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3	On the paragenetic modes of minerals:
4	A mineral evolution perspective
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10	ABSTRACT
11	A systematic survey of 57 different paragenetic modes distributed among 5659 mineral
12	species reveals patterns in the diversity and distribution of minerals related to their
13	evolving formational environments. The earliest minerals in stellar, nebular, asteroid, and
14	primitive Earth contexts were dominated by relatively abundant chemical elements,
15	notably H, C, O, Mg, Al, Si, S, Ca, Ti, Cr, and Fe. Significant mineral diversification
16	subsequently occurred via two main processes, first through gradual selection and
17	concentration of rarer elements by fluid-rock interactions (for example, in hydrothermal
18	metal deposits, complex granite pegmatites, and agpaitic rocks), and then through near-
19	surface biologically-mediated oxidation and weathering.
20	We find that 3349 mineral species (59.2 %) are known from only one paragenetic
21	context, whereas another 1372 species (24.2 %) are associated with two paragenetic
22	modes. Among the most genetically varied minerals are pyrite, albite, hornblende,
23	corundum, magnetite, calcite, hematite, rutile, and baryte, each with 15 or more known
24	modes of formation.
25	Among the most common paragenetic modes of minerals are near-surface

26 weathering/oxidation (1998 species), subsurface hydrothermal deposition (859 species), 27 and condensation at volcanic fumaroles (459 species). In addition, many species are 28 associated with compositionally extreme environments of highly differentiated igneous 29 lithologies, including agaitic rocks (726 species), complex granite pegmatites (564 30 species), and carbonatites and related carbonate-bearing magmas (291 species). 31 Biological processes lead to at least 2707 mineral species, primarily as a consequence of 32 oxidative weathering but also through coal-related and other taphonomic minerals (597 33 species), as well as anthropogenic minerals, for example as byproducts of mining (603 34 minerals). However, contrary to previous estimates, we find that only  $\sim 34\%$  of mineral 35 species form *exclusively* as a consequence of biological processes. By far the most 36 significant factor in enhancing Earth's mineral diversity has been its dynamic 37 hydrological cycle. At least 4583 minerals - 81 % of all species - arise through water-38 rock interactions.

A timeline for mineral-forming events suggests that much of Earth's mineral diversity was established within the first 250 million years. If life is rare in the universe, then this view of a mineralogically diverse early Earth provides many more plausible reactive pathways over a longer timespan than previous models. If, however, life is a cosmic imperative that emerges on any mineral- and water-rich world, then these findings support the hypothesis that life on Earth developed rapidly in the early stages of planetary evolution.

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- 48 **Keywords:** philosophy of mineralogy; classification; mineral evolution; natural kinds;
- 49 meteorite mineralogy; thermal metamorphism; aqueous alteration; biomineralization

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### INTRODUCTION

51 The minerals that form Earth arose from a variety of physical, chemical, and 52 biological processes over a time span now known to exceed 7 billion years (Hazen et al. 53 2008; Heck et al. 2020). Identifying and systematizing these mineral-forming processes is 54 key to understanding planetary evolution through deep time. Accordingly, we have 55 conducted a comprehensive survey of the modes of formation (i.e., "paragenetic modes" 56 - see below) of 5659 mineral species approved by the International Mineralogical 57 Association's Commission on New Minerals, Nomenclature and Classification (IMA-58 CNMNC). The underlying motivation for this effort is to understand how the diversity 59 and distribution of minerals have changed through deep time and to propose a system of 60 mineral classification complementary to IMA-CNMNC protocols that reflects mineral 61 origins in the context of evolving terrestrial worlds.

62 Hazen, Morrison, and colleagues have introduced an "evolutionary system of 63 mineralogy," which classifies "historical natural kinds" (Boyd 1991, 1999; Hawley and 64 Bird 2011; Magnus 2012; Khalidi 2013; Ereshevsky 2014; Godman 2019; Cleland et al. 65 2021) based on modes of mineral formation, as manifested in their distinctive 66 combinations of physical and chemical attributes (Hazen 2019; Hazen and Morrison 67 2020, 2021; Morrison and Hazen 2020, 2021; Hazen et al. 2021). In this effort, we adopt 68 the principle that any alternative system of mineral classification should complement and 69 amplify IMA-CNMNC protocols (Hatert et al. 2021; Hazen 2021), as they are based on 70 the rigorous time- and origin-independent attributes of idealized chemical compositions 71 and crystal structures (e.g., Burke 2006; Mills et al. 2009; Schertl et al. 2018; Cleland et 72 al. 2021).

The alternative evolutionary approach to mineral classification relies on identifying diagnostic suites of mineral properties and the inferred historical processes by which those distinctive properties arose. Accordingly, it is important to enumerate the variety of mineral paragenetic modes, as well as the temporal context of those diverse mechanisms of formation. The principal objective of this contribution is to identify and catalog the paragenetic modes of all known minerals and to attempt to place those mineral-forming mechanisms in the chronological context of Earth's 4.567-billion-year evolution.

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## What is a paragenetic mode?

82 The term "paragenesis" has been applied in different ways to minerals and their 83 formation. Paragenesis is often used in the context of a specific mineral assemblage: "A 84 characteristic association or occurrence of minerals or a mineral assemblage, especially in ore deposits, connoting a specific equilibrium assemblage or contemporaneous 85 86 formation" (https://mindat.org, accessed 26 June 2021). "Paragenesis" or "paragenetic 87 sequence" is also commonly used to mean the sequence of mineral formation in a suite of 88 rocks, such as a regional metamorphic terrain with a range of temperature-pressure 89 conditions or an ore deposit with stages of mineralization (e.g., Craig and Vaughan 1994; 90 Guilbert and Park 2007). And, in a broader context, paragenesis "may be applied to any 91 sequence or assemblage of minerals related by common formation conditions" (Ebel and 92 Kamilli 2018).

Our definition of "paragenetic mode" is most closely related to the latter sense of the term. We define a "mineral paragenetic mode" as *a natural process* by which a collection of atoms in solid and/or fluid form are reconfigured into one or more new solid forms. 96 This definition emphasizes an individual mineral's mode of formation rather than a 97 mineral assemblage or a sequence of mineralization. Consequently, paragenetic modes 98 encompass a wide variety of mineral formation processes, including the formation of 99 primary phases by condensation from a gas; the crystallization of minerals through 100 evaporation of an aqueous solution; the transformation of preexisting phases by lightning, 101 impacts, or other sudden events; and biomineralization.

In its broadest sense, our definition of paragenetic mode applies to processes that form a range of solids that are not usually incorporated in IMA-CNMNC protocols, including volcanic glass, kerogen, amber, and other amorphous and/or biologically-formed phases. Note that sharp boundaries do not always exist between natural crystalline compounds and other solid phases; for example, we are still grappling with how to incorporate biological crystal/organic composite materials of the Phanerozoic Eon, such as bones, teeth, and shells.

