Mercury (Hg) mineral evolution: A mineralogical record of supercontinent assembly, changing ocean geochemistry, and the emerging terrestrial biosphere

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ABSTRACT

Analyses of the temporal and geographic distribution of earliest recorded appearances of the 88 IMA-approved mercury minerals plus two potentially valid species exemplify principles of mineral evolution. Metacinnabar (HgS) and native Hg are the only two species reported from meteorites, specifically, the primitive H3 Tieschitz chondrite with an age of 4550 Ma. Since the first terrestrial appearance of cinnabar more than 3 billion years ago, mercury minerals have been present continuously at or near Earth's surface.

Mercury mineral evolution is characterized by episodic deposition and diversification, perhaps associated with the supercontinent cycle. We observe statistically significant increases in the number of reported Hg mineral localities and new Hg species at ~2.8–2.6, ~1.9–1.8, and ~0.43–0.25 Ga—intervals that correlate with episodes of presumed supercontinent assembly and associated orogenies of Kenorland (Superia), Columbia (Nuna), and Pangea, respectively. In contrast, few Hg deposits or new species of mercury minerals are reported from the intervals of supercontinent stability and breakup at ~2.5–1.9, ~1.8–1.2, and 1.1–0.8 Ga. The interval of Pangean supercontinent stability and breakup (~250–65 Ma) is also marked by a significant decline in reported mercury mineralization; however, rocks of the last 65 million years, during which Pangea has continued to diverge, is characterized by numerous ephemeral near-surface Hg deposits.

The period ~1.2–1.0 Ga, during the assembly of the Rodinian supercontinent, is an exception because of the absence of new Hg minerals or deposits from this period. Episodes of Hg mineralization reflect metamorphism of Hg-enriched marine black shales at zones of continental convergence. We suggest that Hg was effectively sequestered as insoluble nanoparticles of cinnabar (HgS) or tiemannite (HgSe) during the period of the sulfidic “intermediate ocean” (~1.85–0.85 Ga); consequently, few Hg deposits formed during the aggregation of Rodinia, whereas several deposits date from 800–600 Ma, a period that overlaps with the rifting and breakup of Rodinia.

Nearly all Hg mineral species (87 of 90 known), as well as all major economic Hg deposits, are known to occur in formations ≤400 million years old. This relatively recent diversification arises, in part, from the ephemeral nature of many Hg minerals. In addition, mercury mineralization is strongly enhanced by interactions with organic matter, so the relatively recent pulse of new Hg minerals may reflect the rise of a terrestrial biosphere at ~400 Ma.

Keywords: Ocean geochemistry, cinnabar, tiemannite, biosphere, supercontinent cycle, mercury (Hg) isotopes

INTRODUCTION

The evolution of the mineral kingdom is a topic that has engaged Earth scientists for more than two centuries, since debates raged between supporters of steady-state uniformitarianism and episodic catastrophism (Rudwick 1972; Greene 1982). Radiometric measurements of the extreme antiquity of some mineral specimens (Strutt 1910), coupled with recognition of the deterministic evolutionary sequence of igneous rocks and their minerals (Bowen 1915, 1928), placed the chronology of Earth's

changing near-surface mineralogy on a more quantitative footing. Subsequent elaborations of these concepts point to the central importance of time as a dimension in mineralogical research (Ronov et al. 1969; Zhabin 1981; Nash et al. 1981; Wenk and Bulakh 2004; Krivovichev 2010; Tkachev 2011).

“Mineral evolution,” the study of Earth's changing near-surface mineralogy through time, is an approach to Earth materials research that seeks to frame mineralogy in a historical context by focusing on a variety of Earth's near-surface characteristics, including mineral diversity; mineral associations; the relative abundances of mineral species; compositional ranges of their major, minor, and trace elements and isotopes; and grain sizes

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and morphologies (Hazen et al. 2008; Grew and Hazen 2009, 2010a; Hazen 2010; Hazen and Ferry 2010). In particular, temporal variations in mineral diversity have been shown to reflect tectonic, geochemical, and biological changes in Earth's near-surface environment (Hazen et al. 2009, 2011; Grew and Hazen 2010b; McMillan et al. 2010; Grew et al. 2011). The evolving mineral kingdom also displays many features common to other complex evolving systems, including diversification, punctuation, and extinction (Hazen and Eldredge 2010).

The minerals of mercury exemplify both the opportunities and challenges of the mineral evolution approach. The rare element Hg is present in Earth's upper, middle, and lower crust at concentrations of ~0.05, 0.0079, and 0.014 ppm, respectively (Rudnick and Gao 2004), and in the oceans at $<5 \times 10^{-7}$ ppm (Emsley 1991; Li and Schoonmaker 2004). In spite of this relative scarcity, there are 88 minerals approved by the Commission on New Minerals, Nomenclature and Classification (CNMNC) of the International Mineralogical Association (IMA), plus two minerals published but not yet approved by CNMNC, in which mercury is an essential or important constituent (Table 1). These species, which are tabulated in the International Mineralogical Association (IMA) database (<http://truff.info/ima>) as well as in the Mindat database (<http://mindat.org>), include native metals and intermetallic alloys, halides, sulfides, arsenides, selenides, antimonides, tellurides, sulfosalts, oxides, carbonates, and sulfates, and occur in various magmatic, hydrothermal, evaporitic, and surface weathering environments (Tunell 1968; White 1981; Barnes 1997; Parsons and Percival 2005a, 2005b). Domarev (1984) reviewed the temporal distribution of mercury ore deposits, but our contribution goes much further: its principal objective is to survey individual mercury minerals through time, with an emphasis on earliest appearances and temporal distributions of these diverse phases. Such an investigation of individual mineral localities holds the promise of revealing larger-scale geological and geochemical processes, including those associated with the evolving biosphere.

CRYSTAL CHEMISTRY OF MERCURY MINERALS

The crystal chemistry of the chalcophile element mercury is unique (Tunell 1968). Mercury cations in minerals are known to bond to oxygen, chalcogenides (S, As, Sb, Se, and Te), and halides (Cl, Br, and I), as well as with phosphate, sulfate, silicate, arsenate, carbonate, and other anionic species (Table 1). The diversity of mercury minerals, including their color, luster, state, habit, and associations, thus reflects the element's richly varied crystal chemistry (Figs. 1a–1d).

Mercury occurs in three common valence states: 0, 1+, and 2+. Divalent mercury, which occurs in II, IV, VI, and VIII coordination with effective ionic radii ranging from 0.83 to 1.28 Å, forms recognizable coordination polyhedra in a few minerals; for example, $\text{Hg}(\text{S}_2\text{X}_4)$ octahedra ($\text{X} = \text{Cl}, \text{Br}, \text{I}$) in corderoite $\{[\text{Hg}^{2+}]_3\text{Cl}_2\text{S}_2\}$ and other Hg halide-sulfides, and HgS_4 tetrahedra in metacinnabar and hypercinnabar (the wurtzite and sphalerite isomorphs of HgS , respectively). Hg^{2+} minerals also often contain linear S–Hg–S configurations, for example in cinnabar, which has infinite helical chains $(-\text{S}-\text{Hg}-\text{S}-)_{\infty}$.

In the monovalent state mercury's effective ionic radii for III and IV coordination are 1.11 and 1.33 Å, respectively. These val-

ues are comparable to those of Ag^+ (1.14 Å in IV coordination), though larger than for Cu^+ (0.74 Å in IV coordination). The electronegativities of Hg, Ag, and Cu are all 1.9; as a consequence of their crystal-chemical similarities, 29 of 90 recognized mercury minerals contain Ag and/or Cu, often in solid solution with Hg.

The coordination chemistry of minerals with Hg^{1+} typically involves cation clusters. The majority of the 21 known Hg^{1+} minerals and 8 known mixed Hg^{1+} – Hg^{2+} minerals contain $(-\text{Hg}-\text{Hg})^{2+}$ dimers with Hg–Hg distances 2.5 to 2.7 Å (Pervukhina et al. 1999a, 1999b). Each end of the mercury dumbbells in these structures is linked to one or two anions (O, Cl, Br, or I); for example, the principal structural motifs in calomel $\{\text{HgCl}\}$, or sometimes $[\text{Hg}^{1+}]_2\text{Cl}_2$ are linear Cl–Hg–Hg–Cl groups.

Larger cation clusters in mercury minerals include $[\text{Hg}_3]^{4+}$ triangular groups in kuznetsovite $\{[\text{Hg}^{1+}]_2[\text{Hg}^{2+}][(\text{AsO}_4)\text{Cl}]\}$ and Ag_3Hg tetrahedral clusters in tillmannsite $\{\text{Ag}_3[\text{Hg}^{1+}]\text{VO}_4\}$ (Sarp et al. 2003). Given the affinity of Hg to bond to other cations—a trait exemplified by the several natural mercury alloy and amalgam minerals—Borisov and coworkers (Borisov et al. 2005; Magarill et al. 2007) have identified larger structural units with anion-centered polyhedra in some mercury compounds. Oxygen centered Hg_6O tetrahedra, for example, occur as edge-sharing units in terlinguacreekite $\{[\text{Hg}^{2+}]_3\text{Cl}_2\text{O}_2\}$ and pinchite $\{[\text{Hg}^{2+}]_3\text{Cl}_2\text{O}_4\}$, thus making distinctive Hg_6O_2 clusters. Vasilyevite $\{[\text{Hg}^{1+}]_{20}[\text{O}_6\text{I}_3\text{Br}_2\text{Cl}(\text{CO}_3)]\}$, poyarkovite $\{[\text{Hg}^{1+}]_3\text{OCl}\}$, and aurivilliusite $\{[\text{Hg}^{1+}][\text{Hg}^{2+}]\text{OI}\}$ have Hg_6O_2 clusters linked by Hg_2 dumbbells in a framework arrangement, whereas in terlinguaite $\{[\text{Hg}^{1+}][\text{Hg}^{2+}]\text{OCl}\}$ the Hg_6O_2 clusters are linked by $[\text{Hg}_3]^{4+}$ triangles. Additional structural complexity is displayed by hanawaltite $\{[\text{Hg}^{1+}]_6[\text{Hg}^{2+}][\text{O}_3\text{Cl}_2]\}$, which has a framework of corner-linked individual Hg_4O tetrahedra, Hg_6O_2 clusters, and Hg–Hg dumbbells (Borisov et al. 2005). Note that as in other complex framework structures such as zeolites, these structures can also be described in terms of intersecting chains or rings. Thus, for example, hanawaltite can be characterized by infinite chains $[-\text{O}-(\text{Hg}-\text{Hg})^{2+}-\text{O}-\text{Hg}^{2+}-\text{O}-(\text{Hg}-\text{Hg})^{2+}-]_{\infty}$ (Pervukhina et al. 1999a), whereas edoylerite $\{[\text{Hg}^{2+}]_3(\text{CrO}_4)\text{S}_2\}$ and deansmithite $\{[\text{Hg}^{1+}]_2[\text{Hg}^{2+}]_3(\text{CrO}_4)\text{OS}_2\}$ can be described with interconnected networks Hg_4S_4 and Hg_6S_6 rings, respectively (Borisov et al. 2005). Given this rich crystal-chemical variety of Hg minerals, one objective of this study is to document possible trends in the temporal distribution of structural motifs, especially anionic clusters.

TYPES OF MERCURY MINERALIZATION

The principal geochemical mechanism for the concentration and precipitation of mercury minerals is hydrothermal reworking of marine black shales (White 1981; Barnes 1997). Marine black shales extending back to at least 2.5 Ga are typically enriched in Hg compared to other sedimentary rocks (Lehmann et al. 2004; Parsons and Percival 2005a; Sanei et al. 2012), probably as a consequence of the affinity of Hg for organic matter, notably through binding with organic thiols (Xia et al. 1999; Hesterberg et al. 2001; Haitzer et al. 2002; Rytuba 2005). Average Hg-values for black shales seem to be characteristic of different geologic eras: 40 ppb in the Paleozoic (543–251 Ma), 430 ppb in the Paleoproterozoic (2.5–1.6 Ga), and 150 ppb in the Archean (>2.5 Ga; Cameron and Jonasson 1972; Cameron and Garrels 1980;

TABLE 1. IMA recognized minerals of mercury (Hg) with mineral localities,* arranged chronologically by earliest known appearance†

