ongitud	le"	"pres	sure, c	lbar"	"Pb, pmol/kg"
543	s.d.	n							
544	12 "37°4	45.14'S,	57°37	.13'E"	11	21.3	3.1	2	
545	12	25	25.9	1.4	2				
546	12	50	20.6	0.7	2				
547	12	100	20	1.1	2				
548	12	151	20.9	1.6	2				
549	12	200	25.4	3	2				

550	12	402	22.1	4	2				
551	12	601	19.3	2.6	2				
552	12	801	19	0.9	4				
553	12	1000	20	1.8	3				
554	12	1251	11.8	1.6	2				
555	12	1500	10.2	0.2	2				
556	12	2001	7.6	0.6	2				
557	12	2500	8.1	3	2				
558	12	3000	4.5	2.8	2				
559	12	3499	5.3	3.5	2				
560	12	4001	5.4	0.7	3				
561	12	4499	?32.3	4.6	2				
562	12	4501	4.1		1				
563	12	5001	6.1	0.9	3				
564	12	5473	6.8	1.3	2				
565									
566	Station	"Latitı	ude, Lo	ongitud	le"	"pres	sure, d	bar"	"Pb, pmol/kg"
567	s.d.	n							
568	14 "61°5	9.95'S,	40°05.	.98'E"	10	7.2	0.7	2	
569	14	26	8.1	2.4	4				
570	14	51	8.9	1.8	3				
571	14	99	6	0.2	2				
572	14	152	11.7	1	2				

573	14	201	12.6	0.9	2
574	14	399	7.3	2.3	2
575	14	599	5.2	1.5	2
576	14	1001	6.3	0.5	2
577	14	1250	5.9	0.8	2
578	14	1500	6.2	1.7	2
579	14	2002	3.8	1.2	2
580	14	2500	5.5	4.4	2
581	14	2999	6.7	0.2	2
582	14	3501	4.3	1.5	2
583	14	4001	6.1	1.1	2
584	14	4499	?32.4	3.2	3
585	14	5000	6.9	0.6	2
586	14	5271	6.2	1.7	3