109 Our definition of paragenetic modes implicitly requires knowledge of three aspects of110 the mineral-forming process.

111 (1) The initial state of the system: Minerals form from precursor chemical systems at a 112 wide range of initial pressure-temperature-composition (P-T-X) conditions. It is 113 often useful to define minerals that emerge through condensation or precipitation 114 from a gas, liquid, or supercritical fluid as "primary minerals," whereas phases that 115 arise through modification of preexisting minerals (the majority of species) are 116 called "secondary minerals." Note, however, that intermediate formation processes may occur and the distinction between primary and secondary is not always 117 118 unambiguous (e.g., Hazen and Morrison 2021).

(2) <u>The mechanism of transformation:</u> Mineral-forming mechanisms range from rapid
energetic events, such as bolide impacts and lightning strikes with durations on the
order of seconds or less, to much more gradual processes, such as solid-state
reactions that occur during regional metamorphism or fluid-rock interactions
associated with metasomatism. These mechanisms also operate at diverse spatial
scales from micrometers (e.g., microbial biomineralization) to hundreds of
kilometers (regional metamorphism).

(3) <u>The resulting solid phases:</u> Each combination of initial state and transformation
 mechanism yields one or more mineral phases.

128 These three characteristics of the paragenesis of every mineral - initial state, 129 transformation mechanism, and final state – are reflected in typical descriptions of 130 mineral occurrences, notably in the Handbook of Mineralogy (Anthony et al. 1990-2003) 131 and Rock-Forming Minerals (Deer et al. 1982-2013). For example, periclase (MgO) is "a 132 relatively high temperature mineral, resulting from the metamorphism of dolomites and 133 magnesian limestones" (Bowles et al. 2011), whereas malachite [Cu<sub>2</sub>(CO<sub>3</sub>)(OH)<sub>2</sub>] is "a 134 common secondary mineral formed in the oxidation zone of copper deposits" (Anthony et 135 al 2003).

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# 137 Uncertainties related to the classification and timing of paragenetic modes

In attempting to classify the paragenetic modes of minerals, we have adopted fourgeneral principles:

- Every mineral species approved by the IMA-CNMNC must be associated with at
   least one paragenetic mode (e.g., the copper arsenate mineral popovite forms by
   sublimation at volcanic fumaroles).
- 1432. Each paragenetic mode applies to a group of minerals that share a formation144 process (e.g., more than 400 minerals form by sublimation at volcanic fumaroles).
- Mineral-forming environments that represent continuous variations of temperature
  and pressure, for example regional metamorphic facies (greenschist and
  amphibolite) or hydrothermal zones (epithermal and mesothermal) have been
  lumped together.

4. Related mineral-forming environments that represent an evolutionary sequence
have been split; for example, ultramafic igneous rocks are distinguished from
mafic igneous rocks that often arise by decompression melting of ultramafic rocks.

152 In spite of these guidelines, our preliminary chronological tabulations of the 153 paragenetic modes of minerals (Table 1; Supplementary Database 1) are subjective in at 154 least two important respects related to: (1) the classification of paragenetic modes, and (2) the relative timing of the initiation of Earth's principal mineral-forming processes. 155 156 Consequently, enumerating and defining different paragenetic modes must be an ongoing 157 process that involves a broad spectrum of mineralogists and petrologists. In this context, 158 we welcome advice and discussions regarding our provisional classification of 159 paragenetic modes.

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161 <u>The classification of paragenetic modes:</u> A significant source of subjectivity in our study 162 is intrinsic to any classification system – to what extent does one lump or split the varied 163 paragenetic modes of minerals? At the broadest scale, descriptors such as "intrusive

164 igneous," "regional metamorphic," and "hydrothermal alteration" are commonly 165 associated with the formation of minerals in different environments. These and similar 166 general terms may be applied to the characterization of paragenetic modes of most 167 minerals (e.g., Philpotts and Ague 2009; Wenk and Bulakh 2016). However, any attempt 168 to further subdivide mineral formation mechanisms, for example into compositional 169 categories of intrusive igneous rocks, facies of regional metamorphism, or gradations of 170 aqueous mineral alteration among metasomatic, hydrothermal, authigenic, and near-171 surface weathering processes, will be inherently subjective because minerals form over 172 continuous *P*-*T*-*X* regimes.

173 Three prior efforts that detail systems of paragenetic modes for important subsets of 174 minerals exemplify the utility and diversity of specialized approaches that can be 175 developed. Guilbert and Park (2007) presented a "litho-tectonic" classification that 176 recognizes more than 100 paragenetic categories of ore deposits based on a hierarchy of 177 (1) tectonic setting (e.g., mid-ocean ridge; cratonic; convergent margin), (2) host 178 lithologies (granite; massive sulfide; skarn), and (3) commodities (copper-gold; coal; 179 chromite). In their system, Guilbert and Park differentiate hydrothermal "Cyprus-type" 180 sulfide deposits from those at mid-ocean ridges, "proximal" massive sulfide deposits 181 from those at subducting margins, and sulfide deposits at back-arc basins versus those 182 from cratons. This detailed and comprehensive system is undoubtedly useful for placing 183 ore deposits in their plate tectonics contexts. However, it is not as appropriate for 184 applications to the full sweep of Earth's evolution for at least three reasons. First, 185 Guilbert and Park subdivide some types of mineralization, including volcanogenic 186 massive sulfide deposits, iron formations, and carbonatites, into multiple tectonic

187 contexts (e.g., cratons, back-arc basins, or subducting margins) – divisions that only 188 apply to Earth after the initiation of plate tectonics. By contrast, we differentiate 189 paragenetic modes primarily based on pressure-temperature-composition regimes, as 190 opposed to tectonic settings. Second, they narrowly subdivide many deposit types into 191 compositional groups (e.g., Cu, Cu-Mo, Mo, and Cu-Au porphyry copper deposits are 192 listed separately), whereas we do not employ composition as extensively to distinguish 193 among deposit types. And third, numerous non-commercial paragenetic modes, including 194 pre-terrestrial nebular minerals, products of near-surface weathering, impact 195 metamorphism, and biomineralization, are understandably not included in the Guilbert 196 and Park (2007) economic classification.

197 Another specialized list of paragenetic modes was offered in the comprehensive 198 survey of meteorite minerals by Rubin and Ma (2021, their chapter 10). They catalog 22 199 paragenetic modes representing minerals from the full range of meteorite finds and falls, 200 including eight processes related to primary crystallization from gases or melts of 201 different compositions, six variants of aqueous and/or thermal secondary alteration, four 202 variants of solid-state exsolution, shock metamorphism, annealing of amorphous 203 materials, and two forms of meteorite weathering (space weathering versus terrestrial 204 alteration). This revealing list is illuminating in terms of the variety of processes that 205 might produce new solid phases; however, in many instances the exact process by which 206 a meteorite mineral formed is ambiguous and a matter of debate, at times because 207 multiple mechanisms may come into play simultaneously or sequentially. We consolidate 208 Rubin and Ma's 22 modes into seven subsets that we apply specifically to meteorite 209 minerals: condensation of refractory phases in stellar atmospheres; condensation of

210 molecular solids in interstellar environments; condensation from nebular gas; 211 crystallization from chondrule melts; primary minerals formed during asteroid 212 differentiation; thermal, aqueous, and impact alteration of prior meteorite phases in an 213 asteroid body; and terrestrial weathering.