Name	Formula	Select Localities (see Table 2 for key)	Occurrences	
			Oldest (Ma)	Youngest (Ma)
Cinnabar	HgS	>2000 known localities: AU02, AU04, AT02, AT04, AT05, BG01, CA01, CL01, CL02, CL03, CN01, CN05, CZ02, FR02, FR04, GE01, DE02, DE03, DE04, DE05, HU01, IR01, IR02, IE01, IT01, IT02, IT03, IT04, IT05, JP03, KG01, KG02, MK01, MX01, MX02, MX03, MX04, MA01, MA02, NA01, NZ01, PE01, RU01, RU02, RU03, RU07, RU10, SK01, SK02, SK03, SK04, SK05, SI01, ZA02, ZA03, ES01, ES02, SE01, CH01, US-AK01, US-AZ01, US-AZ02, US-AR01, US-CA01, US-CA02, US-CA03, US-CA04, US-CA05, US-CA06, US-CA07, US-CA09, US-ID01, US-NV01, US-NV02, US-NV03, US-NV04, US-NV05, US-NV06, US-NV07, US-NV08, US-NV09, US-TX01, US-UT01, UZ01, UZ02	3043	0
Mercury	Hg	>300 known localities: AT05, CL02, CZ02, DE02, DE03, DE05, HU01, IT01, IT02, IT03, IT05, KG01, KG02, MX01, MX02, MX04, RU01, RU02, SK02, SK04, SI01, ZA02, ES01, ES02, SE01, US-AZ02, US-AR01, US-CA02, US-CA03, US-CA05, US-CA06, US-CA07, US-CA08, US-CA09, US-CO01, US-NV02, US-NV04, US-NV07, US-NV08, US-NV09, US-TX01, US-UT01	2900	0
Hypercinnabar	HgS	KG02, ZA02, US-CA01, US-NV06	2900	0
Metacinnabar	HgS	>220 known localities: AT02, AT05, GE01, DE02, DE03, DE05, IR01, IT02, IT03, IT04, IT05, KG01, KG02, MK01, MX01, MX04, MA02, NA01, RU01, RU03, RU08, RU10, SI01, ZA02, ES02, US-AK01, US-AZ02, US-AR01, US-CA01, US-CA02, US-CA03, US-CA04, US-CA05, US-CA06, US-CA07, US-CA08, US-CA09, US-ID01, US-NV02, US-NV05, US-NV07, US-NV09, US-TX01, US-UT01, UZ01	2900	0
Eglestonite	[Hg ¹⁺] ₆ O(OH)Cl ₃	DE02, DE03, KG01, KG02, MX01, RU01, RU02, ZA02, US-AZ02, US-AR01, US-CA03, US-CA05, US-CA09, US-NV04, US-TX01	[2900]‡	0
Temagamite	Pd ₃ HgTe ₃	CA02, NO01, ZA04, US-MT01, US-WY01	2739	500
Potiarite	PdHg	AU03, AT01, BR01, BR03, GY01, JP01, NC01, RU04, RU06, RU09, ZA01, US-MT01, US-NV01	2716	34
Coloradoite	HgTe	~110 known localities: AU01, CA01, CA03, CL02, IR01, IT04, MX03, ZA04, CH01, US-CA10, US-CO01, US-NV05, ZW01	2704	0
Vaughanite	Tl[Hg ¹⁺]Sb ₂ S ₇	CA01	2638	2621
Aktashite§	Cu ₆ [Hg ²⁺] ₃ As ₂ S ₁₂	CA01, CN05, FR03, IR01, KG01, MX03, RU03, RU07, RU010, US-NV05	2638	18
Galkhaite	(Cs,Tl)(Hg,Cu,Zn) ₆ (As,Sb) ₄ S ₁₂	CA01, IR01, KG01, KG02, RU03, US-NV03, US-NV05	2638	14
Routhierite	TlCu[Hg ²⁺] ₂ As ₂ S ₆	CA01, FR03, MX01, RU07	2638	3.9
Tvalchrelidzeite	[Hg ²⁺] ₃ SbAs ₃	CA01, GE01, KG02, US-NV05	2638	0
Atheneite	Pd ₂ (As _{0.75} Hg _{0.25})	BR01, BR02, RU04, ZA01	2058	600
Tiemannite	HgSe	~60 known localities: AR01, AU03, BO01, CA04, CZ01, CZ03, CZ04, DE01, DE02, DE03, DE05, IT04, MX03, MX04, RU08, US-CA03, US-CA06, US-NV03, US-UT01, UZ01	1850	0
Eugenite	Ag ₁₁ Hg ₂	AT03, CL01, MA01, MA02, NA01, PL01, RU03, SE01, US-AZ01	1800	112
Paraschacherite	Ag _{1.2} Hg _{0.8}	DE02, MX03, RU01, RU03, SK05, SE01	1800	10
Schacherite	Ag _{1.1} Hg _{0.9}	CZ02, FR04, DE02, MX03, SK02, SK05, SE01	1800	10
Luanheite	Ag ₃ Hg	AR01, CL01, CN02, IT05, MX03, MA02, RU03, SK03, SE01	1800	2.6
Moschellandsbergite	Ag ₂ Hg ₃	AT05, CL03, CZ02, FR04, DE02, DE03, HU01, JP02, RU01, SE01, US-NV10	1800	1.8
Imiterite	Ag ₂ HgS ₂	AT04, HU01, IT05, MA02, US-CA10, US-MT02	563	0
Perroudite	5HgS·Ag ₄ Cl ₂	AU02, AU04, FR02, DE03, HU01, NA01, ES01, US-NV01	541	0
Balkanite	Cu ₃ Ag ₅ HgS ₈	AT03, AT05, BG01, IT05, US-NV06	520	45
Calomel	HgCl	~70 known localities: CL02, CZ01, DE02, DE03, IT01, IT03, KG01, KG02, MX01, MX03, MX04, RU01, RU02, ES02, US-AZ02, US-AR01, US-CA03, US-CA08, US-CA09, US-CO01, US-NV02, US-NV04, US-NV07, US-NV09, US-TX01	430	0
Schuetite	Hg ₃ O ₂ (SO ₄)	ES02, US-CA01, US-CA03, US-CA06, US-CA08, US-NV02, US-NV03, US-NV09	430	0
Petrovite	Cu ₃ HgPbBiSe ₅	BO01, CZ03, CZ04, MX03	416	18
Terlinguaite	[Hg ¹⁺][Hg ²⁺][OCl]	AT02, DE02, KG01, KG02, MX03, RU01, US-AR01, US-CA03, US-CA05, US-CA09, US-NV04, US-TX01	416	0
Weishanite	(Au,Ag) _{1.2} Hg _{0.8}	CN03	386	360
Gortdrumite	Cu ₁₈ FeHg ₆ S ₁₆	AT05, IE01, US-NV03	385	39
Leadamalgalam	Hg _{0.3} Pb _{0.7}	CN04, MX03	367	18
Arzakite§	[Hg ²⁺] ₃ [(Br,Cl) ₂ S ₂]	RU01	366	354
Grechishchevite	[Hg ²⁺] ₃ S ₂ BrCl _{0.5} I _{0.5}	RU01	366	354
Kadyrelite	[Hg ¹⁺] ₆ Br ₃ O _{1.5}	RU01	366	354
Lavrentievite	[Hg ²⁺] ₃ [Cl ₂ S ₂]	RU01	366	354
Kuznetsovite	[Hg ¹⁺] ₂ [Hg ²⁺][(AsO ₄)Cl]	KG01, KG02, RU01	366	267
Kuzminite	[Hg ¹⁺] ₂ (Br,Cl) ₂	DE02, RU01	366	248
Poyarkovite	[Hg ¹⁺] ₃ OCl	DE03, KG01, KG02, RU01	366	248
Corderoite	[Hg ²⁺] ₃ Cl ₂ S ₂	DE03, KG01, KG02, RU01, ES01, US-NV03, US-NV04, US-TX01	366	32
Montroydite	HgO	IT02, KG01, KG02, MX01, MX04, RU01, US-AR01, US-CA03, US-CA05, US-CA09, US-NV04, US-TX01	366	0
Artsmithite	[Hg ¹⁺] ₄ Al(PO ₄) _{1.74} (OH) _{0.26}	US-AR01	359	299
Livingstonite	HgSb ₄ S ₈	DE02, JP03, KG01, KG02, MX02, NZ01, US-AR01, US-NV07, US-NV09	359	0.01
Edgarbaileyite	[Hg ¹⁺] ₆ [Si ₂ O ₇]	US-AR01, US-CA03, US-NV09, US-TX01	359	0
Moschelite	[Hg ¹⁺] ₂ I ₂	DE02	354	248
Shakhovite	[Hg ¹⁺] ₄ SbO ₃ (OH) ₃	DE02, DE03, KG01, KG02, RU02	354	235
Kleinite	[Hg ²⁺] ₃ N(ClSO ₄) _n H ₂ O	DE02, US-NV04, US-TX01	354	15
Belendorffite	Cu ₇ Hg ₆	DE02, HU01	354	2.6
Capgaronnite	AgHgClS	AU02, FR02, DE03, HU01	354	0
Coccinite	[Hg ²⁺] ₂ I ₂	AU02, DE02	354	0
Hakite	Cu ₁₀ Hg ₂ Sb ₅ Se ₁₃	AR01, CN02, CZ01, CZ03, CZ04, DE01, MX03	349	2.6
Tischendorfite	Pd ₈ Hg ₃ Se ₉	DE05	296	289

(Continued on next page)

TABLE 1.—CONTINUED

Name	Formula	Select Localities (see Table 2 for key)	Occurrences	
			Oldest (Ma)	Youngest (Ma)
Chursinite	[Hg ¹⁺] ₃ [AsO ₄]	KG01, KG02	273	267
Velikite	Cu ₂ HgSnS ₄	KG01, KG02, US-AZ01	273	163
Gruzdevite	Cu ₆ [Hg ²⁺] ₃ Sb ₃ S ₁₂	KG02, MX03, US-NV03	273	18
Laffittite	Ag[Hg ²⁺]AsS ₃	FR03, KG02, US-NV05	273	18
Marrucciite	[Hg ²⁺] ₃ Pb ₁₆ Sb ₁₈ S ₄₆	IT04, SK04	260	8
Rouxelite	Cu ₂ HgPb ₂₂ Sb ₂₈ S ₆₄ (O,S) ₂	IT04, SK01	260	8
Christite	Tl[Hg ²⁺]AsS ₃	CN01, MK01, US-NV03, US-NV05	260	3.9
Tillmannsite	Ag ₃ [Hg ¹⁺]VO ₄	FR01	251	200
Iltisite	[Hg ²⁺]S·AgCl	FR02, HU01	251	2.6
Kelyanite	[Hg ¹⁺] ₁₂ (SbO ₆)BrCl ₂	RU02	250	235
Stalderite	TlCu(Zn,Fe,Hg ²⁺) ₂ As ₂ S ₆	CH01	245	241
Kolymite	Cu ₇ Hg ₆	CL01, HU01, RU01, RU05, US-NV10	161	2.6
Donharrisite	Ni ₈ Hg ₃ S ₉	AT05, US-CA03	144	0
Fettelite	Ag ₂₄ [Hg ¹⁺]As ₅ S ₂₀	CL03, DE04	114	0
Kenhsuite	[Hg ²⁺] ₃ Cl ₂ S ₂	ES01, US-NV04	88	15
Danielsite	(Cu,Ag) ₁₄ HgS ₈	AU04	65	<65
Magnolite	[Hg ¹⁺] ₂ TeO ₃	US-CO01	65	0
Polhemusite	(Zn,Hg)S	IR02, US-ID01, US-NV05	48	11
Comancheite	[Hg ²⁺] ₁₃ O ₉ (Cl,Br) ₈	US-TX01	38	32
Pinchite	[Hg ²⁺] ₃ Cl ₂ O ₄	US-TX01	38	32
Terlinguacreekite	[Hg ²⁺] ₃ Cl ₂ O ₂	US-NV04, US-TX01	38	15
Gianellaite	[Hg ²⁺] ₄ SO ₄ N ₂	US-CA03, US-TX01	38	0
Mosesite	{[Hg ²⁺] ₂ N}(Cl,SO ₄ ,MoO ₄ ,CO ₃)·H ₂ O	MX01, MX04, US-CA03, US-NV08, US-TX01	38	0
Mazzettiite	Ag ₃ [Hg ²⁺]PbSbTe ₃	US-CO02	28	23
Daliranite	Pb[Hg ²⁺]As ₂ S ₆	IR01	27	14
Grumiplucite	HgBi ₂ S ₄	IT03	27	8
Simonite	Tl[Hg ²⁺]As ₂ S ₆	IR01, MK01	27	3.9
Brodtkorbite	Cu ₂ HgSe ₂	AR01	23	2.6
Radtkite	[Hg ²⁺] ₃ [ClIS ₂]	US-NV04	16	15
Aurivilliusite	[Hg ¹⁺][Hg ²⁺]OI	US-CA03	5.3	0
Clearcreekite	[Hg ¹⁺] ₃ (OH)(CO ₃)·2H ₂ O	US-CA03	5.3	0
Deansmithite	[Hg ¹⁺] ₂ [Hg ²⁺] ₃ (CrO ₄)OS ₂	US-CA03	5.3	0
Edoylerite	[Hg ²⁺] ₃ (CrO ₄)S ₂	US-CA03	5.3	0
Hanawaltite	[Hg ¹⁺] ₆ [Hg ²⁺][O ₃ Cl ₂]	US-CA03	5.3	0
Peterbaylissite	[Hg ¹⁺] ₃ [(OH)(CO ₃)·2H ₂ O]	US-CA03	5.3	0
Szymańskiite	[Hg ¹⁺] ₁₆ Ni ₆ (CO ₃) ₁₂ (OH) ₁₂ (H ₂ O) ₆ ·3H ₂ O	US-CA03	5.3	0
Tedhadleyite	[Hg ¹⁺] ₁₀ [Hg ²⁺]O ₄ Cl ₂ (Cl,Br) ₂	US-CA03	5.3	0
Vasilyevite	[Hg ¹⁺] ₂₀ (O ₆ Br ₂ Cl)(CO ₃)	US-CA03	5.3	0
Wattersite	[Hg ¹⁺] ₄ [Hg ²⁺][(CrO ₄)O ₂]	US-CA03, US-CA09	5.3	0
Vrbaite	Tl ₄ [Hg ²⁺] ₃ Sb ₂ As ₈ S ₂₀	MK01	5.1	3.9

* Chemical formula from IMA database ruff.info/IMA/; locality data compiled from mindat.org.

† Age data compiled from the Mineral Evolution Database; see mindat.org for additional references.

‡ Eglestonite is a secondary halide mineral, likely younger than the age of primary mineralization listed here.

§ Aktashite and arzakite are inadequately described species not yet IMA approved.

Blum and Anbar 2010). However, the form of the Hg in black shales is not well established. It may be bound to organic matter, incorporated into pyrite, or present as a distinct Hg mineral such as cinnabar.

Hydrothermal reworking of organic-rich sedimentary rocks leads to Hg-enriched brines, which in turn form three distinctive types of Hg deposits (Rytuba 2005). The world's largest Hg deposit, the Almadén district of central Spain, is representative of concentrations that form when submarine mafic volcanism occurs adjacent to Hg-enriched marine sediments (Hernandez et al. 1999). Such deposits form near continental margins, where black shales are disrupted by volcanic activity.

New Almadén and New Idria in California, the largest Hg mining districts in North America, represent post-Miocene (<5.3 Ma) silica-carbonate deposits, a second common type of economically important Hg concentration. These bodies feature Hg mineralization in silica and carbonate that form during low-temperature hydrothermal alteration and replacement of serpentine (Bailey 1946; Eckel and Myers 1946). As in other types of Hg deposits, the Hg-, halide-, and hydrocarbon-rich fluids are derived from a nearby marine sedimentary basin.

Hot-springs-type mercury deposits, a third mode of Hg mineralization, are found associated with most Hg-rich regions. In these shallow to surface ore bodies Hg is concentrated by volcanically heated, often silicic, near-surface waters, which vapor-precipitate mercury minerals in the cooler near-surface environment (Cox and Singer 1986). Many such deposits are quite young and active today. For example, native mercury has been observed forming at ocean floor hydrothermal vents off the north shore of New Zealand's North Island (Stoffers et al. 1999).

Hydrothermal activity often complicates the dating of mercury minerals. For example, hydrothermal reworking of the mercury-hosting rocks of the Almadén district (primary mineralization 427–380 Ma) led to at least one additional pulse of hydrothermal Hg mineralization at 360 Ma (Hall et al. 1997). Thus no single date can be applied to Hg minerals from Almadén, as well as to many other mercury mineral localities.

It should be noted that commercial quantities of mercury are also obtained from many other mineralized zones that may lack separate Hg minerals, notably sedimentary exhalative deposits (SEDEX), volcanic-hosted epithermal (<200 °C) deposits, and volcanogenic massive sulfide (VMS) deposits that principally

concentrate other metals (e.g., White 1981; Barnes 1997). In these ore bodies, mercury commonly occurs as a trace or minor element in solid solution with zinc and copper sulfide ores, such as Hg-rich tetrahedrite $[(\text{Cu,Hg})_{12}\text{Sb}_4\text{S}_{13}]$; e.g., Dana 1958], as well as in amalgams with copper, lead, silver, and gold (e.g., Chen et al. 1985).

MERCURY MINERAL AGE AND LOCALITY DATA

Progress in mineral evolution depends on the availability of detailed information on valid mineral species and their localities of known ages and geologic settings (Hazen et al. 2011). Here we rely on data recently added to the Mineral Evolution Database, which is embedded in the mindat.org platform. According to mindat.org, more than 3000 separate “localities” host mercury minerals. However, many different localities may be associated with one larger contemporaneous mineralized region. Most notably, mindat.org records more than 300 separate mercury mines, prospects, dumps, placers, and other localized Hg mineral sites associated with the Pliocene to Recent (5.3 to 0 Ma) hydrothermal systems of west-central California. This important mining district, including the New Almadén mine in Santa Clara County and the New Idria mine in San Benito County (two of the world’s largest mercury producers), represents a single mineralized province with ages restricted to the last 5.3 million years (Bailey 1962; White 1981; Studemeister 1984; Varekamp and Buseck 1984; Barnes 1997; Smith et al. 2008). Similarly, hundreds of separate localities in southwest Alaska (Szumigala 1996), central Arizona (Eastoe et al. 1990), west Texas (Thompson 1954; Henry et al. 1997), and southwest Utah (Cunningham et al. 1982), as well as clusters of localities in many countries, represent individual mineralized districts.

Table 1 lists, in order of their oldest recorded occurrence, the 87 mineral species in which Hg is considered an essential element and approved by the IMA CNMNC (<http://ruff.info/>) and two Hg minerals not yet approved by CNMNC but probably valid, aktashite $[\text{Cu}_6[\text{Hg}^{2+}]_3\text{As}_4\text{S}_{12}]$ and arzakite $\{[\text{Hg}^{2+}]_3[(\text{Br,Cl})_2\text{S}_2]\}$, together with mineral locality information for each species. The ninetieth mineral in our list is atheneite $[\text{Pd}_2(\text{As}_{0.75}\text{Hg}_{0.25})]$, which is a valid mineral, but Hg may not be an essential constituent. Arsenic exceeds Hg at the only crystallographic site occupied by Hg (Bindi 2010) and the Hg-free analog has been synthesized (Schubert et al. 1963). Nonetheless, we have included atheneite as an Hg mineral because of the substantial Hg contents (14–16 wt%) at the type locality of Itabira, Minas Gerais, Brazil, and at Serra Pelada (Serra Leste) Au-(Pd-Pt) deposit, Pará, Brazil (Bindi 2010). However, we have not included the questionable mineral toconalite [supposedly $(\text{Ag,Hg})\text{I}$], originally reported from Chanarcilla, Chile, and later from Broken Hill, Australia (Mason 1972, Fleischer 1973); the mineral from Broken Hill is now suspected to be misidentified capgaronnite (AgHgClS ; Mason et al. 1992). We have documented 128 mercury mineral localities, including all known localities for 84 of the 90 known species and at least a dozen age and geographically representative localities for native mercury, calomel, coloradoite (HgTe), cinnabar, metacinnabar, and tiemannite (HgSe), each of which is known from numerous worldwide localities. Table 2 presents a key to the 128 mineral localities listed in Table 1, while Table 3 lists 127 Hg mineral localities in chronological order (the age

of the placer locality at Potaro River, Guyana, is indeterminate). These data are the basis for much of the subsequent analysis.