214 A third specialized tabulation of mineral-forming processes was provided by Hill and 215 Forti (1997), who illustrate more than 20 paragenetic modes associated with cave 216 minerals. Their comprehensive survey of 255 cave mineral species distinguishes 217 formation by varied mineral-precipitating aqueous fluids (e.g., rich in silica, carbonate, 218 sulfate, and/or metals); sublimates from subsurface volcanic fumarole vents; oxidation or 219 reduction of prior minerals; bacterial fixation, for example of Fe or Mn minerals; 220 subsurface evaporation to form halides or carbonates; numerous phosphate, nitrate, and 221 organic minerals related to reactions with guano; and freezing of ice. They also present a 222 division into six types of aqueous depositional environments: flowing water, dripping 223 water, pool water, seeping water, condensation water, and thermal water. Like Hill and 224 Forti (1997), we do not consider "cave minerals" a separate paragenetic mode; rather we 225 lump subsets of these shallow subsurface minerals with minerals formed by analogous 226 subaerial processes. Thus, we include halite formed by evaporation in a dry cave with 227 other evaporite minerals. Similarly, we lump subsurface cave fumarole minerals with 228 those that occur at volcanic vents at the surface.

Our preliminary tabulation recognizes 57 paragenetic modes, each assigned a reference number (cited as "[#xx]" in this contribution; see Table 1). This classification of paragenetic modes is admittedly subjective in several respects. In particular, when minerals form over a wide range of pressures, temperatures, compositions, and/or time

we have, of necessity, selected somewhat arbitrary subdivisions based on the followingcriteria:

235 1) Depth of formation: In most instances, we distinguish near-surface mineralization 236 that is presumed to form in direct atmospheric contact, either at the surface or in 237 shallow subsurface settings (e.g., volcanic lavas [#9], fumaroles [#11], hot-spring 238 precipitates [#14], subaerial aqueous alteration [#16], and evaporites [#25]), versus 239 subsurface processes not in atmospheric contact (e.g., intrusive igneous formations 240 [#19], subsurface aqueous alteration [#22], primary hydrothermal vein minerals 241 [#33], and regional metamorphism [#40]). Exceptions include Earth's earliest 242 lithologies (most of which are still being produced today) - ultramafic and mafic 243 igneous rocks – for which we lump intrusive and extrusive lithologies (e.g., 244 peridotite and komatiite [#7], and basalt and gabbro [#8]).

245 2) Metamorphic facies: Rather than differentiating among all facies of regional 246 metamorphic rocks, we recognize three broad P-T trends [e.g., as illustrated by 247 Philpotts and Ague (2009), their figure 16.6]. We lump facies of regional 248 metamorphic rocks (e.g., greenschist, amphibolite, and granulite facies) into one 249 paragenetic mode [#40]. We distinguish those facies from metamorphic rocks 250 formed at relatively high pressure and low temperature, including blueschist, 251 eclogite, and ultrahigh-pressure (UHP) facies [#39]; these lithologies are tied to 252 subduction tectonics and thus may be constrained to post-Archean time intervals of 253 Earth history. Finally, in the case of very low-pressure, high-temperature 254 "pyrometamorphic" minerals (Grapes 2006), we recognize four distinct paragenetic 255 environments, including: (1) thermally metamorphosed xenolith minerals (sanidinite,

or sometimes hornfels, facies) in volcanic and hypabyssal rocks [#9]; (2) nearsurface minerals altered by regional fires, as exemplified by the distinctive mineralogy of the Hatrurim Formation in the Middle East (Gross 1977; Sokol et al. 2019) [#51]; (3) anthropogenic pyrometamorphic minerals formed by coal mine fires [#54]; and (4) slag minerals associated with the smelting of ores [#56]. In the cases of regional and high-pressure metamorphic rocks, our conservative approach could be expanded by further subdivision into multiple facies in a future treatment.

263 3) Igneous lithologies: The classification of igneous rocks is a classic problem with no 264 unique solution. At one extreme, Johannsen (1932–1938) proposed hundreds of rock 265 types based on modest variations in mineralogy, bulk composition, and textures. 266 Significant order to the problem was achieved by the Subcommission on the Systematics of Igneous Rocks of the International Union of Geological Sciences (the 267 268 "IUGS classification": Streckeisen 1976, 1979; Le Bas et al. 1986; Le Bas and 269 Streickeisen 1991), which recognizes approximately 60 igneous rock types, again 270 based on mineralogy, composition, and texture. We adopt a more parsimonious 271 approach, with eight paragenetic modes chosen to reflect diagnostic suites of igneous 272 minerals. Ultramafic igneous rocks such as peridotite, pyroxenite, and komatiite 273 [#7], which are composed primarily of Mg-Fe minerals, represent the earliest near-274 surface rocks on Earth, though these lithologies still form today. We distinguish 275 ultramafic lithologies from plagioclase-rich mafic igneous rocks such as basalt and 276 gabbro, which are derived primarily from decompression melting of prior ultramafic 277 lithologies [#8]. We list both intrusive granitic igneous rocks [#19] and their acidic 278 volcanic equivalents such as rhyolite and dacite [#20], and we differentiate mineral-

279 rich complex pegmatites of granitic [#34] and agpaitic [#35] rocks. We also 280 recognize mineralogically-distinctive carbonatites and related carbonate-rich igneous 281 rocks [#36], as well as layered igneous formations because of their unusual richness 282 in minerals of platinum group elements (PGE: O'Driscoll and VanTongeren 2017) 283 [#37]. Five additional paragenetic modes are closely related to igneous processes: 284 pyrometamorphosed xenoliths in lava [#9], basalt-hosted zeolite minerals [#10], 285 volcanic fumarole minerals [#11] and [#45], and the highly-altered ultramafic rocks 286 of obducted ophiolites [#38]. Note that we do not treat a number of important 287 igneous lithologies, including andesite, anorthosite, monzonite, and syenite 288 (Streckeisen 1976; Le Bas and Streckeisen 1991; Ashwal 1993; Philpotts and Ague 289 2009), as separate paragenetic modes because they do not appear to incorporate new 290 mineral species or distinct formation mechanisms.