The most abundant mercury compound is HgS , which in nature occurs in three polymorphs: cinnabar, metacinnabar, and hypercinnabar. Cinnabar has been found at more than 2000 localities in at least 300 mineral districts of all ages from the Mesoarchean through Recent (Table 1) in 59 countries (mindat.org)—more than all other mercury minerals combined. Metacinnabar is widespread (~220 localities), but hypercinnabar is rare, only reported from four localities. The HgS polymorphs are also among the four or five oldest terrestrial Hg minerals. In the pure HgS system, cinnabar inverts to metacinnabar at $345 \pm 2^\circ\text{C}$, and metacinnabar to hypercinnabar, at $481 \pm 3^\circ\text{C}$ (Dickson and Tunell 1959; Potter and Barnes 1978). However, impurities such as Fe and Zn, as well as non-stoichiometry, lower the inversion temperatures (Barnes 1997), which explains the occurrence of the high-temperature polymorphs in terrestrial environments.

Obtaining reliable and unambiguous ages for many Hg mineral species is difficult. Some Hg mineral species, including Hg alloys, sulfides, tellurides, and sulfosalts, can occur as primary phases with massive habits associated with igneous activity (Figs. 2a and 2b) and can thus be dated with relative confidence as contemporaneous with the associated intrusion or volcanic events (Berman and Harcourt 1938; Cabri et al. 1973; Guillou et al. 1985; Nickel 1985; Harris et al. 1989; Hall et al. 1997). However, many Hg minerals arise from hydrothermal remobilization and deposition of mercury in epithermal zones (e.g., Foord et al. 1974; Johan et al. 1974, 1976; Leonard et al. 1978; Steed 1983; Kucha 1986; Orlandi et al. 1998, 2005, 2007). These Hg deposits must postdate their host lithologies, but ages of emplacement are not always provided in the literature. For example, the Hg mining district of Pike County, Arkansas, is hosted in Carboniferous sediments (359–299 Ma; Lowe 1985; Roberts et al. 2003a), but we have been unable to find a reliable age range for the subsequent Hg mineralization.

Several mercury minerals occur as a consequence of alteration, including recent surface weathering, of previous Hg species (Figs. 2c and 2d). For example, perroudite ($5\text{HgS} \cdot \text{Ag}_4\text{I}_2\text{Cl}_2$) and capgaronnite occur by alteration of Hg- and Ag-bearing tennantite ($\text{Cu}_{12}\text{As}_4\text{S}_{13}$) or tetrahedrite ($\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$) by halide-bearing solutions of marine origin (Sarp et al. 1987). Similarly, corderoite and kenhsuite (the α and γ forms of $\text{Hg}_3\text{S}_2\text{Cl}_2$, respectively), radkeite ($\text{Hg}_3\text{S}_2\text{ClI}$), eglestonite $\{[\text{Hg}^{1+}]_6\text{O}(\text{OH})\text{Cl}_3\}$, and many other Hg minerals occur as hydrothermal alteration products of cinnabar (Tunell et al. 1977; Vasil’eva and Lavrent’ev 1980; Roberts et al. 1981, 1990, 1993, 2003a, 2005; McCormack et al. 1991; McCormack and Dickson 1998; Pervukhina et al. 2008). For example, perroudite and danielsite $[(\text{Cu,Ag})_{14}\text{HgS}_8]$ occur in a supergene assemblage in gossan at Coppin Pool, Western Australia, hosted by sedimentary host rocks of the Fortescue Group (Nickel 1985, 1987; Sarp et al. 1987), which has been dated 2.765–2.697 Ga elsewhere in the Hamersley Basin (Arndt et al. 1991). The gossan is most likely due to deep weathering during the Cretaceous or Tertiary, when goethite deposits formed in the banded iron formations of the Hamersley basin near Mount Tom Price, 41 km from Coppin Pool (Taylor et al. 2001; Thorne et al. 2004).

Reports of capgaronnite, coccinite $\{[\text{Hg}^{2+}]\text{I}_2\}$, and perroudite

FIGURE 1. The diversity of mercury minerals, including their color, luster, state, habits, and associations, reflects various crystal-chemical environments of Hg. (a) Native mercury (Hg), RRUFF 070277, in massive and drusy quartz from the Socrates mine, Sonoma County, California; (b) cinnabar (HgS), RRUFF 070532, on calcite from Charcas, San Luis Potosi, Mexico; (c) imiterite (Ag_2HgS_2), RRUFF 080014, with calcite and dolomite from Imiter, Morocco; (d) silver var. amalgam (Ag,Hg), RRUFF 070463, with malachite and calcite, from the Tsumeb mine, Namibia.

in the famous deposits of Broken Hill, New South Wales, Australia, which are dated at 1.695–1.685 Ga (Frost et al. 2005; Page et al. 2005), provide a second example. It is unlikely that these minerals are this old, as they are found in kaolinite that resulted from the weathering of primary aluminosilicates (Sarp et al. 1987; Birch 1999). A possible source of iodine is seawater when Broken Hill was 50 km from the coast and salt spray was blown inland in the last 5 Ma (Sarp et al. 1987; Plimer 1999). Thus, the secondary Broken Hill Hg minerals are most likely no older than 5 Ma. Alternatively, Cl, Br, and I in the Hg halides could have originated from fluid inclusions in the Pb-Zn-Ag ores (Slack et al. 1993), and thus the age of the oxidation and of the Hg halides is less constrained, i.e., to between 65 and 5 Ma (Plimer 1984, 1999; Stevens 1986).

Yet another example is the reported occurrence of the common mercury chloride, calomel, with cinnabar and native Hg from deposits hosted by Precambrian metasediments in the Mazatzal Mountains, Sunflower District, Gila County, Arizona (Lausen 1926; Anthony et al. 1995). In this case, the source of Hg for the deposits has been inferred to be Tertiary volcanics

rather than a nearby Precambrian intrusion (Lausen 1926; Faick 1958). It is unlikely that the Hg minerals would be related to nearby volcanogenic massive sulfide deposits dated at 1.7 Ga (Eastoe et al. 1990).

Given these examples and their associated uncertainties, the ages for the earliest reported occurrence of 90 mercury minerals (Table 1) and for 127 mercury mineral localities (Table 3) indicate upper age limits based on reported occurrences, but actual ages for some mercury minerals may be significantly younger.

MERCURY MINERAL EVOLUTION

The temporal distribution of 127 mercury mineral localities (Table 3) and earliest reported occurrences of the 90 known Hg minerals (Table 1; Fig. 3) reveal episodes of increased Hg deposition separated by long intervals with relatively little Hg mineralization. A review of this punctuated history points to possible correlations between mercury mineralization and the evolution of Earth's near-surface environment, particularly in the context of the supercontinent cycle, as well as changes in ocean and atmosphere chemistry and the emergence of the ter-

TABLE 2. Mercury mineral locality register*

Location no.†	Country	Locality	References
AR01	Argentina	Tumiñico Mine, Sierra de Cacho, Sierra de Umango, La Rioja	Paar et al. (2002, 2005), de Brodtkorb (2009)
AU01	Australia	Kalgoorlie, Goldfields-Esperance Region, Western Australia	Kent and McDougall (1995)
AU02		Broken Hill, Yancowinna County, New South Wales	Birch (1999), Frost et al. (2005), Page et al. (2005)
AU03		Copper Hills, Pilbara Region, Western Australia	Bagas and Lubieniecki (2000), Nickel et al. (2002)
AU04		Coppin Pool, Western Australia	Nickel (1985), Sarp et al. (1987), Arndt et al. (1991)
AT01	Austria	Kraubath, Leoben, Styria	Malitch et al. (2001)
AT02		Geyer-Silberberg District, Inn Valley, North Tyrol	Matura and Summesberger (1980), Arlt and Diamond (1998)
AT03		Röhrebühel Mountain, Fieberbrunn, North Tyrol	Ebner (1998)
AT04		Ruden, Asten Valley, Goldberg Group, Hohe Tauren Mtns, Carinthia	Paar and Niedermayr (1998)
AT05		Schwarzle District, Saalfelden, Salzburg	Pohl and Belocky (1994)
BO01	Bolivia	El Dragón Mine, Potosí Department	Grundmann and Lehrberger (1990)
BR01	Brazil	Serra Pelada Deposit, Carajás mineral prov., Pará, North Region	Grainger et al. (2008)
BR02		Itabira, Iron Quadrangle, Minas Gerais	Cabral et al. (2002)
BR03		Serro, Minas Gerais	Richardson (1988), Cabral and Lehmann (2006)
BG01	Bulgaria	Sedmochislenitsi Mine, Balkan Mountains, Vratsa, Oblast	Atanassov and Kirov (1973)
CA01	Canada	Hemlo gold deposit, Marathon, Thunder Bay Dist., Ontario	Pan and Fleet (1995), Muir (2002), Davis and Lin (2003)
CA02		Copperfields Mine (Temagami Mine), Nipissing District, Ontario	Cabri et al. (1991), Bowins and Heaman (1991)
CA03		Robb-Montbray Mine, Rouyn-Noranda TE, Québec	Gibson and Galley (2007)
CA04		Shirley Peninsula (Fish Hook Bay area), Lake Athabasca, Saskatchewan	Cabri et al. 1991, O'Hanley et al. 1991, Rees 1992
CL01	Chile	Pabellón, Pampa Larga District, Copiapó Province	Marschik and Fontboté (2001), Kojima et al. (2007)
CL02		La Coipa Mine, Chañaral Province, Atacama Region	Oviedo et al. (1991)
CL03		Chañarcillo, Copiapó Province, Atacama Region	Sillitoe (2007)
CN01	China	Lanmuchang Tl-(Hg) Deposit, Xingren County, Guizhou Provi.	Zhang et al. (2000a, 2000b)
CN02		Luan River Valley, Chengde Prefecture, Hebei Province	Huang et al. (1996), Mao et al. (1999)
CN03		Weishancheng ore field, Nanyang Prefecture, Henan Province	Jiang et al. (2009)
CN04		Xiaonanshan Pt-Cu-Ni deposit, Wuchuan County, Inner Mongolia	Jiang et al. (2003)
CN05		Lianhecut Au-Hg-(As-Sb) deposit, Western Qinling Gold Belt, Sichuan Prov.	Xia et al. (2006)
CZ01	Czech Rep	Pedbořice, Central Bohemia Region	Křibek et al. (1999), Škácha et al. (2009)
CZ02		Radnice, Plzeň Region, Bohemia	Křibek et al. (1999), Škácha et al. (2009)
CZ03		Petrovice, Vysočina Region, Moravia	Křibek et al. (1999), Škácha et al. (2009)
CZ04		Rožná deposit, Vysočina Region, Moravia	Křibek et al. (2009)
FR01	France	Roua Mines, Alpes Maritimes, Provence-Alpes-Côtes d'Azur	Sarp et al. (1994), Sarp and Cerný (1999)
FR02		Cap Garrone Mine, Var, Provence-Alpes-Côtes d'Azur	Cathelineau et al. (1990)
FR03		Pelvoux Mtn, Hautes Alpes, Provence-Alpes-Côtes d'Azur	Johan and Mantienne (2000), Gasquet et al. (2010)
FR04		Allemont, Isère, Rhône Alpes	Feybesse et al. (2004)
GE01	Georgia	Gomi As-Sb-Hg deposit, Racha-Lochkhum-Kvemo Svaneti Region	Kekelia et al. (2008)
DE01	Germany	Alberoda, Schlema-Hartenstein District, Erzgebirge, Saxony	Förster and Haack (1995), Förster and Rhede (2002), Förster et al. (2005), Krupp (1984), Krupp et al. (1989)
DE02		Landsberg Mt., Obergoschel, Rhineland-Palatinate	Krupp (1984), Krupp et al. (1989)
DE03		Other Hg deposits, Rhineland-Palatinate	Krupp (1984), Krupp et al. (1989)
DE04		Glasberg Quarry, Odenwald, Hesse	Kissan et al. (1993), Pfaff et al. (2010)
DE05		Harz Mountains, Saxony-Anhalt	Möller et al. (1984), Baumann et al. (1991)
GY01	Guyana	Potaro River, Kangaruma District	Spencer (1928)
HU01	Hungary	Rudabánya, Borsod-Abaúj-Zemplén County	Fügedi et al. (2010)
IR01	Iran	Zareh Shuran Mine, Takab, West Azarbaijan Province	Mehrabi et al. (1999), Daliran (2008)
IR02		Agh-Darreh Mine, Takab (Takan Tepe), West Azarbaijan Province	Daliran (2008)
IE01	Ireland	Gortdrum Mine, Monard, County Tipperary	Duane et al. (1986), Duane (1988)
IT01	Italy	San Quirico, Gotra Valley, Albareto, Parma Province	Dini et al. (1995)
IT02		Amiata Mt., Grosseto Province, Tuscany	Bigazzi et al. (1981)
IT03		Levigliani Mine, Lucca Province, Tuscany	Dini et al. (1995, 2001)
IT04		Buca della Vena Mine, Lucca Province, Tuscany	Dini et al. (1995)
IT05		San Giovanni Mine, Gonnese, Carbonia-Iglesias Prov., Sardinia	Caron et al. (1997)
JP01	Japan	Inatsumiyama, Tottori Prefecture, Chugoku Region, Honshu Island	Watanabe et al. (1998)
JP02		Yamagano Mine, Kagoshima Prefecture, Kyushu Island	Watanabe (2005)
JP03		Matsuo Mine, Iwate Prefecture, Honshu Island	Imai (2004), Ohba et al. (2007)
KG01	Kyrgyzstan	Khaidarkan Sb-Hg Deposit, Osh Oblast	Pirajno et al. (2009), Dobretsov et al. (2010)
KG02		Chauvai Sb-Hg deposit, Alai Range, Osh Oblast	Pirajno et al. (2009), Dobretsov et al. (2010)
MK01	Macedonia	Allchar, Roszdan	Volkov et al. (2006)
MX01	Mexico	San Luis Mine, Hauhauxtla, Mun. de Taxco, Guerrero	Alaniz-Álvarez et al. (2002)
MX02		Huitzco de los Figueroa, Guerrero	Campa and Ramirez (1979), Camprubi et al. (2003), Moran-Zenteno et al. (2004)
MX03		Moctezuma, Mun. de Moctezuma, Sonora	Deen and Atkinson (1988), Camprubi et al. (2003)
MX04		El Doctor, Queretaro	Ferrari et al. (1999), Aguirre-Diaz and Lopez-Martinez (2001)
MA01	Morocco	Bou Azzer District, Tazenakht, Ouazarate Province	Gasquet et al. (2005), El Ghorfi et al. (2006), Oberthür et al. (2009)
MA02		Imiter Mine, Boumalne-Dadès, Ouazarate Province	Cheilletz et al. (2002)
NA01	Namibia	Tsumeb Mine, Otjikoto Region	Kamona et al. (1999), Haest and Muchez (2010)
NC01	New Caledonia	Ouen Island Ophiolite, Southern Province	Cluzel et al. (2001), Paquette and Cluzel (2007)
NZ01	New Zealand	Puhipuhi, Northland, North Island	Craw et al. (2000)
NO01	Norway	Kamøy, Rogaland	Pedersen and Hertogen (1990), Dunning and Pedersen (1988)
PE01	Peru	Huancavelica Department	Mckee et al. (1986)
PL01	Poland	Sierosowice Mine, Polkowice District, Lower Silesia	Kucha and Przybyłowicz (1999), Piestryński and Wodzicki (2000)
RU01	Russia	Kadyrel, Pii-Khem District, Tuva Republic, Eastern Siberian Region	Tretiakova et al. (2010)
RU02		Kelyana Hg deposit, Bount District, Eastern Siberian Region	Berger et al. (1978)
RU03		Privol'noye and Gal-khaya Mines, Saha Rep., Eastern Siberian Region	Parfenov et al. (1999), Stepanov and Moiseenko (2008)
RU04		Yoko-Dovyrensky Massif, Prebailkalia, Eastern Siberian Region	Kislov et al. (1989), Kislov (2005)

(Continued on next page)

TABLE 2.—CONTINUED

Location no.†	Country	Locality	References
RU05		Kolyma River Basin, Magadanskaya, Far-Eastern Region	Volkov et al. (2008)
RU06		Uktus Complex, Middle Urals	Krause et al. (2005)
RU07		Vorontsovskaya, Turjusk, Middle Urals	Sazonov et al. (1998, 2001)
RU08		Uchaly, Bashkortostan Republic, Southern Urals	Chernyshev et al. (2008)
RU09		Nurali Complex, Bashkortostan Republic, Southern Urals	Grieco et al. (2007)
RU010		Aktashskoye Sb-Hg deposit, Altai Republic, Western Siberian Region	Borisenko et al. (2003), Pavlova and Borisenko (2009)
SK01	Slovakia	Magurka, Partizánska Lupča, Liptovský Mikuláš Co., Žilina Region	Kohút and Stein (2005), Hurai et al. (2006)
SK02		Dobšiná Mining District, Rožňava County, Košice Region	Hurai et al. (2006, 2008)
SK03		Novoveská Huta U-Cu deposit, Košice Region	Rojkovič et al. (1993)
SK04		Gelnica Ore Belt, Gelnica County, Košice Region	Kohút and Stein (2005), Hurai et al. (2006)
SK05		Kremnica Mtns, Žiar nad Hronom Co., Banská Bystrica Region	Lexa (2005)
SI01	Slovenia	Idria Mine, Idria	Palinkaš et al. (2004)
ZA01	South Africa	Bushveld Complex, Limpopo Province	Melcher et al. (2005), Scoates and Friedman (2008), Olsson et al. (2010)
ZA02		Monarch Cinnabar Mine, Gravelotte, Murchison Range	Muff 1978, Davies et al. 1986, Boocock et al. 1988)
ZA03		Barberton District, Mpumalanga Province, Kaalrug Farm	Poujol et al. (1996), Schwarz-Schampera et al. (2010)
ZA04		Uitkomst Complex, Mpumalanga Province	Pearnton (1986), Cairncross (2004), Toulkeridis et al. (2010)
ES01	Spain	Bellota Ravine and El Hembrar, Castellón, Valencia	de Waal et al. (2001)
ES02		Almadén Mine, Ciudad Region, Castile-La Mancha	Tritilla and Solé (1999), Tritilla and Cardellach (2003)
SE01	Sweden	Sala Silver Mine, Sala, Västmanland	Hall et al. (1997)
CH01	Switzerland	Lengenbach Quarry, Imfeld, Wallis	Kieft et al. (1987), Zakrzewski and Burke (1987), Allen et al. (1996), (Erik Jonsson, University of Uppsala, personal communications)
US-AK01	U.S.A.	Aniak District, southwestern Alaska	Hoffmann and Knill (1996), Schroll (2005)
US-AZ01		Bisbee, Cochise County, Arizona	Szumigala (1996)
US-AZ02		Sunflower District, Gila County, Arizona	Lowell (1974)
US-AR01		Funderburk Prospect, Pike Co., Arkansas	Beckman and Kerns (1965), Eastoe et al. (1990)
US-CA01		Hg mines, Contra Costa County, California	Lowe (1985), Roberts et al. (2003a)
US-CA02		Patrick Creek District, Del Norte County, California	Bailey (1962), Studemeister (1984)
US-CA03		New Idria District, Fresno and San Benito Counties, Calif.	Bailey (1962), Studemeister (1984)
US-CA04		Chloride Cliff Mine, Inyo County, California	Bailey (1962), Studemeister (1984)
US-CA05		Parkfield District, Kings and Montgomery Counties, Calif.	Bailey (1962), Studemeister (1984)
US-CA06		East Mayacmas District, Lake County, California	Smith et al. (2008)
US-CA07		Adelaide District, San Luis Obispo County, California	Bailey (1962), Studemeister (1984)
US-CA08		Cambria-Oceanic District, San Luis Obispo, California	Bailey (1962), Studemeister (1984)
US-CA09		Emerald Lake Area, San Mateo County, California	Bailey (1962), Studemeister (1984)
US-CA10		Golden Rule Mine, Tuolumne County, California	Bailey (1962), Studemeister (1984)
US-CO01		Magnolia District, Boulder County, Colorado	Kelly and Goddard (1969)
US-CO02		Bonanza District, Saguache County, Colorado	Pride and Hasenohr (1983), Rose (2010)
US-ID01		Big Creek District, Valley County, Idaho	Leonard et al. (1978), Leonard and Marvin (1982), Criss et al. (1984)
US-MT01		Stillwater Complex, Stillwater County, Montana	DePaolo and Wasserburg (1979), Premo et al. (1990)
US-MT02		Warm Springs District, Fergus County, Montana	Marvin et al. (1980), Zhang and Spry (1994)
US-NV01		Goodsprings District, Clark County, Nevada	Church et al. (2005)
US-NV02		Ivanhoe District, Elko County, Nevada	Wallace (2003)
US-NV03		Elko, Lynn District, Eureka County, Nevada	Arehart et al. (2003)
US-NV04		McDermitt Mine, Opalite District, Humboldt County, Nevada	Noble et al. (1988)
US-NV05		Getchell Mine, Potosi District, Humboldt County, Nevada	Tretbar et al. (2000)
US-NV06		Manhattan District, Nye County, Nevada	Shawe et al. (1986, 2003)
US-NV07		Tybo District, Nye County, Nevada	Best et al. (1989), Best and Christiansen (1991), Sawyer et al. (1994)
US-NV08		Willard District, Pershing County, Nevada	Noble et al. (1987), Coolbaugh et al. (2005)
US-NV09		Antelope Springs District, Pershing County, Nevada	Noble et al. (1987), Coolbaugh et al. (2005)
US-NV10		Comstock Lode, Storey County, Nevada	Vikre et al. (1988)
US-TX01		Mariposa Mine, Terlingua District, Brewster County, Texas	Thompson (1954), Henry et al. (1997)
US-UT01		Marysville District (Marysville Uranium area), Piute County, Utah	Cunningham et al. (1982)
US-WY01		New Rambler District, Albany County, Wyoming	McCallum et al. (1976), Premo and Loucks (2000)
UZ01	Uzbekistan	Kul'dzhuk deposit, Central Kyzylkum Region, Kyzylkum Desert	Wilde et al. (2001)
UZ02		Muruntau Mine, Zarafshan, Central Kyzylkum Region, Kyzylkum Desert	Wilde et al. (2001)
ZW01	Zimbabwe	Commoner Mine, Kadoma District, Mashonaland West	Twemlow (1982)

* For additional details on localities, including lists of mineral species and additional references, see mindat.org.

† Locality abbreviations employ the two-letter scheme of the International Organization for Standardization (<http://www.iso.org/>).

restrial biosphere.

Data on the temporal distribution of mineral localities and species should be approached with caution in one important regard. As has been more fully explored by the paleontological community, even a relatively comprehensive database may suffer from distortions owing to collection bias (e.g., Alroy et al. 2008; Kiessling et al. 2010; Peters and Heim 2010; Alroy 2010). Evidence from the Mineral Evolution Database for pulses of mineralization, for the appearance or disappearance of mineral-forming processes, or even for presumed episodes of “mineral

extinction,” requires statistical treatments to tease out real events from noise (Sepkoski 1997; Bambach et al. 2004; Hazen et al. 2011). However, while we can point to statistically significant temporal episodes of mercury mineralization, we do not yet have a broad enough coverage of worldwide localities and ages to undertake a comprehensive analysis.

MERCURY IN METEORITES (~4.5 Ga)

Meteorites preserve the earliest stages (>4.5 Ga) of Earth's mineral evolution (Hazen et al. 2008; McCoy 2010). In spite of

TABLE 3. Mercury mineral locality register arranged chronologically*

Location no.	Country	Locality	Age range* (Ma)
ZA03	South Africa	Kaalrug Farm, Barberton District, Mpumalanga Province	3043
ZA02	South Africa	Monarch Cinnabar Mine, Gravelotte, Murchison Range	2900
CA02	Canada	Copperfields Mine (Temagami Mine), Nipissing District, Ontario	2739–2735
US-MT01	U.S.A.	Stillwater Complex, Stillwater County, Montana	2716–2693
CA03	Canada	Robb-Montbray Mine, Rouyn-Noranda, Québec	2704–2696
ZW01	Zimbabwe	Commoner Mine, Kadoma District, Mashonaland West	2700
AU01	Australia	Kalgoorlie, Goldfields-Esperance Region, Western Australia	2680–2600
CA01	Canada	Hemlo gold deposit, Marathon, Thunder bay District, Ontario	2638–2621
ZA01	South Africa	Bushveld Complex, Limpopo Province	2058
ZA04	South Africa	Uitkomst Complex, Mpumalanga Province	2052–2036
BR01	Brazil	Serra Pelada Deposit, Carajás province, Pará, North Region	1885–1879
CA04	Canada	Shirley Peninsula (Fish Hook Bay area), Lake Athabasca, Saskatchewan	1850
SE01	Sweden	Sala Silver Mine, Sala, Västmanland	1800
US-WY01	U.S.A.	New Rambler District, Albany County, Wyoming	1778–1750
AU03	Australia	Copper Hills, Pilbara Region, Western Australia	~800
BR02	Brazil	Itabira, Iron Quadrangle, Minas Gerais	800–600
RU04	Russia	Yoko-Dovyrensky Massif, Prebailkalia, Eastern Siberian Region	794–684
AT01	Austria	Kraubath, Leoben, Styria	780
BR03	Brazil	Serro, Minas Gerais	700–450
MA01	Morocco	Bou Azzer District, Tazenakht, Ouarzazate Province	600–550
MA02	Morocco	Imiter Mine, Boumalne-Dadès, Ouarzazate Province	563–544
NA01	Namibia	Tsumeb Mine, Otjikoto Region	541–519
IT05	Italy	San Giovanneddu Mine, Gonnese, Carbonia-Iglesias Province, Sardinia	520–465
NO01	Norway	Kamøy, Rogaland	500–465
RU09	Russia	Nurali Complex, Bashkortostan Republic, Southern Urals	472–397
AT03	Austria	Röhrerbühel Mountain, Fieberbrunn, North Tyrol	444–359
RU08	Russia	Uchaly, Bashkortostan Republic, Southern Urals	440–380
ES02	Spain	Almadén Mine, Ciudad Real Region, Castile-La Mancha	430–361
BO01	Bolivia	El Dragón Mine, Potosi Department	416–359
AT02	Austria	Geyer-Silberberg District, Inn Valley, North Tyrol	416–251
RU06	Russia	Uktus Complex, Middle Urals	400–328
CN03	China	Weishancheng ore field, Nanyang Prefecture, Henan Province	386–360
IE01	Ireland	Gortdrum Mine, Monard, County Tipperary	385–320
CN04	China	Xiaonanshan Pt-Cu-Ni deposit, Wuchuan County, Inner Mongolia	367
RU01	Russia	Kadyrel, Pii-Khem Dist, Tuva Rep., Eastern Siberian Region	365–354
US-NV01	U.S.A.	Goodsprings District, Clark County, Nevada	359–318
US-AR01	U.S.A.	Funderburk Prospect, Pike Co., Arkansas	359–299
DE02	Germany	Landsberg Mt., Obermoschel, Obermoschel, Rhineland-Palatinate	354–248
DE03	Germany	Other Hg deposits, Rhineland-Palatinate	354–248
CZ01	Czech Rep	Předbořice, Central Bohemia Region	348–150
CZ02	Czech Rep	Radnice, Plzeň Region, Bohemia	348–150
CZ03	Czech Rep	Petrovice, Vysočina Region, Moravia	348–150
CZ04	Czech Rep	Rožná deposit, Vysočina Region, Moravia	314–223
RU07	Russia	Vorontsovskova, Turjusk, Middle Urals	310–290
DE05	Germany	Harz Mountains, Saxony-Anhalt	296–289
UZ01	Uzbekistan	Kul'dzhuk deposit, Central Kyzylkum Region, Kyzylkum Desert	286–220
UZ02	Uzbekistan	Muruntau Mine, Zarafshan, Central Kyzylkum Region, Kyzylkum Desert	286–220
KG01	Kyrgyzstan	Khaidarkan Sb-Hg Deposit, Osh Oblast	273–267
KG02	Kyrgyzstan	Chauvai Sb-Hg deposit, Alai Range, Osh Oblast	273–267
CN01	China	Lanmuchang Tl-(Hg) Deposit, Xingren County, Guizhou Province	260–235
SK04	Slovakia	Gelnica Ore Belt, Košice Region	260–76
SK01	Slovakia	Magurka, Partizánska Lupča, Liptovský Mikuláš Co., Žilina Region	260–76
FR02	France	Cap Garrone Mine, Var, Provence-Alpes-Côtes d'Azur	251–245
FR01	France	Roua Mines, Alpes Maritimes, Provence-Alpes-Côtes d'Azur	251–200
FR03	France	Pelvoux Mtn, Hautes Alpes, Provence-Alpes-Côtes d'Azur	251–18
RU02	Russia	Kelyana Hg deposit, Bount District, Eastern Siberian Region	250–235
PL01	Poland	Sierosowice Mine	250–230
CH01	Switzerland	Lengenbach Quarry, Imfeld, Wallis	245–241
SI01	Slovenia	Idria Mine, Idria	245–235
BG01	Bulgaria	Sedmochislenitsi Mine, Balkan Mountains, Vratsa, Oblast	245–235
RU10	Russia	Aktashskoye Sb-Hg deposit, Altai Republic, Western Siberian Region	232–230
CN05	China	Lianhecun Au-Hg-(As-Sb) deposit, Western Quinling Gold Belt, Sichuan Prov.	232
RU03	Russia	Privol'noye and Gal-khaya Mine, Saha Republic, Eastern Siberia	202–145
DE01	Germany	Alberoda, Schlema-Hartenstein District, Erzgebirge, Saxony	190
US-AZ01	U.S.A.	Bisbee, Cochise County, Arizona	163
RU05	Russia	Kolyma River Basin, Magadanskaya, Far-Eastern Region	161–145
CN02	China	Luan River Valley, Chengde Prefecture, Hebei Province	152–131
AT05	Austria	Schwarzleo District, Saalfelden, Salzburg	144–65
CL01	Chile	Pabellón, Pampa Larga District, Copiapó Province	130–112
SK03	Slovakia	Novoveská Huta U-Cu deposit, Košice Region	125–105
SK02	Slovakia	Dobšiná Mining District, Rožňava County, Košice Region	120–76
CL03	Chile	Chañarcillo, Copiapó Province, Atacama Region	114–95
ES01	Spain	Bellota Ravine and El Hembrar, Castellón, Valencia	88–80
NC01	New Caledonia	Ouen Island Ophiolite, Southern Province	83–34
US-MT02	U.S.A.	Warm Springs District, Fergus County, Montana	74–54

(Continued on next page)