291 4) Water-rock interactions: Mineral formation processes related to water-rock 292 interactions pose a significant challenge in classifying mineral paragenetic modes. A 293 number of potentially ambiguous and partially overlapping terms, including aqueous 294 alteration, authigenesis, diagenesis, hydrothermal mineralization, metasomatism, and 295 serpentinization, have been applied to these formation environments of minerals 296 (Heinrich and Henley 1989; Pirajno 2009; Steele-MacInnis and Manning 2020). We 297 are not able to fully resolve this difficulty, but we have adopted the following definitions: 298

<u>Authigenesis:</u> Authigenesis refers to mineral formation *in situ* by direct
 precipitation from an aqueous solution, for example as a carbonate cement in
 detrital sediments. We apply this term exclusively to low-*T* (< ~100 °C)</li>

302 sedimentary environments, in contrast to some prior uses in metamorphic 303 petrology. We distinguish marine authigenesis [#17], which must have 304 commenced early in the Hadean Eon, from authigenesis in terrestrial sediments 305 [#24].

- Diagenesis: Diagenesis refers to the alteration of sediments in an aqueous environment by increasing *T* and *P*, resulting in compaction, lithification, dissolution/precipitation reactions, and some alteration of mineral chemistry by interactions with fluids. Our tabulations do not include diagenesis as a separate paragenetic process, as it may be viewed as the initial stages of metamorphism prior to the formation of new mineral assemblages.
- 312 • Hydrothermal mineralization: We define hydrothermal mineralization as any 313 precipitation of minerals from an aqueous phase that is > 100 °C or, in the case 314 of near-surface waters, significantly above ambient temperature. We 315 distinguish among mineralization associated with near-surface hot and/or 316 subaerial boiling fluids such as hot springs and geysers [#14]; seafloor hydrothermal vents, known as black smokers and white smokers [#15]; and 317 318 subsurface mineralization, usually deposited in veins and other fracture 319 systems [#12] and [#33]. Hydrothermal minerals are typically compositionally 320 distinct from their host lithologies. Note that we do not differentiate among 321 different P-T regimes of subsurface hydrothermal mineralization (e.g., 322 epithermal, mesothermal, and hypothermal).
- Metasomatism: Metasomatism is a general term for the subsurface
   hydrothermal alteration of minerals through mass transfer in an open system

325 (Harlov and Austrheim 2013). As such, the term can be applied to varied chemical processes (e.g., hydration/dehydration, dissolution/precipitation, and 326 327 ion exchange reactions). Metasomatism may affect the full spectrum of 328 subsurface lithologic settings, including metamorphism and diagenesis of 329 sediments, alteration of plutonic and volcanic rocks, skarn formation, and 330 alteration of a wide variety of ore deposits. Given this broad umbrella of 331 processes and environments, "metasomatism" is not particularly helpful as a 332 paragenetic descriptor. Therefore, we employ it in only in the specific case of 333 "mantle metasomatism" (Luth 2003; O'Reilly and Griffin 2012), which is a 334 term well embedded in the literature related to the alteration of mantle 335 lithologies by complex C-O-H fluids [#41].

Serpentinization: Serpentinization is the low-T (typically  $< 100 \,^{\circ}$ C, but at times 336 337 to T > 200 °C) aqueous alteration process by which anhydrous ferromagnesian 338 minerals in mafic and ultramafic rocks are transformed to hydrous phases, 339 including serpentine polytypes and brucite (Shrenk et al. 2013). 340 Serpentinization is just one of many mineral-producing water-rock 341 interactions; however, it is possibly the most ubiquitous chemical reaction on 342 wet terrestrial planets (e.g., Holm et al. 2015), and it has transformed at least 343 70 % of Earth's crust throughout its 4.5-billion-year history [#13].

• <u>Aqueous alteration:</u> "Aqueous alteration" is the most general term for any process that changes minerals by interactions with a water-based solution, irrespective of *P-T-X* conditions. We adopt "aqueous alteration" as a fallback term for the paragenesis of hundreds of mineral species that do not clearly fit

348	one of the above categories of water-rock interaction, especially when reported
349	details of formation processes are vague. We recognize five subsets of low- $T$
350	(< 100 °C) aqueous alteration, based on age and depth of formation: subsurface
351	and subaerial aqueous alteration of Hadean lithologies (> 4 Ga) $[#16]$ ;
352	subsurface aqueous alteration, prior to the Great Oxidation Event (GOE, >2.5
353	Ga) [#22]; subaerial aqueous alteration, pre-GOE [#23]; subsurface aqueous
354	alteration, post-GOE (< 2.5 Ga) [#46]; and subaerial aqueous alteration, post-
355	GOE [#47].

In addition to the above paragenetic mechanisms, we recognize five near-surface processes related to water-rock interactions that produce distinctive suites of minerals: freezing of aqueous solutions [#18]; pre-biotic surface precipitation of carbonate, phosphate, and iron formations [#21]; evaporite minerals [#25]; seafloor manganese nodules [#42]; and leaching zone soil minerals, including laterites and bauxites [#48].

361 5) Composition: The lumping and splitting of compositional variants is an admittedly 362 subjective aspect of our proposed system. For example, we distinguish between 363 minerals formed in complex granite pegmatites [#34] versus agaitic pegmatites 364 [#35], because the compositions of their host lithologies (and the resulting mineral 365 species) are dramatically different, even though the mechanisms of mineral 366 formation are similar. We divide contact metamorphic deposits into two somewhat 367 overlapping compositional groups: 356 metamorphosed carbonate (i.e., skarn), 368 phosphate, and iron-formation minerals [#31], are treated as distinct from 412 phases 369 associated with mineral-rich metamorphic Ba, Mn, Pb, and/or Zn deposits [#32]. 370 Also, as noted above, we separate 129 hydrothermal sulfides with common transition

metals [#12] from most other chalcogenides [#33] on the basis of their assumed chronology. However, with the exception of distinct platinum groups element (PGE) enrichments in layered intrusions [#37], we do not further subdivide hydrothermal metal deposits according to their principal metals or lithologic context.

375 6) Chronology – crust formation: In several instances, we have split minerals derived from mafic/ultramafic lithologies, and thus assumed to have first appeared early in 376 377 the Hadean Eon (Stage 3 of Table 1), from minerals likely derived from subsequent 378 granitic lithologies and the formation of continental crust (Stage 4) and/or after the 379 GOE (Stage 7). Thus, we distinguish minerals formed by authigenesis in marine 380 sediments as early as the Hadean Eon [#17] from authigenic minerals of Earth's 381 earliest continental crust, which likely appeared later [#24]. Similarly, we 382 differentiate the presumed earliest suites of Hadean minerals formed by subaerial 383 aqueous alteration of basaltic crust [#16] from those that more likely first formed subsequently in granitic crust [#23] or after the GOE (see below) [#47]. We also split 384 385 (with significant uncertainties) some presumably ancient hydrothermal vein sulfides 386 [#12] versus other chalcogenides [#33], because the extensive development of 387 sulfide deposits appears to predate that of other much less abundant chalcogenides, 388 such as those dominant in As, Sb, Se, and Te. Note that in all of these examples, 389 minerals that first formed early in the Hadean Eon continue to form today; our 390 sequence of mineral paragenetic modes thus reflects a gradual accumulation of more 391 and more mineral-forming processes, rather than a sequential replacement of 392 processes.