TABLE 3.—*CONTINUED*

Location no.	Country	Locality	Age range* (Ma)
US-AK01	U.S.A.	Aniak District, southwestern Alaska	72
JP01	Japan	Inatsumiyama, Tottori Prefecture, Chugoku Region, Honshu Island	69–59
AU02	Australia	Broken Hill, Yancowinna County, New South Wales	<65–5†
HU01	Hungary	Rudabánya, Borsod-Abaúj-Zemplén County	<65–2.6
DE04	Germany	Glasberg Quarry, Odenwald, Hesse	<65–2.6
US-AZ02	U.S.A.	Sunflower District, Gila County, Arizona	<65–2.6†
US-CO01	U.S.A.	Magnolia District, Boulder County, Colorado	<65–0
AU04	Australia	Coppin Pool, Western Australia	<65†
US-NV06	U.S.A.	Manhattan Districts, Nye County, Nevada	50–45
US-ID01	U.S.A.	Big Creek District, Valley County, Idaho	48–45
MX03	Mexico	Moctezuma, Mun. de Moctezuma, Sonora	48–18
US-NV05	U.S.A.	Getchell Mine, Potosi District, Humboldt County, Nevada	41–37
US-NV03	U.S.A.	Elko, Lynn District, Eureka County, Nevada	40–39
FR04	France	Allemont, Isère, Rhône Alpes	39–36
US-TX01	U.S.A.	Mariposa Mine, Terlingua District, Brewster County, Texas	38–32
MX02	Mexico	Huitzaco de los Figueroa, Guerrero	34–5.3
MX01	Mexico	San Luis Mine, Hauhauxtla, Mun. de Taxco, Guerrero	33–30
US-CO02	U.S.A.	Bonanza District, Saguache County, Colorado	28–23
IR01	Iran	Zareh Shuran Mine, Takab, West Azarbaijan Province	27–14
IT03	Italy	Levigliani Mine, Lucca Province, Tuscany	27–8
IT04	Italy	Buca della Vena Mine, Lucca Province, Tuscany	27–8
IT01	Italy	San Quirico, Gotra Valley, Albareto, Parma Province	27–8
AT04	Austria	Ruden, Asten Valley, Goldberg Group, Hohe Tauern Mtns, Carinthia	27
US-NV07	U.S.A.	Tybo District, Nye County, Nevada	27
US-UT01	U.S.A.	Marysville District, Piute County, Utah	23–15
AR01	Argentina	Tumiñico Mine, Sierra de Cacho, La Rioja	23–2.6
CL02	Chile	La Coipa Mine, Chañaral Province, Atacama Region	23–17
MX04	Mexico	El Doctor, Queretaro	17–0
US-NV04	U.S.A.	McDermitt Mine, Opalite District, Humboldt County, Nevada	16–15
IR02	Iran	Agh-Darreh Mine, Takab (Takan Tepe), West Azarbaijan Province	16–11
US-NV02	U.S.A.	Ivanhoe District, Elko County, Nevada	15
US-NV10	U.S.A.	Comstock Lode, Storey County, Nevada	14–13
SK05	Slovakia	Kremnica Mtns, Žiar nad Hronom Co., Banská Bystrica Region	12–10
PE01	Peru	Huancavelica Department	7–3
US-NV08	U.S.A.	Willard District, Pershing County, Nevada	6.4–5.8
US-NV09	U.S.A.	Antelope Springs District, Pershing County, Nevada	6.4–5.8
NZ01	New Zealand	Puhipuhi, Northland, North Island	5.3–0.01
US-CA01	U.S.A.	Hg mines, Contra Costa County, California	5.3–0
US-CA02	U.S.A.	Patrick Creek District, Del Norte County, California	5.3–0
US-CA03	U.S.A.	New Idria District, Fresno and San Benito Counties, California	5.3–0
US-CA04	U.S.A.	Chloride Cliff Mine, Inyo County, California	5.3–0
US-CA05	U.S.A.	Parkfield District, Kings and Montgomery Counties, California	5.3–0
US-CA07	U.S.A.	Adelaide District, San Luis Obispo County, California	5.3–0
US-CA08	U.S.A.	Cambria-Oceanic District, San Luis Obispo, California	5.3–0
US-CA09	U.S.A.	Emerald Lake Area, San Mateo County, California	5.3–0
US-CA10	U.S.A.	Golden Rule Mine, Tuolumne County, California	5.3–0
MK01	Macedonia	Allchar, Roszdan	5.1–3.9
GE01	Georgia	Gomi As-Sb-Hg deposit, Racha-Lochkumi-Kvemo Svaneti Region	5–0
US-CA06	U.S.A.	East Mayacmas District, Lake County, California	2.9–0
JP02	Japan	Yamagano Mine, Kagoshima Prefecture, Kyushu Island	1.96–1.8
JP03	Japan	Matsuo Mine, Iwate Prefecture, Honshu Island	1–0.1
IT02	Italy	Amiata Mt., Grosseto Province, Tuscany	0.29–0
GY01	Guyana	Potaro River, Kangaruma District	placert‡

* "Age range" records the range of ages reported for Hg mineralization. For example, if two studies report radiometric ages of 300 ± 10 and 290 ± 8 Ma, then we record 310–282 Ma as the age range. If a deposit is reported as from a certain time period, e.g. Pliocene, then we use the appropriate age range from the 2009 GSA Geologic Timescale.

† Host rocks at Coppin Pool and Broken Hill, Australia, and the Sunflower District, Arizona, are Precambrian, but Hg mineralization is Tertiary.

‡ Ages for placer mercury deposits are uncertain. The primary source of Hg minerals from the Potaro River may be associated with the Transamazonian Orogeny (~2 Ga).

the diversity of minerals found in meteorites (a number currently approaching 300 species, according to Rubin 1997a, 1997b; Brearley and Jones 1998; Papike 1998), until very recently the only Hg mineral reported in meteorites was HgS with no information as to which polymorph, e.g., in carbonaceous chondrites (CI, CV3) by Ulyanov (1991) and in Hg-rich chondrules in the Mighei CM chondrite by Lauretta et al. (1999), who suggested that HgS in Mighei resulted from aqueous alteration on the CM parent body. Caillet Komorowski et al. (2009, 2010) described HRTEM evidence for nanoscale native Hg and metacinnabar in the primitive H3 Tieschitz chondrite, and proposed formation of

these phases by cold accretion of previously condensed particles that survived due to lack of subsequent heating. These two are the oldest Hg minerals reported to date; at 4550 Ma, approximately 1500 Ma older than any recorded terrestrial occurrence.

Given the scarcity of discrete Hg phases in meteorites, an intriguing question—one applicable to most rare elements—is where does Hg reside? Mercury is highly volatile and metallic Hg has a high vapor pressure, so its condensation would have occurred at very low temperatures in the solar nebula. Mercury condensation into troilite requires temperatures below 300 K [e.g., Lodders (2003) calculated 252 K]. To explain the 50%

FIGURE 2. The massive habits of some primary mercury minerals, including (a) coloradoite (HgTe), RRUFF 070326, from the Herald mine, Sugarloaf, Boulder County, Colorado, U.S.A.; and (b) livingstonite (HgSb_4S_8), RRUFF 050453, from Huitzuc, Guerrero, Mexico; contrast with euhedral crystals of secondary alteration phases, including (c) montroydite (HgO), RRUFF 070235, on quartz from the Clear Creek Claim, southern San Benito County, California; and (d) kleinite $\{[\text{Hg}^{2+}]_2\text{N}(\text{Cl}, \text{SO}_4) \cdot n\text{H}_2\text{O}\}$, RRUFF 060179, from the McDermott mine, Humboldt County, Nevada.

FIGURE 3. Cumulative plot of the reported oldest occurrences in the Earth's near surface of 106 Be minerals (Grew and Hazen 2009, 2010a; Hazen et al. 2011; unpublished data) and 90 Hg minerals (this paper, Table 1). Both curves are based on literature searches. The plot is cumulative because each reported new appearance is added to the number of minerals that had been reported before this new appearance. The y-axis indicates the number of new minerals that are reported to have appeared by a certain time; it does not indicate the number of minerals forming at that time. Note the significant increases in number of different species for both Be and Hg at 2.8 to 2.5 Ga and 0.6 Ga to present, but major increases for Be minerals at 1.8–1.6 Ga and 1.2–1.0 Ga correspond to minimal or no increases in Hg minerals.

condensation temperature of 350 K for the Allende (CV) chondrite, Lauretta et al. (1999) performed calculations that ruled out Hg condensation in Fe-Ni metal, as Fe-Ni compounds, as HgO, or as one of the HgS polymorphs at reasonable condensation temperatures. They carried out other calculations that lent support to their suggestion that Hg may chemisorb onto Fe-Ni alloy surfaces at temperatures up to 515 K in CM chondrites, with later formation of HgS during aqueous alteration on the CM parent body, presumably at the nanoscale in most cases, but occasionally at the microscale as on Mighei (see above). Additionally, studies of thermal Hg release from the Murchison (CM) chondrite are consistent with Hg almost entirely in HgS, while similar measurements on the Allende (CV) chondrite suggest a mixture of HgS and Hg adsorbed on internal mineral surfaces, possibly silicate minerals (Lauretta et al. 2001).

Although the cosmochemical behavior of Hg has been studied since before 1960, questions regarding its distribution in extra-terrestrial materials have persisted (Lauretta et al. 2001). In a compilation of Hg abundances measured by neutron activation and wet chemistry, Lauretta et al. (1999) found that values scattered by over 3 orders of magnitude, even for samples of a single meteorite. Extreme values such as the 500 ppm Hg reported in the Orgueil CI chondrite by Ozerova et al. (1973) most likely resulted from laboratory contamination, but in other meteorites compositional heterogeneity is a possible explanation for the reported variation (Lauretta et al. 1999; Lodders 2003). Natural terrestrial contamination has been also suggested in one case, meteorite Yamamoto 82050 [a CO3 type chondrite (Kumar et al. 2001)]. Even though agreement seems to be converging on 0.35 ppm for the average CI chondritic Hg abundance (Lodders 2003, 2010), the question arises why this abundance is an order of magnitude greater than in Earth's crust or mantle? This difference points to significant and as yet incompletely explained Hg losses during Earth's accretion. Perhaps 50% was lost through volatilization, but much of the primordial Hg content represented by chondritic sources is unaccounted for (Lauretta et al. 1999, 2001).

MERCURY AND THE ARCHEAN SUPERCONTINENT CYCLE (~3.3–2.5 GA)

The terrestrial mineralogical record extends back at least 4.4 Ga (Cavosie et al. 2007; Harrison 2009; Papineau 2010). However, no Hg minerals have been reported in any terrestrial samples older than ~3.1 Ga. A survey of the first appearances and distribution of mercury minerals through time reveals several intriguing statistically significant trends, most notably a correlation between the appearance of new Hg mineral species and periods of supercontinent assembly (Table 4; Fig. 3). Specifically, the data in Table 3 may be fit to 5 Gaussian curves with the following means $\pm$ standard deviations: 2.69 ± 0.04 , 1.81 ± 0.05 , 0.53 ± 0.05 , 0.32 ± 0.07 , and 0.05 ± 0.05 Ga.

Varied evidence from geologic, geomagnetic, tectonic, and paleontological data point to a quasi-periodic cycle roughly 750 Ma in duration of assembly and dispersal of Earth's continents that has operated for at least the last 2.8 billion years, and may extend back >3.2 billion years (Gurnis 1988; Nance et al. 1988; Murphy and Nance 1992; Rogers and Santosh 2002, 2004, 2009; Zhao et al. 2002, 2004; Condie et al. 2009; de Kock et

al. 2009; Murphy et al. 2009; Santosh et al. 2009; Shirey and Richardson 2011). Three overlapping tectonic stages characterize the supercontinent cycle. First, during periods of continental aggregation, global tectonics is dominated by convergence, continental collision, and associated orogenic events. Many of modern Earth's largest mountain chains, including the Himalayas, the Alps, the Urals, and the Appalachians, arose during such periods of continental collision. Second, during periods of stable aggregation, supercontinents experience marginal subduction of oceanic crust and associated near-coastal acidic volcanism. Finally, because supercontinents act as "thermal lids," heat builds up mid-continent over periods of 10^8 years. Thus, continental rifting and the formation of new ocean basins characterize the breakup of supercontinents.

These three stages—assembly, stability, and breakup—commonly overlap, as all three modes of tectonic activity may occur simultaneously at different regions of the globe (as they do today), and it is difficult to define exact chronologies for each event in the supercontinent cycle. Rogers and Santosh (2004, 2009) used the concept of "maximum packing" of supercontinents for the situation when a single landmass includes the greatest amount of available continental lithosphere. Five episodes of supercontinent formation dating to ~2.8 billion years ago have been proposed, as well as possibly one or two intervals prior to 2.8 Ga named variously as Ur and/or Vaalbara (e.g., Rogers 1996; Cheney 1996; Wingate 1998; Rogers and Santosh 2002; Pesonen et al. 2003; Zhao et al. 2004; Bogdanova et al. 2009; Shirey and Richardson 2011). Moreover, there is disagreement on the nature of the early Paleozoic supercontinent; we have chosen to include Pannotia, whereas Gondwana is considered to have been a part of Pannotia and Pangea rather than a separate supercontinent (Table 4; Fig. 3; cf. Santosh et al. 2009). We emphasize that the duration of each stage listed in Table 4 is distilled from different papers that give a range of ages. Just as authors disagree on the detailed configurations of the supercontinents (except Pangea), they also disagree on the time intervals inferred for assembly,

TABLE 4. Chronological overview of the supercontinent cycle, mercury mineral localities, and first terrestrial appearances of Hg species

Supercontinent	Status	Interval (Ga)	Duration (Ma)	Number of Hg localities*	Number of new Hg mineralst
Ur/Vaalbara	Uncertain	>2.8		2	5
Kenorland	Assembly	2.8–2.5	300	6	8
	Stable	2.5–2.4	100	0	0
	Breakup	2.4–2.0	400	2	1
Columbia	Assembly	2.0–1.8	200	3	6
	Stable	1.8–1.6	200	1	0
	Breakup	1.6–1.2	400	0	0
Rodinia	Assembly	1.2–1.0	200	0	0
	Stable	1.0–0.75	250	4	0
	Breakup	0.75–0.6	150	2	0
Pannotia	Assembly	0.6–0.56	40	1	1
	Stable	0.56–0.54	20	1	1
	Breakup	0.54–0.43	110	6	3
Pangea	Assembly	0.43–0.25	180	29	35
	Stable	0.25–0.175	75	7	1
	Breakup	0.175–0.065	110	13	4
Cenozoic†		0.065–present	65	50	25

* See Table 3.

† See Table 1.

‡ The last 65 million years have been characterized by simultaneous continental rifting and convergence.

stability, and breakup of the supercontinents.

The five oldest reported Hg minerals are found in the Barberton and Murchison greenstone belts of the Kaapvaal craton, South Africa (Table 1). The oldest known terrestrial occurrence of cinnabar is a former mine on Kaalrug Farm, Mpumalanga Province, South Africa, in the Barberton belt (Pearnton 1986; Cairncross 2004). This cinnabar occurs in quartzite and vein quartz, and most plausibly formed during a hydrothermal event dated by Pb–Pb age of 3043 ± 59 Ma, which is related to an episode of extensive plutonism at about 3.1 Ga. (Toulkeridis et al. 2010) and roughly coeval with Au mineralization at ~ 3.1 Ga to the southwest in the Barberton belt (de Ronde et al. 1991, 1992; Kakegawa and Ohmoto 1999). This mineralization has been interpreted as related to extension tectonism that followed an extended history of accretion and convergence in the Barberton belt (de Ronde and de Wit 1994; Dirks et al. 2009).

Native mercury (Hg), hypercinnabar and metacinnabar (the two high-temperature polymorphs of cinnabar), and eglestonite are reported in the Monarch Cinnabar Mine, located a short distance south of the “antimony line” in the Murchison Range, Limpopo Province, South Africa (Pearnton 1986; Cairncross and Dixon 1995; Schwarz-Schampera et al. 2010). Livingstonite [HgSb_4S_7] was reported from the “antimony line” itself (Boese 1964; also in the list of Davis et al. 1986 and Boocock et al. 1988), but in a detailed study of these deposits, Muff (1978) did not find livingstonite, and cited Boese’s (1964) report as “identity not certain.” Consequently, we have not included livingstonite in our list of Mesoproterozoic Hg minerals. Pearnton (1986) reported that the epigenetic Hg mineralization at the Monarch Mercury Mine is of hydrothermal origin and is localized along a shear zone in schists that are interpreted to result from alteration of komatiitic rocks. Cinnabar is the most abundant ore; Pearnton’s (1986) isotropic unknown intergrown with cinnabar is probably metacinnabar. Cairncross and Dixon (1995) also list hypercinnabar and eglestonite from the Monarch Mine, the latter as a yellow powder associated with native Hg and cinnabar. As eglestonite is typically a secondary product of cinnabar (see above), it probably formed later than the HgS polymorphs, but possibly during the Archean epigenetic event. Poujol et al. (1996) reported a zircon U–Pb data age of 2900 Ma for a granite intrusion and deformation related to Sb–Au mineralization in the “antimony line.” This age provides the best constraint for the age of the epigenetic Hg minerals at the Monarch mercury and antimony mine; it is consistent with the 3020 ± 50 Ma Pb/Pb age reported as a possible maximum age for the mineralization (Saager and Köppel 1976).

Little is known about possible pre-2.8 Ga supercontinent assemblages (e.g., de Kock et al. 2009), so we are unable to relate Hg mineralization to these Archean tectonic events. Kenorland (also called Superia) is the oldest widely recognized supercontinent. Assembly (~ 2.8 –2.5 Ga) was accompanied by extensive hydrothermal activity and emplacement of volcanic massive sulfide mineralization (Barley et al. 2005). In the 100-million-year interval between about 2.74 and 2.64 Ga the number of mercury minerals more than doubled with a pulse of 8 new phases, mostly in deposits associated with greenstone belts and igneous complexes in the Superior and Wyoming provinces of North America and in the Yilgarn Craton, Western Australia,

e.g., temagamite (Pd_3HgTe_3) in the 2.739–2.735 Ga Copperfields Mine from the Nipissing District in Ontario, Canada (Cabri et al. 1973; Bowins and Heaman 1991) and coloradoite in the Abitibi greenstone belt near Kirkland Lake about 100 km to the north and similar in age (Ispolatov et al. 2008). An additional 5 mercury sulfide, sulfosalt, and telluride minerals are reported from the Archean Hemlo gold deposits at Marathon in the Thunder Bay District of Ontario, Canada (Pan and Fleet 1995; Muir 2002): aktashite, galkhaite $[(\text{Cs}, \text{Ti})(\text{Hg}, \text{Cu}, \text{Zn})_6(\text{As}, \text{Sb})_4\text{S}_{12}]$, routhierite $\{\text{TiCu}[\text{Hg}^{2+}]_2\text{As}_2\text{S}_6\}$, tvalchrelidzeite $\{[\text{Hg}^{2+}]_3\text{SbAsS}_3\}$, and vaughanite $\{\text{Ti}[\text{Hg}^{1+}]\text{Sb}_4\text{S}_7\}$. Pan and Fleet (1995) gave the age of Hg mineralization as 2643–2632 Ma during low-grade calc-silicate skarn alteration (see also Corfu and Muir 1989; Muir 2002), although it is possible that the 2681–2676 Ma zircon U–Pb age reported by Davis and Lin (2003) to bracket granite plutonism, gold mineralization, deformation, and metamorphism at Hemlo could also date the Hg minerals. The eighth new mineral, potarite (PdHg), as well as temagamite, are reported from the ~ 2.7 Ga Stillwater Igneous Complex, a layered intrusion exposed in southern Montana (DePaolo and Wasserburg 1979; Premo et al. 1990; Zientek et al. 1990). These North American localities are associated with craton convergence and the Algonian orogeny (also known as the Kenoran orogeny) during the assembly of the Kenorland supercontinent between ~ 2.8 and 2.5 Ga, a time characterized by a worldwide increase in igneous activity (e.g., Murphy and Nance 1992). Coloradoite is also reported to have formed at 2665 Ma at in the Golden Mile deposit, Kagoorlie in the Yilgarn craton, Western Australia (Shackleton et al. 2003). The pulse of 6 new Hg mineral localities during this interval may be fit with a Gaussian distribution (mean $\pm$ standard deviation = 2.69 ± 0.04 Ga; standard error of the mean = 0.017 Ga). We conclude that there was a marked diversification of Hg minerals associated with the assembly of Kenorland, well before the inferred stabilization of this supercontinent (Table 4; Fig. 3).

THE BREAKUP OF KENORLAND AND ASSEMBLY OF COLUMBIA (~ 2.5 –1.8 Ga)

The next 500 million years from 2.5 to 2.0 Ga, a time roughly correlated with the stable stage of the Equator-straddling Kenorland supercontinent and its subsequent breakup, is represented by the ~ 2.05 Ga Bushveld and Uitkomst Complexes in adjacent Limpopo and Mpumalanga Provinces, South Africa, respectively (de Waal et al. 2001; Scoates and Friedman 2008; Olsson et al. 2010). The Bushveld complex hosts two Hg species, potarite and atheneite (Cousins and Kinloch 1976; Kinloch 1982; Fleet et al. 2002; Melcher et al. 2005), of which atheneite is new. Jambor and Puziewicz (1989) suggested that an unnamed mineral having the composition Au_3Hg reported from the Sumidouro locality in Brazil dated at 2.14 Ga (Vial et al. 2007) could be weishanite $[(\text{Au}, \text{Ag})_{1.2}\text{Hg}_{0.8}]$, but also noted that Baptista and Baptista (1986), who described the Au_3Hg mineral in a museum sample from Sumidouri, had reported that no other mercury minerals are found in this deposit and that the mineral could have been an anthropogenic product of mining activity. Consequently, the evidence for weishanite having formed at 2.14 Ga is too tenuous to include, which leaves the period 2.5–2.0 Ga host to only one new Hg species.

Assembly of the next supercontinent, Columbia (also called

Nuna), commenced approximately 2.0 billion years ago, when five separate continents are thought to have converged into a single landmass. Each new suture resulted in an orogenic event associated with granitoid magmatism, continental crust formation, and hydrothermal mineralization (Condie et al. 2009). In the 50-million-year interval between ~1.85 and 1.80 Ga, 6 more Hg species occur for the first time (Table 1; Fig. 3). Four new Hg localities from this interval fit to a Gaussian peak (mean $\pm$ standard deviation = 1.81 ± 0.05 Ga; standard error of the mean = 0.024 Ga). The oldest reported occurrence of tiemannite is associated with pitchblende in a drill core from the Shirley Peninsula (Fish Hook Bay area), Lake Athabasca, Saskatchewan, Canada (Cabri et al. 1991); at 1850 Ma (Rees 1992; O'Hanley et al. 1991). Five new silver-mercury amalgams—eugenite ($\text{Ag}_{11}\text{Hg}_2$), luanheite (Ag_5Hg), moschellandsbergite (Ag_2Hg_3), paraschachnerite ($\text{Ag}_{12}\text{Hg}_{0.8}$), and schachnerite ($\text{Ag}_{11}\text{Hg}_{0.9}$)—are found at the Sala Silver mine, Västmanland, Sweden, where the age of Hg mineralization is estimated to be ~1.8 Ga (Allen et al. 1996; Erik Jonsson, personal communication). Additional evidence of mercury mineralization during this interval comes from the Serra Pelada gold deposit in Pará, North Region, Brazil (1.885–1.879 Ga; Grainger et al. 2008), where atheneite and potarite are found in association with gold, as well as numerous minerals of Cu, Ni, and the platinum group elements, and the 1.778–1.750 Ga New Rambler District of Wyoming, where temagamite has been found (Anthony et al. 1990; Premo and Loucks 2000).

Livingstonite has been reported from Broken Hill, Australia, where coccinite, capgaronnite, and perroudite are also recorded. Primary mineralization at Broken Hill has been dated at 1.695–1.685 Ga (Frost et al. 2005; Page et al. 2005). Note, however, that the latter three species likely represent alteration minerals of a much later age (see above), whereas livingstonite is a mineral whose occurrence at Broken Hill is “in doubt without further work,” because the method by which it had been identified was not specified (Birch 1999).

This significant ~1.90–1.80 Ga pulse of Hg mineral diversification on three continents is contemporaneous with the widespread orogenic activity related to the final assembly of Columbia (Rogers and Santosh 2002; Zhao et al. 2002, 2004). Paleomagnetic and geological reconstructions, which identify convergent margins between South America and West Africa, between Laurentia (central North America) and Baltica, between southern Africa and western Australia, and between Laurentia and Central Australia, suggest that the mercury mineral localities noted above are spatially and temporally close to presumed Columbian orogenic zones (Zhao et al. 2004).

RODINIA AND THE SULFIDIC INTERMEDIATE OCEAN (~1.8–0.75 GA)

Following the Paleoproterozoic pulse of mercury mineralization, the 1.2-billion-year period from 1.80 to 0.60 Ga, which roughly overlaps the time (~1.85–0.85 Ga) known variously as the “intermediate ocean,” the “Canfield ocean,” or the “boring billion” (Canfield 1998; Anbar and Knoll 2002; Poulton and Canfield 2011; Hazen 2012), saw a dearth of Hg mineral localities and the appearance of no new Hg species (Tables 1 and 3). This distinctive interval is marked by the termination of major banded iron formation deposition, and a redox stratified ocean,

characterized by an oxic near-surface but anoxic deep-ocean with widespread euxinic conditions, as well. Such sulfidic deep ocean conditions are known to have scavenged both Fe and Mo (Scott et al. 2008), thus affecting biological productivity. We argue that the same circumstance applied to Hg.

Note that Semenov et al. (1967) described the rare sulfosalt vrbaitte $\{\text{Ti}_4[\text{Hg}^{2+}]_3\text{Sb}_2\text{As}_8\text{S}_{20}\}$ as inclusions in chalcocite from the 1.175–1.123 Ga Mesoproterozoic Ilímaussaq complex of South Greenland, which is associated with continental rifting (Waight et al. 2002; Upton et al. 2003). However, the report has been questioned (Makovicky et al. 1980; Petersen 2001). The Ilímaussaq alkaline complex is famous for its enrichments in Zr, Nb REE, Be, and other rare elements, including Tl, but bulk Hg contents range only from 0.62 to 11.4 ppb depending on rock type (Bailey et al. 2001), well below the upper crust average of 50 ppb. Consequently, we have not included this report of vrbaitte in our cumulative plot (Fig. 3).

Rodinia was assembled between 1.2 and 1.0 Ga and lasted roughly 150–250 million years before breaking up between 750 and 600 Ma (Li et al. 2008; Bogdanova et al. 2009; Santosh et al. 2009). Unlike the previous two episodes of supercontinent assembly, no Hg minerals are recorded from this interval. Why is the period of Rodinian assembly different from that of Kenorland and Columbia, when apparent pulses of Hg mineralization are recorded? The paucity of Hg mineralization during the 1.2-billion-year interval from 1.8 to 0.6 Ga may be related to dramatic changes in ocean chemistry at that time. In modern times the oceans contain volatile biologically reduced Hg^0 and methyl Hg species, which are released into the atmosphere—a process that represents the largest single source of atmospheric mercury (Mason and Sheu 2002; Mason and Gill 2005). These species are oxidized in the atmosphere to Hg^{2+} species such as HgCl_2 , which are deposited back to the ocean surface where biological activity can reduce it and methylate it, after which it may be concentrated in ocean floor sediments and subject to remobilization.

Today's oceans contrast with those of the Mesoproterozoic. According to Canfield and coworkers (Canfield 1998; Canfield et al. 2000, 2007; Poulton et al. 2004; Poulton and Canfield 2011), the ocean 1.8 billion years ago was sulfidic (and possibly selenic, as well?)—an unprecedented state that may have scavenged atmospherically deposited mercury as insoluble nano-cinnabar and/or nano-tiemannite in the water column, after which it would be sequestered in ocean-floor sediments. The direct formation of tiemannite has been inferred in modern deep-sea cores in turbidites and sapropels (Mercone et al. 1999).

The relative stability of cinnabar at highly reducing surface conditions containing sulfides is evident in calculated f_{O_2} -pH diagrams in Figure 4. We calculated these diagrams with the aid of the software package Geochemists Workbench, using thermodynamic data from the literature as follows: cinnabar, tiemannite, and coloradoite (Mills 1974; Bethke 1996); montroydite (HgO) and calomel (Cox et al. 1989); aqueous Hg species (Shock et al. 1997). These diagrams suggest that in the Proterozoic ocean a particle carrying HgCl_2 from the atmosphere settling into a water column at 25 °C containing H_2S would result in the immediate precipitation of HgS , which should settle to the deep ocean floor as part of the sediment. This stability could account

FIGURE 4. Calculated f_{O_2} -pH diagrams illustrate relative stabilities of Hg minerals at surface conditions and at high temperatures. In **a**, the mineral calomel ($HgCl$) appears only because the activity of Hg^{2+} is unusually high. In **b–d**, the only stable Hg minerals are Hg (liquid) and cinnabar (HgS), and it can be seen that the stability field of cinnabar diminishes with increasing temperature. However, more complex Hg-S-As-Sb minerals, which may be stable at elevated temperatures, cannot be considered in the model because thermodynamic data are lacking.

for the anomalously high-Hg contents of the Paleoproterozoic shales compared with Archean or Paleozoic shales.

Even when the Paleoproterozoic cinnabar-bearing shales were subducted they may not have released their Hg to devolatilization fluids as readily as if mercury had been bound to organic matter or contained in pyrite. The stability of cinnabar at higher temperatures is illustrated in Figures 4c and 4d. At even higher temperatures during metamorphism it is likely that Hg could be incorporated into sulfosalts in the rock that could persist to at least 600 °C (Powell and Pattison 1997).

If so, then there may have been a long interval when mercury mobilization and the appearance of new Hg minerals was inhibited by cinnabar and tiemannite formation in marine black shales. In this scenario the availability of Hg would have increased at the end of the billion-year interval of the sulfidic intermediate ocean, with the oxygenation of successively deeper ocean layers (Canfield 1998; Scott et al. 2008). Under these circumstances deposition of particles carrying $HgCl_2$ from the atmosphere into the ocean would not have resulted in the immediate precipitation and removal of HgS , but instead Hg^{2+} could have been re-reduced and or methylated as in the modern Hg cycle. In this regard, it would be useful to determine how deeply rooted microbial mercury methylation pathways might be and, thus, the age when biological processes began to exert a significant influence on

the global Hg cycle.

In contrast to the period of Rodinia assembly, four occurrences of Hg minerals, including tiemannite, atheneite, and potarite, occur during the subsequent period of Rodinian stability from 1.0 to 0.75 Ga: Copper Hills, which is associated with the Camel-Tabletop Fault Zone of Western Australia (~800 Ma; Bagas and Lubieniecki 2000; Nickel 2002); Itabira, Minas Gerais, southeastern Brazil (800–600 Ma; Cabral et al. 2002; Cabral and Beaudoin 2006); the Yoko-Dovyrensky Massif of the Eastern Siberian region, Russia (794–684 Ma; Kislov 2005); and the Kraubath ultramafic body, Styria, Austria (~780 Ma; Malitch et al. 2001). These are the only localities for Hg minerals that our study has documented during the billion-year interval between 1.75 and 0.75 Ga.

RIFTING OF RODINIA AND THE SHORT-LIVED SUPERCONTINENT OF PANNOTIA (~750 TO 430 MA)

The global tectonic period from ~750 to ~430 Ma was complicated by regions of simultaneous continental rifting and convergence, including the breakup of the Rodinian supercontinent and the brief assembly and subsequent fragmentation of the partial supercontinents of Pannotia and Gondwana. The initial phase of Rodinia breakup at ~750 Ma generated three large landmasses—Proto-Laurasia and Proto-Gondwana separated by

the widening Proto-Tethys Ocean, and the smaller Congo Craton. Proto-Laurasia subsequently rifted into three continents, Laurentia, Siberia, and Baltica, separated by the Iapetus and Paleasian Oceans. Thus, by ~650 Ma, Earth's surface featured at least five major continents with three large intervening oceans.

In spite of the relatively chaotic global tectonic pattern between 750 and 430 Ma, the episodic temporal distribution of Hg mineralization and the first appearances of Hg minerals may once again reflect Earth's supercontinent cycle. Between 750 and 600 Ma, during the breakup of Rodinia, we record only one Hg mineral locality (and no new mercury mineral species). Mercury mineralization at that locality, the poorly dated 700–450 Ma Serro district of Minas Gerais, Brazil (Richardson 1988; Cabral and Lehmann 2006), may in fact postdate this interval of Rodinia's disaggregation.

From ~600 to 560 Ma several continents—portions of what are now Africa, India, the Middle East, and South America—converged to form the short-lived supercontinent Pannotia (also known as the Vendian supercontinent), which was situated primarily at both poles, with only a narrow strip of Equatorial land connecting the southern and northern landmasses (Pisarevsky et al. 2008). We document 6 localities approximating this interval in age; they yield a Gaussian peak (mean = 0.53 ± 0.05 Ga; standard error of the mean = 0.018 Ga). Within 60 million years, by the beginning of the Cambrian Period at ~540 Ma, Pannotia had begun to fragment into 4 main pieces: the Equatorial continent of Laurentia, the northern continents of Baltica and Siberia, and the southern supercontinent of Gondwana, which itself consolidated in a series of orogenies between ~550 and 500 Ma. The next 70 million years, from 500 to 430 saw continued rifting (e.g., Condie 1989; Merali and Skinner 2009). At ~480 Ma Avalonia split from Gondwana and moved northward toward Laurentia (now preserved along the coast in New England, the Canadian Maritimes, and Newfoundland, as well as in the British Isles).

Only 3 new Hg minerals appear between 600 and 430 Ma: imiterite (Ag_2HgS_2) from the 563–544 Ma Imiter mine, Ouarzazate Province, Morocco (Cheilletz et al. 2002); perrouditite from the 541–519 Ma Tsumeb mine, Otjikoto Region, Namibia; and balkanite ($\text{Cu}_9\text{Ag}_5\text{HgS}_8$) from the 520–465 Ma San Giovanni mine, Sardinia, Italy (Caron et al. 1997; Boni et al. 2000). The only other Hg mineral localities from this interval noted in our study (Table 3) are the Bou Azzer District of Morocco (600–550 Ma); Nurali Complex (572–397 Ma) and Uchaly (440–380 Ma), Southern Urals, Russia; Röhrerbühel Mountain, Tyrol, Austria (444–359 Ma); and the Rogaland district of Norway (500–465 Ma). Thus the 170 million year interval of Pannotia's assembly, stability, and breakup saw relatively little mercury mineralization.