393	7) <u>Chronology – the Great Oxidation Event:</u> Late Archean (< 3 Ga) atmospheric
394	oxygenation (the "Great Oxidation Event" or GOE) represents a major transition in
395	the formation of near-surface minerals on Earth. Therefore, in four instances we
396	distinguish suites of relatively reduced, near-surface/subaerial phases, presumably
397	formed prior to the GOE from suites of more oxidized minerals that most likely
398	formed after the GOE. We split fumarolic minerals into more reduced phases [#11],
399	such as sulfides that were likely associated with volcanism early in the Hadean Eon,
400	and more oxidized species such as sulfates and arsenates [#45], that presumably
401	formed after atmospheric oxygenation. We differentiate between low-temperature,
402	subsurface aqueous alteration prior to the GOE [#22] and post-GOE [#46]. Similarly,
403	we divide low-temperature, subaerial aqueous alteration pre-GOE [#23] and post-
404	GOE phases [#47]. We also enumerate 11 minerals formed by anaerobic microbial
405	metabolism prior to the GOE [#44], compared to 77 biominerals produced directly
406	through cellular processes post-GOE [#49].
407	8) Near-surface oxidation/weathering: Because of the large number (~ 2000 species) of



416 The relative timing of Earth's principal mineral-forming processes: An important 417 objective of our study is to present a chronological tabulation of modes of mineral 418 formation, with quantitative geochronological constraints where possible. However, this 419 effort is faced with significant uncertainties. In a few instances, for example with primary 420 meteorite minerals [#1] to [#5] (> 4.56 Ga); Phanerozoic biomineralization and 421 taphonomic minerals (i.e., minerals formed via decay/alteration of biological materials) 422 [#49], [#52], and [#53] (< 541 Ma); coal minerals [#50] (< 360 Ma); and anthropogenic 423 minerals [#54] to [#57] (< 10 Ka), the ages of minerals are well defined by the nature of 424 the processes in question. In addition, over the past decade we have developed the 425 Mineral Evolution Database (MED; https://rruff.info/evolution, accessed 17 March 2021; 426 Golden et al. 2016; Golden 2020), with more than 190,000 minerals species/age/locality 427 data for more than 1000 different mineral species. However, in the cases of most mineral 428 species and their formation mechanisms, the absolute and relative timings of paragenetic 429 modes are poorly constrained.

430 Two well-known factors contribute to these uncertainties. First is the paucity of any 431 geological formations older than ~3.5 Ga; therefore, solid evidence for the formation and 432 timing of Hadean and Paleoarchean minerals is literally all but lacking. The ages of many 433 mineral-forming processes might be inferred if we knew the timings of significant 434 episodes in Earth history, such as the formation of continents, the establishment of 435 oceans, and the transition from vertical plume tectonics to lateral plate tectonics. 436 However, the nature and timing of these and other events from Earth's first billion years 437 are matters of intense ongoing debates. Among the uncertainties are:

438	• The composition and extent of Earth's earliest crust: The mineral evolution of
439	differentiated Earth began with subaerial solidification of the magma ocean that
440	formed immediately after the Moon-forming impact (> 4.50 Ga; Barboni et al. 2017;
441	Thiemans et al. 2019), coupled with initiation of mantle convection, possibly under a
442	"stagnant lid" (i.e., prior to significant recycling of surface layers; e.g., Debaille et al.
443	2013; Piper 2013; Stern 2018; Korenaga 2021). General agreement exists that
444	Earth's earliest crust featured globe-spanning mafic/ultramafic igneous lithologies;
445	however, significant debate remains regarding that crust's composition and thickness
446	- parameters that may have strongly influenced the earliest stages of mineral
447	evolution (Rollinson 2007b; Korenaga 2021). The earliest crust was likely more
448	mafic (i.e., with a higher average Mg content) than today (Taylor and McLennan
449	1995; Kemp and Hawksworth 2003; Keller and Schoene 2012). If a relatively thick
450	mafic crust (i.e., similar in composition to modern oceanic crust) prevailed, then a
451	gradual tectonic transition from stagnant lid to subduction seems plausible. If, on the
452	other hand, Earth's first crust was dominated by a relatively thin ultramafic layer, as
453	advocated by Korenaga (2021), then sinking dense crustal slabs may have triggered
454	an active early style of subduction-driven plate tectonics in the Hadean Eon. In this
455	study, we assume that some combination of mafic and ultramafic crustal lithologies
456	dominated Earth's earliest mineral diversity by ~4.50 Ga, without adopting a
457	position on the compositional and tectonic details.

The composition and extent of Earth's earliest oceans: Most models of early Earth
posit the rapid evolution of a hydrosphere, culminating in globe-spanning oceans by
460 4.45 Ga (Wilde et al. 2001; Mojzsis et al. 2001; Elkins-Tanton 2011). Anoxic

Hadean oceans must have differed significantly from more recent marine
environments. For example, estimates of Hadean ocean chemistry suggest
significantly higher salinity than the modern oceans (Holland 1984; Knauth 2005;
Marty et al. 2018) – conditions that might have led to enhanced evaporite deposition
at coastal margins. In this study, we assume that oceans were well established by
4.45 Ga.

467 The timing and extent of continental crust formation: A major uncertainty is the 468 timing of initiation and evolution of buoyant, felsic continental crust that was 469 relatively enriched in incompatible elements – a significant source of mineral 470 diversity. Stable, long-lived continental crust is thought to form principally through 471 extensive partial melting of a hydrated mafic layer in the upper mantle. Virtually all 472 models posit formation of some felsic crust by 4.4 Ga, but estimates of the rate of 473 subsequent continental "net crustal growth" vary wildly. At one extreme, some 474 authors suggest rapid granitic production, achieving close to modern extent of 475 continental crust by the early Hadean Eon (Armstrong 1991; Rosas and Korenaga 476 2018; Guo and Korenaga 2020). Others argue for a more gradual development of net 477 crustal volume, with modern levels not achieved until the mid-Proterozoic or later 478 (Belousova et al. 2010; Dhuime et al. 2012; Greber et al. 2017; see Korenaga 2018, 479 his Figure 1 and references therein). In this study, we assume that felsic continental 480 crust sufficient to enhance Earth's mineral diversity developed by 4.40 Ga, without 481 weighing in on the debate regarding the rate of continental evolution.