PANGAEA (430–65 MA)

The dynamic subsequent 180-million-year period, between ~430 and ~250 Ma, was notable for the assembly of the well-documented supercontinent of Pangea through a series of continental collisions and subsequent orogenic events (Condie 1989), including the Caledonian, Guangxian, Variscan, Alleghanian, and Uralian orogenies. By ~430 Ma Baltica and Laurentia had collided, forming the minor supercontinent of Euramerica (or, equivalently, Laurussia) and initiating the northern Appalachian

Orogeny in the process. This mountain-building event can be considered the first step in the assembly of Pangea. The modest Avalonia landmass was accreted next to the East coast of Euramerica (~370 Ma) as the Iapetus Ocean between Euramerica and Gondwana continued to close.

The assembly of the bulk of Pangea occurred following the collision of Gondwana and Euramerica (~360–320 Ma), which also caused the Variscan (also termed the Hercynian) Orogeny (von Raumer et al. 2003). This extensive mountain-building event, which included the elevation of the Appalachians, was associated with a new pulse of Hg mineralization, for example in the Almadén district of Spain (Hall et al. 1997; Hernandez et al. 1999). The Pangean supercontinent continued to grow with accretion of smaller separate landmasses, including North China, South China, Kazakhstan, and Siberia; Pangean assembly was nearly completed by the end of the Pennsylvanian Period (~300 Ma), though associated convergent tectonics such as the Uralian and Cimmerian orogenies persisted into the late Permian (~250 Ma) and Jurassic Periods (~200 Ma), respectively. A total of 38 Hg mineral localities from this interval fit to a Gaussian peak (mean $\pm$ standard deviation = 0.32 ± 0.07 Ga; standard error of the mean = 0.011 Ga).

Almost 40% of known mercury minerals—35 of 90 species—appeared for the first time during this relatively brief interval of Pangea's assembly (~430–250 Ma; Tables 1 and 4). Of special note are the occurrence of 9 new species in the Pii-Khem District of Eastern Siberia, Russia (365–354 Ma; Tretiakova et al. 2010); 5 new species in the Landsberg Mountain district, Rhineland-Palatinate, Germany (354–248 Ma; Krupp 1984, 1989); and 4 new species in the Chauvai Sb-Hg deposit, Alai Range, Osh Oblast, Kyrgyzstan (273–267 Ma; Pirajno et al. 2009; Dobretsov et al. 2010). In addition, 26 other Hg localities of this age range occur in Asia, Europe, and North and South America. Note that most of these 29 localities are found in mid- to late-Paleozoic orogenic belts associated with the assembly of Pangea.

In sharp contrast to this period of extensive Hg deposits, we record a significant decrease in mercury mineralization during the periods of Pangean stability (~250–175 Ma) and rifting (~175–65 Ma)—a 185-million-year interval corresponding to the Mesozoic era that saw only 5 new mercury mineral species. This decline in mercury mineralization may be reflected in the data of Sanei et al. (2012), who document a significant increase in the Hg content of black shale deposited during the late Permian extinction (~250 Ma). This interval of Hg sequestration may represent a brief reprise of the billion-year Proterozoic gap in mercury deposits. In any event, the dramatic contrast between the Paleozoic time of Pangean assembly and its subsequent age of stability and breakup provides evidence for the important role of convergent tectonics on mercury mineralization.

THE CENOZOIC ERA (65 MA TO RECENT)

The last 65 million years have been a period of complex continental rearrangement, with simultaneous convergent and divergent margins. The Cenozoic Era has also been a time of unprecedented Hg mineralization. More than one quarter of all known mercury minerals (25 of 90 species) first appear in the last 65 million years (Tables 1 and 3). At least three factors contribute to this relative abundance. First, the rock record of the

Cenozoic Era is much more complete and well preserved than earlier eras. Many older mercury mineral localities, including shallow crustal and surface localities with earlier occurrences of many Hg minerals, must have been lost through erosion, subduction, and other winnowing processes.

Second, this history must in part reflect the ephemeral nature of Hg minerals, some of which are soluble in water, including many of the 27 known Hg halides (e.g., Parks and Nordstrom 1979), or gradually evaporate at STP (e.g., native mercury; Rytuba 2005). The significant vapor pressure of many mercury minerals may have played an important role in the distribution of Hg minerals through time (Rasmussen 1994; Zehner and Gustin 2002; Gustin 2003; Rasmussen et al. 2005). Given this volatility, near-surface Hg deposits may become significantly depleted and some Hg minerals may simply evaporate over geological timescales. Gustin (2003) cites several factors in the rate of evaporation, including the type of mineral species, exposure to sunlight, precipitation, and other weather-related parameters. While Hg-rich black shales and exhalations from volcanoes and Hg-enriched geothermal systems contribute more atmospheric Hg per unit area (Hinkley 2003; Gustin and Lindberg 2005), even relatively Hg-poor soils are major contributors to Earth's atmospheric mercury inventory because of their relatively large total area. The principal atmospheric Hg species (~95%) released from soils and rocks is monatomic elemental mercury, while HgCl_2 (also known as "reactive gaseous mercury," or RGM) accounts for most of the rest (Gustin 2003).

A third important factor in the relative abundance of new mercury minerals in the last 400 million years, as well as the distribution of major economic Hg deposits (all of which are ≤ 400 Ma in age; Table 5), is the rise of a terrestrial biosphere. Mercury is concentrated, and thus Hg mineralization is enhanced, by interaction with buried organic matter (Xia et al. 1999; Rytuba 2005). Thus the content of Hg in coal (0.1 ppm) and black shale (0.18 ppm), is an order of magnitude greater than in most other crustal lithologies, including sandstone (0.01 ppm), limestone (0.02 ppm), ocean ridge basalt (0.01 ppm), granite (0.03 ppm), and other sedimentary, igneous, and metamorphic rocks (Reimann and De Caritat 1998). The highest burial rates of organic carbon in the geologic record during the Phanerozoic Eon occurred from about 450 to 250 Ma (Berner and Canfield 1989), which corresponds to the dramatic increase in the number of Hg minerals and localities over the same time span (Fig. 5; Tables 1 and 3). Increased rates of organic burial since the rise of terrestrial biota in the Silurian Period have thus played a significant role in redistributing and concentrating Hg.

DISCUSSION

Mercury mineralization, the supercontinent cycle, and preservation of the mineralogical record

Correlations between the supercontinent cycle and mercury mineralization and the appearance of new mercury minerals are summarized in Table 4 and Figure 6. This phenomenon of episodic mineralizations, perhaps first articulated by Zhabin in 1979 [as translated in Zhabin (1981)], has been placed on a quantitative basis by several authors who have noted striking correlations between the supercontinent cycle and other types

of mineralization (e.g., Grew and Hazen 2009; Goldfarb et al. 2010; Tkachev 2011). Examples include minerals in igneous formations such as granitic pegmatites, alkaline complexes, and submarine volcanic exhalative deposits as manifest by the episodic age distribution of beryllium and boron minerals (Fig. 3; Grew and Hazen 2010a, 2010b), zircon crystals (Fig. 6; Hawkesworth et al. 2010; Condie and Aster 2010; Condie et al. 2011), and molybdenite (McMillan et al. 2010).

Episodic pulses of Hg mineralization reveal some similarities to this pattern (Table 4, Figs. 3 and 6) notably during the intervals that correlate with the aggregation of the supercontinents Kenorland, Columbia, and Pangea (~2.8–2.5, ~2.0–1.8, and ~0.43–0.25 Ga, respectively), and significant hiatuses during periods of supercontinent stability and breakup (~2.5–2.0, 1.8–1.2, 1.0–0.6, and 0.250–0.065 Ga), when few Hg deposits or new mercury mineral species appeared. Of the 60 Hg minerals that first appeared between 2.8 and 0.065 billion years ago, 50 (83% of species) formed during five intervals of supercontinent assembly totaling ~920 million years (34% of total interval; Tables 3 and 4). Similarly, 39 of 75 Hg deposits documented from this interval occurred during the relatively brief periods of continental aggregation.

The correlations evident in Figures 3 and 6 suggest that Hg mineralization follows periods of continental collision and orogeny, as tracked by supercontinent assembly. However, it is probable that destruction of older Hg deposits by geological activity has skewed the record so that most Hg mineralization appears to be associated with the last 430 Ma (99 of 127 localities; 65 of 90 species). Indeed, Goldfarb et al. (2010) noted that belts in Rodinia have been eroded down to high-grade metamorphic rocks, that is, to depths well below zones where most mercury deposits are formed.

An alternative (in our view more convincing) explanation for the sparsity of Hg deposits during the one billion year interval between ~1.8 and 0.8 Ga is that chalcophile Hg appears not to have been mobilized, perhaps owing to elevated oceanic sulfide levels (Canfield 1998; Anbar and Knoll 2002). This hypothesis is supported by data from the time of the late Permian extinction (~250 Ma)—a time characterized by a significant decrease in Hg mineralization (Fig. 6) correlated with an increase in the Hg sequestration in marine black shale (Sanei et al. 2012).

It is instructive to compare the mineralization history of Hg vs. Be (Fig. 3). Diversification of Hg minerals during the 1.8–0.8 Ga Proterozoic interval lagged behind diversification of Be minerals, which correlates strongly with assembly and stabilization of Columbia and Rodinia. This billion-year period contrasts with most of the last 430 million years, during which Hg mineral diversification accelerated to a greater extent than Be minerals, perhaps in part owing to the ephemeral nature of

TABLE 5. Principal mercury mining districts and their ages

Deposit	Age (Ma)	Reference
Almadén, Spain	430–361	Hall et al. (1997)
Idrija Mine, Slovenia	245–235	Palinkaš et al. (2004)
Amiata, Italy	0.30–0	Bigazzi et al. (1981)
Huancavelica, Peru	7–3	McKee et al. (1986)
New Almadén, California	5.3–0	Bailey (1962), Studemeister (1984)
New Idria, California	5.3–0	Bailey (1962), Studemeister (1984)
McDermitt, Nevada	16–15	Noble et al. (1988)

Note: The largest deposit is listed first.

FIGURE 5. The appearance of new Hg minerals over the past 400 Ma (a) reveals a pulse of mineralization that correlates with the Paleozoic increase in the burial of organic carbon, and consequent rise of atmospheric oxygen between ~370 and 250 Ma (b). The model of organic carbon burial through the Phanerozoic is based on relative abundances of sedimentary rocks (Bernier and Canfield 1989). This result is consistent with independently derived observations from carbon and sulfur isotopic studies.

many Hg phases, a phenomenon that could also explain the apparently accelerated diversification of B minerals, particularly evaporitic borates, in the Phanerozoic (Grew and Hazen 2010b; Grew et al. 2011).

Another difference between diversification of Hg and Be minerals is that 69% of the known Hg minerals (62 of 90 species) have been reported in rocks of Miocene age or younger (<23 Ma, Table 1) vs. 24% of Be minerals, many of which have been reported from only one locality worldwide (Grew and Hazen 2009, 2010a, unpublished data). In other words, current mineral diversity is closer to the cumulative diversity shown in Figure 3 for Hg than for Be. That a lower proportion of Be minerals than Hg minerals form very close to Earth's surface could also be a factor; this difference would also explain in part why proportionally fewer Hg minerals are preserved in older rocks.

FIGURE 6. A histogram of the number of new mercury minerals (top) and Hg mineral localities (middle) vs. time (50-million-year bins) reveals pulses of mercury mineralization that correlate with three periods of supercontinent assembly. Mineral locality data may be fit with five Gaussian peaks with means $\pm$ standard deviations as follows: 2.69 ± 0.04 , 1.81 ± 0.05 , 0.53 ± 0.05 , 0.32 ± 0.07 , and 0.05 ± 0.05 Ga. These episodes of Hg mineralization correlate with some, but not all, periods of increased zircon formation (bottom; data from Hawkesworth et al. 2010).

Mercury mineral evolution and the Great Oxidation Event

An important conclusion of previous mineral evolution studies is that a significant fraction of known minerals, perhaps exceeding two-thirds of the >4500 IMA approved species, are an indirect consequence of biological activity (Hazen et al. 2008, 2009; Sverjensky and Lee 2010). The principal cause of this biologically driven diversification is the Great Oxidation Event (GOE)—the rise of atmospheric oxygen after ~2.4 Ga owing to oxygenic photosynthesis.

Global oxidation affects Hg mineral formation in at least two important ways. The most obvious influence of atmospheric oxygenation after the GOE was creation of near-surface conditions where Hg oxides could form. The influence of atmospheric oxidation is thus reflected in the temporal distribution of mercury

minerals. With the exception of an occurrence of eglestonite that is probably a recent secondary weathering product of primary Archean cinnabar, all known Hg oxides and oxy-halides date from the last 430 million years.

Several lines of geochemical evidence suggest pervasive anoxic conditions prior to ~2.4 Ga. The presence of unweathered pebbles of siderite, uraninite, and pyrite in conglomerates (Rasmussen and Buick 1999; England et al. 2002; Hessler et al. 2004; Frimmel 2005), paleosol iron and cerium compositions (Holland and Rye 1997; Rye and Holland 1998; Murakami et al. 2001), mass-independent sulfur isotope anomalies (Farquhar et al. 2000, 2007, 2010; Papineau et al. 2007; Halevy et al. 2010), models of a ferruginous ocean (Holland 1984, 2002; Klein 2005), and reaction path calculations (Sverjensky and Lee 2010) point to an early Archean near-surface environment essentially devoid of molecular oxygen. Based on these data, the effective oxygen fugacity of the upper crust was thus buffered close to hematite-magnetite, with $\log f_{O_2} \sim -72$ at standard temperature and pressure (e.g., Hazen et al. 2009; Sverjensky et al. 2010; Sverjensky and Lee 2010). Purported hints of a “whiff of oxygen” at 2.5 Ga, based on the presumed mobilization by weathering of Mo and Re in the Mount McRae black shale of Western Australia (Anbar et al. 2007), are consistent with $\log f_{O_2} < -60$ (Sverjensky et al. 2010; Sverjensky and Lee 2010).

Under these circumstances the near-surface environment on the Archean Earth could easily have helped preserve early formed Hg minerals such as cinnabar, coloradoite, tiemannite, and potarite. Although thermodynamic data for these phases are limited in scope, there is sufficient experimental information to permit the calculation of aqueous activity diagrams at temperatures >200 °C for cinnabar, 200 °C for tiemannite, and at 25 °C for coloradoite. Several f_{O_2} -pH stability diagrams for cinnabar at different temperatures are given in Figure 4 and for tiemannite (HgSe), coloradoite, and montroydite in Figure 7. It can be seen in these figures that cinnabar, coloradoite, and tiemannite could all be stable at or near the relatively reducing Archean Earth's surface or at hydrothermal conditions. However, minerals such as montroydite and calomel would not be stable under these conditions.

We suggest that Earth's atmospheric oxygenation after the GOE is reflected in the temporal distribution of Hg minerals. Montroydite, terlinguaite, comancheite $\{[Hg^{2+}]_{13}O_9(Cl,Br)_8\}$, hanawaltite, and several other halide-oxides appear for the first time in the last 500 million years. It can be seen in the calculated $\log f_{O_2}$ -pH diagrams (Fig. 7a) that montroydite is only stable in a sulfur-free system with extremely high dissolved Hg concentrations (10^{-3} M). Furthermore, the montroydite stability field lies at $\log f_{O_2} > -20$, which is significantly greater than the maximum near-surface $\log f_{O_2}$ estimated prior to ~2.4 Ga. Therefore, we suggest that montroydite and other Hg^{2+} oxide minerals would not have been present prior to the GOE.

Changes in solubility and mobilization of Hg are a second important effect of near-surface oxidation. Mercury in the Hg^0 state is relatively soluble in reduced aqueous solutions with low sulfide content or in a liquid hydrocarbon phase (Krupp 1988). More oxidized ionic species of Hg form aqueous complexes with chloride and sulfate, as well as with organic thiols (Rytuba 2005).

A third potentially significant effect of rising f_{O_2} relates

FIGURE 7. Calculated f_{O_2} -pH diagrams illustrating the relative stabilities of montroydite (HgO), coloradoite (HgTe), and tiemannite (HgSe). (a) Montroydite appears only because the Hg activity is extremely high. In b, only one aqueous Te species and one Te mineral are considered (see text). In c and d, tiemannite stability is seen to be substantial over the temperature range 25 to 200 °C.

to changes in the near-surface sulfur (and to a lesser extent selenium) cycle. As in the production of atmospheric oxygen, microbial metabolism plays a key role in the S cycle, and thus in the coevolution of near-surface Hg mineralogy and the biosphere (see below).

Biological influences on mercury mineral evolution

A central thesis of mineral evolution is that Earth's near-surface mineralogy has coevolved with the biosphere for much of the past 3 billion years. A significant conclusion of this approach is that two-thirds of known mineral species are a consequence of the Great Oxidation Event at ~2.4 to 2.2 Ga, and thus these minerals are an indirect result of oxygenic photosynthesis (Sverjensky and Lee 2010). The minerals of mercury reflect this mineral diversification that occurred following the rise of atmospheric oxygen, as described above.

Many transition elements, including Fe, Ni, Mo, and Mn, are incorporated directly into essential enzymes and participate in biological reactions and metabolic pathways; life has thus significantly affected the geochemical cycling of these elements (e.g., Hazen et al. 2009). Mercury, by contrast, is not a biologically essential element; indeed, the element is highly toxic to many organisms (e.g., Fitzgerald and Lamborg 2004). Nevertheless, we speculate that biological influences may have played a significant role in mercury mineral evolution.

Microbial effects. While Hg is not an essential element in biological reactions, microbial communities are known to "process" environmental Hg through the production of methyl mercury (CH_3Hg^+) and dimethyl mercury $[(\text{CH}_3)_2\text{Hg}^0]$, which significantly affects the near-surface geochemical cycling of mercury (Compeau and Bartha 1985; Choi et al. 1994; Morel et al. 1998; King et al. 2000; Goulding et al. 2002; Gray et al. 2004; Krabbenhoft et al. 2005; Kritee et al. 2008, 2009). The timing of this microbial innovation of mercury methylation is as yet unknown, so we are unable to speculate on its possibly significant effects on the global mercury cycle and Hg mineral evolution.

Microbes also may have a significant effect on Hg mineralization through their metabolic byproducts. It is possible that local microbial production of H_2S raises f_{S_2} into the cinnabar stability field (Fig. 4), as suggested above for the Proterozoic. Nevertheless, sulfur isotope measurements point to a magmatic fluid source in some deposits (Lavric and Spangenberg 2003).

Effects of the terrestrial biosphere. Biology plays an important role in the Hg cycle by providing effective processes for mercury concentration and transport. Mercury has a strong affinity for organic matter, especially organic thiols, and it thus concentrates in black shales and coal (Krupp 1988; Hesterberg et al. 2001; Haitzer et al. 2002; Bergquist and Blum 2009). A particularly close link with Hg has been observed in petroleum and natural gas deposits (Peabody and Einaudi 1992; Manning and Gize 1993; Wilhelm 2001; Rytuba 2005); Hg^0 is soluble in, and thus transported by, liquid hydrocarbons (Krupp 1988). Both Hg and hydrocarbons are concentrated in black shales, and both are released through the action of hydrothermal activity.

The rise of the terrestrial biosphere over the past 500 million years, notably the diversification of the plant kingdom (Kenrick and Crane 1997; Beerling 2007), has greatly accelerated the production and deposition of organic carbon (e.g., Berner 2006).

The emergence of vascular land plants has also affected the global Hg cycle by extracting and concentrating soil Hg in leaves and subsequently releasing that Hg to the atmosphere during respiration or forest fires (Freidli et al. 2003; Rytuba 2005). As noted above, the highest burial rates of organic carbon in the geologic record during the Phanerozoic occurred from ~450 to ~250 Ma (Berner and Canfield 1989), which corresponds to the greatest increase in number of new Hg minerals (Fig. 5).

Anthropogenic effects. Finally, human activities have imposed significant changes in the near-surface mercury cycle (Mason et al. 1994; Fitzgerald and Lamborg 2004). Mercury has been mined since the Neolithic Age (~4000 BCE), with varied pre-industrial applications, including use of red cinnabar as a pigment, in medicine, and in gold and silver amalgamation, (Goldwater 1972; Parsons and Percival 2005b; Pacyna and Pacyna 2005). More recent technological applications include scientific instruments, such as barometers, thermometers, and vacuum pumps; amalgams in dentistry; insecticides, herbicides, fungicides, and bactericides; chemical processing, notably in the chlor-alkali industry; and a new generation of compact fluorescent light bulbs. Burning of Hg-enriched coal and petroleum adds to these anthropogenic sources (Wilhelm 2001; Finkelman 2003; Pacyna and Pacyna 2005). Collectively, the anthropogenic release of Hg into the atmosphere by near-surface exposure of mercury deposits, roasting of mercury ores, separation of gold and silver, and varied uses of mercury-bearing products has significantly increased the global atmospheric Hg concentration in the past several centuries (Gray et al. 2004; Hylander and Meili 2005; Rytuba 2005; Bergquist and Blum 2009).

While unambiguous biological influences have not been observed in mercury mineralization to the same extent as several other elements (i.e., uranium; Hazen et al. 2009), it is intriguing to speculate on the role of anthropogenic processes. For example, the secondary mineral schuetteite $[\text{Hg}_3\text{O}_2(\text{SO}_4)]$ is known only as a thin surficial coating on cinnabar exposed to sunlight in arid regions, most commonly in mine dumps, burnt ore, and bricks from old Hg furnaces (Bailey et al. 1959). Similarly, edoyleite (Erd et al. 1993), wattersite (Roberts et al. 1991), peterbaylissite (Roberts et al. 1995), hanawaltite (Roberts et al. 1996), clearcreekite (Roberts et al. 2001), tedhadleyite (Roberts et al. 2002), vasilyevite (Roberts et al. 2003b), and aurivilliusite (Roberts et al. 2004) are known only as secondary minerals and weathering products, spied by keen-eyed collectors in the historic mercury-rich dumps of mines in the New Idria district in California. It is possible that some of these ephemeral phases arise only when mercury-rich ores are exposed to the surface environment, and thus are effectively inadvertent anthropogenic minerals.

The crystal-chemical evolution of mercury

Given the rich crystal-chemical variety of Hg minerals, one objective of this study is to document the temporal distribution of structural motifs, especially anionic clusters. All of the earliest (age >600 Ma) unambiguously primary mercury minerals are either Hg metal and Ag-Hg alloys or chalcogenides (including sulfides, tellurides, arsenides, selenides, and antimonides). This limited crystal-chemical repertoire may in part reflect the diversity of stable Hg bonding environments; low concentrations of Hg can be accommodated as a trace or minor element in many

different minerals. This limited diversity in rocks older than 600 Ma also reflects the relative stability of these phases, for example compared to the numerous soluble halide species that appear only in the more recent record. However, the early appearance of these alloys and chalcogenides, which are characteristic of relatively low f_{O_2} , is consistent with patterns seen in the mineral evolution of other elements (Hazen et al. 2008; Sverjensky and Lee 2010).

Another obvious trend is the relatively late appearance of Hg oxides, oxy-halides, and other minerals with Hg-O bonds. The earliest recorded mineral with Hg-O bonds, eglestonite from the 2.9 Ga Monarch Mine of South Africa, is clearly of secondary origin and thus its age is not certain. Schuetteite occurs at the 430 Ma Almadén mine, Spain, while terlinguaite is reported from the 416 Ma Geyer-Silberberg District of Austria. The earliest recorded occurrence of mercury oxide, montroydite, is not until the 365 Ma Kadyrel Hg deposit of Eastern Siberia.

A similar relatively recent crystal-chemical innovation is the appearance of 7 species with mixed Hg^{1+} and Hg^{2+} : terlinguaite, hanawaltite, aurivilliusite, tedhadleyite, kuznetsovite, watersite, and deansmithite. These minerals, all of which are oxy-halides, arsenates, or chromates, are also reported from deposits no older than 365 million years. Among the most recently formed Hg minerals are the 4 known (possibly ephemeral) mercury carbonates—clearcreekite, peterbaylissite, symanskiite, and vasilyevite (Roberts et al. 1990, 1995, 2001, 2003b), and the 3 known mercury chromates—deansmithite, edoylerite, and watersite (Roberts et al. 1991, 1993; Erd et al. 1993), all of which are found in the Pliocene to Recent (<5.3 Ma) deposits of the New Idria District, California. The late appearance of these and other relatively recent exotic Hg minerals (Table 1) may point to a combination of idiosyncratic geochemical conditions and limited stability ranges.

Mercury isotopes

Mercury is unusual in having 7 stable isotopes— Hg^{196} , Hg^{198} , Hg^{199} , Hg^{200} , Hg^{201} , Hg^{202} , and Hg^{204} , spanning a relative mass difference of 4%. Consequently, mercury isotope systematics, both mass-dependent and mass-independent fractionations, hold great promise for tracking the element's geochemical transformations (Bergquist and Blum 2009). Accordingly, several recent investigations of the dynamic contemporary mercury atmospheric and biogeochemical cycles (Bergquist and Blum 2007; Ghosh et al. 2008; Kritee et al. 2008, 2009; Carignan et al. 2009; Point et al. 2011), though relatively few studies examine Hg isotopes in a mineralogical context (see, however, Hintelmann and Lu 2003; Smith et al. 2005, 2008; Blum and Anbar 2010; Dahl et al. 2010).

Mercury isotopes may prove particularly revealing of the paragenesis and timing of mercury mineralization because Hg compounds readily undergo near-surface phase transformations. Many of the most common mercury minerals, including cinnabar, coloradoite, and tiemannite, are easily altered to a host of secondary minerals—changes that might be reflected in isotope systematics. Thus, for example, the relative ages of presumably primary cinnabar and secondary eglestonite from the 2.9 Ga Monarch Mine, Murchison Range, South Africa, might be established through such an investigation.

The volatilities of many Hg minerals and the significant atmospheric Hg contributions of mining districts (Gustin 2003;

Mason and Sheu 2002; Mason and Gill 2005), suggest that near-surface mercury deposits must experience a significant isotopic evolution—systematic changes that might reveal historical aspects of complex ore deposits. Additional insights might be obtained from possible mass-independent Hg isotope fractionation, for example caused by the selective photolysis of Hg compounds such as $HgCl_2$ in the upper atmosphere. It is plausible, for example, that Hg isotopes in minerals display temporal trends analogous to those of sulfur, whose mass-independent isotope fractionation reveal important atmospheric changes associated with the Great Oxidation Event (e.g., Farquhar et al. 2000, 2007, 2010; Papineau et al. 2007; Halevy et al. 2010).

Additional analytical richness might be provided by the most abundant anions in ancient mercury minerals. The multiple isotope species of tellurium (6 stable isotopes), selenium (6 stable isotopes), and sulfur (4 stable isotopes), point to opportunities in the investigation of complexly clumped isotopes in some of the commonest Hg minerals. Potentially revealing studies might be to examine marine black shales, as well as cinnabar-, coloradoite-, and tiemannite-bearing ores, through 3 billion years of Earth history.

A note on iodide and bromide minerals

The minerals of mercury bear an intriguing relationship to those of the halogens: iodine and bromine (Table 6). Of the 13 IMA approved iodide minerals, 8 contain essential mercury. Similarly, of 10 approved bromine minerals (all bromides), 7 contain essential mercury. With the exception of the rare fumarolic sulfide minerals demicheleite-(Br) ($BiSBr$) and mutnovskite (Pb_2AsS_3I), all other known iodide and bromide minerals lacking mercury contain monovalent silver and/or copper. Mercury and bromine are also closely tied in Earth's atmosphere, where reactive halogens are known to oxidize Hg^0 (Seigneur and Lohman 2008; Holmes 2010; Obrist et al. 2011).

This close affinity of Br^- and I^- for Hg (as well as for Ag^+ and Cu^+) in minerals in part reflects similar concentration mechanisms in hydrothermal fluids derived from organic-rich marine black shales (e.g., Barnes 1997). Indeed, the hydrothermal processing of marine black shale is a recurrent feature of continental collisions during supercontinent accretion and likely explains the close temporal association of Hg minerals and supercontinent assembly.

What is perhaps more intriguing is the apparent complete absence of iodide and bromide minerals in natural alkali or alkaline earth halides. A probable explanation lies in brine compositions. Even in the most I- and Br-enriched brines, chlorine contents greatly exceed that of other halogens: $Cl/I > 1000$ and $Cl/Br > 10000$ (Barnes 1997). Therefore, iodine and bromine enter alkali and alkaline earth chlorides as a minor element in solid solution rather than form their own phases.

An important application of mineral evolution is in the comparative mineralogy of different terrestrial planets and moons, which may advance to different stages of mineral evolution and might also diverge. A frequently asked question is whether there exist any minerals on Mars not found on Earth. We speculate that the anhydrous, acidic, evaporitic near-surface environment of Mars (Squyres et al. 2004) may display a range of halides, including bromides and iodides, not found in near-surface ter-

TABLE 6. IMA approved iodide (I^{-1}) and bromide (Br^{-1}) minerals*

Name	Formula
Halides	
Marshallite	CuI
Miersite	(Ag,Cu)I
Cupro-iodargyrite	(Ag,Cu)I
Iodargyrite	AgI
Bromargyrite	AgBr
Kuzminite	$[Hg^{+1}]_2(Br,Cl)_2$
Coccinite	$[Hg^{+1}]_2I_2$
Moschelite	$[Hg^{+1}]_2I_2$
Halide-Sulfides	
Demicheleite-(Br)	BiSBr
Mutnovskite	Pb_2As_3I
Radtkite	$[Hg^{+1}]_3[ClI_2S_2]$
Grechishchevite	$[Hg^{+1}]_3S_2BrCl_{0.5}I_{0.5}$
Perroudite	$5HgSAg_4I_2Cl_2$
Halide-Oxides	
Aurivilliusite	$[Hg^{+1}][Hg^{+2}]OI$
Comancheite	$[Hg^{+1}]_{13}O_3(Cl,Br)_8$
Kadyrelite	$[Hg^{+1}]_6Br_3O_{1.5}$
Kelyanite	$[Hg^{+1}]_{12}(SbO_6)BrCl_2$
Tedhadelite	$[Hg^{+1}]_{10}[Hg^{+2}]O_4I_2(Cl,Br)_2$
Vasilyevite	$[Hg^{+1}]_{20}[O_6I_3Br_2Cl(CO_3)]$
Barlowite	$Cu_4BrF(OH)_6$

* Not including iodine or iodates [I^{5+} minerals with $(IO_3)^-$ groups], none of which contains Hg.

restrial formations. By contrast, in the absence of extensive hydrothermal processing of the martian crust, it seems unlikely that any mercury minerals will have formed on the red planet.

FUTURE RESEARCH OPPORTUNITIES

The central theme of mineral evolution is that Earth's near-surface mineralogy changes through time as a result of varied geological and biological processes. In the case of mercury, geotectonic events, notably hydrothermal activity associated with orogenic collisional boundaries during supercontinent assembly, appear to play a dominant role, with a clear overprint of the rise of the terrestrial biosphere and changes in near-surface oxidation since the Great Oxidation Event. An important missing piece of this story—one that we have not yet attempted—is to correlate specific Hg mineral localities with their tectonic settings at the time of mineralization. Such paleotectonic analysis represents an important target for future research.

These results also suggest several other future studies on variations of mercury minerals through time. While the first appearances of Hg minerals do not appear to be particularly sensitive to biological influences, their trace and minor element chemistries might be. For example, cinnabar is known to incorporate Se, Tl, Ag, Fe, Sb, Co, and Cd (Chen et al. 1982; Zhang 1996; Kidera et al. 2001; Hampton et al. 2004). Temporal variations of those and other trace elements might thus reveal other aspects of Earth's changing near-surface geochemical environment.

The strong correlation between mercury mineralization and collisional orogens suggests that it will be interesting to explore the behavior of other chemical elements to see if other stages of the quasi-periodic supercontinent cycle promote mineralization. For example, once formed, stable supercontinents no longer experience extensive orogenies, but they are constantly subjected to marginal subduction and associated acidic volcanism. Supercontinent breakup, on the other hand, leads to seafloor spreading with basaltic volcanism and passive continental

margins. These processes may lead to additional episodic mineralization events.

This study employed the Mineral Evolution Database, which facilitates studies of the changing diversity, distribution, associations, and characteristics of individual minerals as well as mineral groups through time. The results of this study thus underscore the potential of the MED to reveal important geophysical, geochemical, and biological events in Earth history. It is worth noting that this study was completed entirely by collating and analyzing data available in previous publications. At a time when funding for mineralogical research is highly competitive and advanced analytical facilities may not be available to all researchers, it is important to recognize that significant mineralogical insights may be awaiting discovery solely through the extensive resources of a good Earth science library and the internet.

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