<u>The extent of dry land</u>: Significant uncertainties exist regarding the volume and
 depth of Hadean surface waters. Notably, recent models suggest that the Hadean

484 ocean may have held significantly more water, perhaps twice today's volume 485 (Kurokawa et al. 2018; Dong et al. 2021), as a consequence of the relatively hot, and 486 therefore dry, peridotitic mantle prior to 4 Ga. Subsequent mantle hydration through 487 plate tectonics gradually decreased the ocean's volume to modern levels (Jarrad 488 2003; Korenaga 2008; Korenaga et al. 2017; Rosas and Korenaga 2021). A 489 voluminous early ocean, possibly coupled with less extreme topography owing to a 490 steeper geothermal gradient and/or lack of orogenesis (e.g., Tang et al. 2021), may 491 have resulted in a largely submerged Hadean world with extensive serpentinization 492 and other water-rock interactions (Voosen 2021, Korenaga 2021). Such a scenario 493 has important implications for the emergence of dry land, which played a number of 494 roles in mineral diversification, for example through the formation of minerals by 495 evaporation, volcanic fumaroles, hot springs and gevsers, photo-oxidation, lightning 496 strikes, and impact mineralization. In spite of uncertainties regarding the extent of 497 dry land, we assume that at least some subaerial continental crust has been present 498 continuously since ~4.4 Ga.

<u>The nature of Earth's earliest atmosphere:</u> That Earth's atmospheric composition has changed dramatically through time is certain. However, the nature and timing of those changes is another enigmatic aspect of early Earth that influences models of Hadean mineral evolution. The widely cited model of Zahnle and colleagues (Zahnle 2006; Zahnle et al. 2007) suggests a Hadean transition from an initially dense CO<sub>2</sub>-rich atmosphere to one dominated by N<sub>2</sub> – a change that must have been accompanied by extensive early deposition of carbonate minerals. We adopt this

506 scenario and assume that carbonate precipitation was an important paragenetic 507 process throughout the Hadean Eon.

508 The nature and timing of the commencement of plate tectonics: The initiation of 509 subduction, possibly tied to a transition from vertical (plume-dominated or "heat 510 pipe") tectonics to modern-style lateral tectonics (Van Kranendonk et al. 2004, 511 2007a; Moore and Webb 2013), dramatically expanded Earth's mineralogical repertoire. However, significant uncertainties are reflected in a range of competing 512 513 models regarding both the style and timing of extensive crustal recycling (Condie 514 and Pease 2008; Brown et al. 2020; Korenaga 2021). Estimates for the initiation of 515 plate tectonics range from > 4 Ga (Hopkins et al. 2008; Rosas and Korenaga 2018; 516 Turner et al. 2020) to < 1 Ga (Stern 2005, 2018; Rollinson 2007a), with additional 517 discussions related to continuous versus episodic processes (O'Neill et al. 2007; 518 Belousova et al. 2010; Moyen and van Hunen 2012). In spite of these varied ideas, 519 many researchers have concluded that a significant transition occurred near  $\sim 3$  Ga to 520 a "modern style" plate tectonics, exemplified by the Wilson cycle (Shirey and Richardson 2011; Dhuime et al. 2012; van Hunen and Moyen 2012; Tang et al. 521 522 2016; Cawood et al. 2018). In this study we assume that significant subduction-523 driven plate tectonics and associated mineral-forming processes began at some point 524 after the earliest appearance of oceans and continental crust (i.e., < 4.3 Ga), but 525 significantly before atmospheric oxygenation (> 3.0 Ga).

526 In conclusion, we assume the following sequence of mineral-forming processes 527 associated with the evolution of Earth's lithosphere and hydrosphere: (1) solidification of 528 a mafic/ultramafic crust (*Stage 3a*; > 4.50 Ga); (2) formation of oceans and an active

hydrosphere (*Stage 3b*; > 4.45 Ga); (3) continental crust formation (*Stage 4*; > 4.40 Ga); and (4) initiation of subduction-driven plate tectonics (*Stage 5*; < 4.3 Ga). Note that this timeline is significantly more compressed than originally proposed by Hazen et al. (2008), thus suggesting that much of Earth's mineral diversity was established within the first 250 million years.

534

535 Preservation bias: A critical factor impeding our understanding of the relative timing of 536 paragenetic modes relates to inevitable biases associated with limited preservation of 537 Archean and Proterozoic rocks, especially near-surface minerals associated with 538 evaporites, fumaroles, hot springs, and other transient environments (e.g., Liu et al. 539 2019). Most minerals from those sources have been preferentially lost – preservation 540 biases that are especially relevant to documenting the first appearance of many rare 541 minerals (Hazen and Ausubel 2016). Therefore, the absolute and relative timing of 542 Earth's earliest minerals must often be inferred from geochemical and geophysical 543 models.

Significant uncertainties in the timing of mineral paragenesis relate to the emergence of minerals incorporating rare elements. In particular, 41 relatively rare mineral-forming elements (defined here as present at < 5 ppm in Earth's crust), including Sb, As, Be, Bi, Br, Cd, Cs, Au, Hf, In, Hg, Mo, Se, Ag, Ta, Te, Tl, Sn, U, W, the platinum group elements, and the rare earth elements, account for almost 2400 mineral species. Thus, fewer than 1 in 10,000 crustal atoms play an essential role in more than 42 % of Earth's total mineral diversity (https://rruff.info/ima, accessed 7 March 2021). An unanswered

551 question is how much time is required for these scarce elements to achieve localized 552 concentrations sufficient to produce new minerals.

Some evidence points to time periods greater than a billion years, as exemplified by the earliest known minerals containing Be (Grew and Hazen 2014), Cs (London 2008), and Hg (Hazen et al. 2012) from 3.00, 2.65, and 3.04 Ga, respectively. The paucity of these and other minerals containing rare elements with ages greater than 3 Ga suggests that it takes hundreds of millions of years for some scarce elements to become sufficiently concentrated, perhaps through fluid-rock interactions in large volumes of crust and upper mantle rocks affected by subduction-generated fluids.

560 On the other hand, at least some minerals incorporating rare elements arose much 561 earlier in highly localized deposits, many of which did not survive or are yet to be 562 exhumed. For example, micrometer-scale "nuggets" of PGE alloys enriched in osmium 563 and ruthenium have been found in meteorites, formed > 4.55 Ga either as primary 564 condensates from the solar nebula (Morrison and Hazen 2020) or exsolved from 565 thermally metamorphosed Fe-Ni metal of differentiated planetesimals and preserved in 566 iron meteorites (Hazen and Morrison 2021). Thus, in spite of the presence of Os and Ru 567 at crustal concentrations of 1 part per billion or less, these platinum group elements 568 rapidly segregated into concentrations sufficient to form PGE minerals. We conclude that 569 much more research will be required to determine how rapidly minerals of rare elements 570 might have formed in Earth's crust.

571

572

## **Systematic Survey of Paragenetic Modes**

573 In this section we describe 57 paragenetic modes of minerals, listed in a suggested 574 order of earliest appearance, as summarized in Table 1. This analysis is based on a newly 575 constructed database of 5659 minerals, each represented by one or more paragenetic 576 modes (see Supplementary xlsx spreadsheet, [EDITOR: Please advise here on format]) – 577 work that would not have been possible without the availability of large and growing 578 open-access data resources (Hazen 2014; Lafuente et al. 2015; Hazen et al. 2019; Golden 579 2020; Prabhu et al. 2021). Paragenetic modes for each IMA-CNMNC-approved mineral 580 species were determined by consulting one or more of the following sources: (1) lists of 581 IMA-approved mineral species and associated links to primary references 582 (https://rruff.info/ima, accessed 25 February 2021; Lafuente et al. 2015); (2) tabulations 583 of mineral species and their localities, associated species, and (in many instances) 584 geological contexts (https://mindat.org, accessed 25 February 2021); (3) detailed 585 descriptions of paragenesis for rock-forming minerals in Deer et al. (1982-2013), Rock-586 Forming Minerals, Second Edition, 11 volumes; (4) brief descriptions of "Occurrence" in 587 Anthony et al. (1990-2003), Handbook of Mineralogy, 6 volumes; and (5) books and 588 articles describing new individual mineral species and their modes of occurrence from 589 primary references (as listed in sources 1 through 4, above). This initial version of the 590 paragenetic mode spreadsheet, with 10,556 combinations of an IMA-approved mineral 591 species and one of 57 paragenetic modes, represents an ongoing effort; we welcome 592 additions and corrections.

593 Several aspects of this treatment (Table 1) should be noted:

• While most of the paragenetic modes encompass numerous mineral species, several unusual categories are represented by 10 or fewer minerals. These relatively rare

paragenetic processes include minerals formed by interstellar condensation [#2] (8
species), near-surface freezing [#18] (4), auto-oxidation through radioactive decay
[#27] (9), photo-oxidation [#28] (10), lightning [#29] (9), and shear stresses such as
mylonitization [#43] (9).

A significant number of durable mineral species, including metal alloys, carbides, and chalcogenides, are known only from detrital deposits, placers, or heavy mineral separates. In several instances, the origins of these grains are unknown. We list "detrital" as a paragenetic mode, even though detrital refers to a local accumulation of mineral grains in a sedimentary environment, as opposed to a formation mechanism, *per se*.

A number of mineral-forming processes, including exsolution, order-disorder
 transitions, and reversible phase transitions, occur in the solid-state during cooling
 and/or annealing. We have chosen to lump these processes with broader igneous and
 metamorphic paragenetic categories rather than treat them as separate formation
 processes. However, solid-state transition mechanisms could be added as one or
 more paragenetic modes in future studies.

Several minerals form only in specific environmental contexts, for example in fluid
 inclusions [e.g., zabuyeite; Li<sub>2</sub>(CO<sub>3</sub>)], in caves [guanine; C<sub>5</sub>H<sub>3</sub>(NH<sub>2</sub>)N<sub>4</sub>O], or as
 epitaxial overgrowths [marsturite; NaCaMn<sub>3</sub>Si<sub>5</sub>O<sub>14</sub>(OH)]. We catalog these
 minerals according to their petrogenetic settings, rather than by their environmental
 contexts.

• Similarly, we do not consider the formation processes of morphologically distinct 618 minerals, such as lava phenocrysts, minerals that occur exclusively as nanophases

- 619 (e.g., protoenstatite; MgSiO<sub>3</sub>), or minerals that display efflorescence, as distinct
- 620 paragenetic modes.
- Finally, we do not list minerals from the Moon or Mars. The comparative mineralogy
  of extraterrestrial bodies will be treated in a separate contribution.
- 623

524 <u>Stage 0. Pre-terrestrial "Ur-minerals"</u> We tabulate 296 IMA-approved species as pre-525 terrestrial minerals on the basis of their occurrences in meteorites (Hazen and Morrison 526 2021; Rubin and Ma 2021). We divide these diverse phases, at least 97 of which are 527 known only from meteorites, into six sequential paragenetic modes ([#1] through [#6]), 528 representing the first three stages of mineral evolution.

629 [#1 - Stellar atmosphere condensates]: The earliest of these phases, dubbed "Ur-630 minerals" by Hazen et al. (2008), include 22 IMA-approved mineral species that formed 631 prior to the solar nebula in the expanding, cooling atmospheres of highly-evolved stars 632 (Zinner 2014; Nittler and Ciesla 2016; Hazen and Morrison 2020). Stellar minerals, 633 which are identified by their extreme isotopic anomalies as pre-solar grains, include the 634 most ancient condensed matter yet identified, with some individual stellar moissanite 635 (SiC) grains dated ~7 billion years old (Heck et al. 2020). Hazen and Morrison (2020) 636 catalogued these stellar minerals and further subdivided them into three probable sources 637 - AGB stars, classic novae, and core-collapse supernovae. Subsequent work by Boujibar 638 et al. (2021) employed cluster analysis on the isotopic attributes of thousands of stellar 639 moissanite grains to identify at least seven distinct SiC groups associated with variants of 640 these stellar types.

641 [#2 - Interstellar condensates]: Morrison and Hazen (2020) listed an additional 8 probable interstellar condensed molecular C-H-O-N "ice" phases, which have been 642 identified in so-called "cool" (T < 20 K), "dense" (P <  $10^{-13}$  atm) molecular clouds by 643 644 telescopic observations of absorption and emission spectra of interstellar molecules in 645 radio, microwave, or infrared wavelengths (Greenberg 1991; Whittet 2003; Gibb et al. 646 2004). All of these condensed phases would have formed prior to the solar nebula (i.e., >647 4.567 Ga). With the exception of water ice, these molecular solids are not currently 648 recognized as minerals by the IMA-CNMNC.

649

650 <u>Stage 1. Primary nebular phases</u> Stage 1 of Earth's mineral evolution, which occurred 651 between 4.567 and 4.561 Ga, encompassed two mineral-forming processes.

[#3 – Solar nebular condensates]: Initial mineral formation occurred as nebular gas
cooled and highly refractory phases condensed at temperatures greater than ~1000 K.
Morrison and Hazen (2020) catalogued 48 IMA-approved minerals that are preserved in
calcium-aluminum-rich inclusions (CAIs; MacPherson 2014), amoeboid olivine
aggregates (AOAs; Krot et al. 2004), and ultra-refractory inclusions (URIs; El Goresy et
al. 2002).

[#4 – Primary chondrule phases]: Overlapping pulses of nebular mineralization are
preserved as the primary igneous minerals of chondrules, which represent solidified
droplets, typically < 0.1 cm diameter, that formed during the first few million years of the</li>
solar nebula, ~4.566 to 4.561 Ga (Brearley & Jones 1998; Krot et al. 2014; Russell et al.
2018; Hazen et al. 2021). Chondrules are preserved in abundant chondrite meteorites,
which may be thought of as the oldest sedimentary rocks in the solar system. We identify

47 primary chondrule minerals, most of which form from 10 abundant chemical elements
- C, O, Mg, Al, Si, S, Ca, Ti, Cr, and Fe.

666

667 <u>Stage 2. Planetesimal differentiation</u> Within the first million years of the solar nebula's 668 formation, the primary condensates of *Stage 1* had begun to accrete into the first 669 generation of planetesimals. Over the next few million years (~4.566 to 4.560 Ga; 670 Blichert-Toft et al. 2010), some of these bodies grew to diameters exceeding 100 km – 671 large enough to melt and differentiate into a metallic core and rocky mantle. Subsequent 672 collisions disrupted some of these objects to produce a rich variety of non-chondritic 673 meteorites.

674 [#5 – Primary asteroid phases]: Morrison and Hazen (2021) tabulate 94 IMA675 approved species that occur as primary igneous minerals in iron, stony-iron, and
676 achondrite meteorites (Mittlefehldt et al. 1998; Krot et al. 2014; Mittlefehldt 2014).

677 [#6 – Secondary asteroid phases]: A final pre-terrestrial phase of mineral evolution is 678 preserved as a diverse variety of secondary phases formed by aqueous, thermal, and/or 679 impact alteration of prior meteorite minerals. These processes produced more than 200 680 IMA-approved mineral species, primarily during an interval from ~4.565 to 4.550 Ga 681 (McSween et al. 1988; Brearley and Jones 1998; Mittlefehldt et al. 1998; Hazen and 682 Morrison 2021; Rubin and Ma 2021). This period of secondary mineralization saw the 683 first occurrence of several important mineral groups, including sulfates, carbonates, 684 hydroxides, and layer silicates (notably clay minerals).

685 An important caveat relates to the distinction between primary and secondary 686 meteorite minerals. Most secondary meteorite minerals are unambiguously the products 687 of aqueous and/or thermal alteration of earlier phases, yet at least two complications may

arise. First, terrestrial weathering may lead to some secondary alteration that is difficult
to distinguish from pre-terrestrial processes. However, most terrestrial weathering
products arise from oxidation (see *Stage 7* below) and are thus distinct from asteroidal
alteration.

692 A subtler situation arises from the gradual alteration of primary asteroid minerals to 693 secondary phases. Such transformations may involve incremental shifts in composition 694 and atomic structure spanning thousands to millions of years. Some suites of secondary 695 mineral properties, such as exsolution, order/disorder, or zoning, are the consequences of 696 thermal and/or aqueous alteration, but sharp boundaries may not exist between the 697 primary and secondary forms of minerals. Furthermore, it has been suggested that some 698 presumably secondary asteroidal minerals might have also formed via primary nebular 699 processes, for example via sulfidization, oxidation, and/or hydration during pre-700 accretionary interactions with a warm nebular gas (Krot et al. 1995, their Table 2; 701 Bischoff 1998; Ciesla at al. 2003). These ambiguities are emblematic of challenges to any 702 comprehensive tabulation of mineral paragenesis.

703

Stage 3a. Earth's earliest Hadean crust Proto-Earth's earliest crust must have solidified before 4.55 Ga, shortly after the planet's accretion and differentiation, though that stage of Earth's mineral evolution is lost. The Moon-forming event (perhaps ~ 4.51 Ga; Barboni et al. 2017) completely obliterated the initial solid outer shell, but a new crust dominated by ultramafic and mafic igneous lithologies crystalized shortly thereafter (> 4.50 Ga). No minerals have been preserved from that Hadean world, save for a few detrital zircon (ZrSiO<sub>4</sub>) crystals with ages as great as 4.4 Ga (Wilde et al. 2001).

Nevertheless, we suggest that five major groups of minerals, totaling approximately 400 mineral species, arose from this earliest terrestrial *Stage 3a* of mineral evolution (modes [#7] through [#11]; Table 1). Many of these phases, previously cataloged by Hazen (2013) and Morrison et al. (2018) in inventories of Hadean "paleomineralogy," are familiar oxides and silicates with the most abundant crustal elements (notably H, Al, Na, Mg, K, Ca, Fe, and Ti), as well as accessory sulfides, possibly arsenides, and other relatively reduced phases.

[#7 – Ultramafic igneous rocks]: Ultramafic igneous rocks, including peridotite, 718 719 pyroxenite, and komatiite, are dominated by ferromagnesian minerals and lack significant 720 plagioclase (Dick 1989; Isley and Abbott 1999; Guilbert and Park 2007; Philpotts and 721 Ague 2009; Lambert et al. 2016). These lithologies played a significant role in the 722 mineralogy of Hadean Earth's crust and mantle (Rollinson 2007b; Van Kranendonk et al. 723 2007b), though significant uncertainty remains regarding the relative volumes of 724 ultramafic versus mafic lithologies (e.g., Korenaga 2021). We list 123 minerals that have 725 been identified in ultramafic intrusive and extrusive igneous rocks.

726 [#8 - Mafic igneous rocks]: Mafic igneous rocks, notably volcanic basalt and its 727 intrusive equivalent gabbro, have been important crustal lithologies throughout Earth's 728 history (Rollinson 2007b; Van Kranendonk et al. 2007b; O'Neil et al. 2008). Most basalt 729 is thought to form by decompression partial melting of ultramafic rocks in Earth's 730 mantle, which produces a buoyant magma. Not only is basalt the dominant lithology of 731 the oceanic crust, but it also is produced in huge volumes in continental settings during 732 the episodic production of large igneous provinces (LIPS; Ernst 2014), which have 733 occurred since at least 2.8 Ga (Prokoph et al. 2004). The mineralogy of basalt is always

dominated by plagioclase and pyroxene; however, a variety of basalt subtypes based on relative amounts of Mg, Fe, Al, Ti, and alkali metals incorporate different accessory minerals and display diagnostic compositional attributes. In this study, we identify 93 mineral species from Precambrian mafic igneous rocks. Further detailed studies of basalt minerals, for example employing cluster analysis on trace and minor elements, may point to additional subdivision of these phases into natural kinds (Hazen 2019; Boujibar et al. 2021).

[#9 – *Lava/xenolith minerals*]: Additional mineralogical variety would have been present in the form of thermally-altered xenoliths that were derived from both mantle and crustal sources in abundant Hadean lavas (Grapes 2006). We identify 127 mineral species that were contributed by this igneous pyrometamorphic (most typically sanidinite facies) process. Note that some of these xenolith minerals, notably those representing altered carbonates and alkalic igneous rocks, must have formed significantly after 4.5 Ga to allow enough time for compositional differentiation.

748 [#10 – Basalt-hosted zeolite minerals]: A significant number of late-stage zeolite 749 group minerals likely formed in the cavities of cooling basalt (Deer et al. 2004). We 750 identify more than 100 such phases, with the caveat that unaltered zeolite group minerals 751 are rarely preserved in Precambrian formations and any tabulation based on ages of 752 formation must remain speculative.

[#11 – Volcanic fumarole minerals, reduced]: A modest suite of a few dozen reduced
phases, predominantly transition metal sulfides, likely formed as sublimates at Hadean
volcanic fumaroles (Grapes 2006; Vergasova and Filatov 2016). These phases represent a

subset of the more than 450 documented fumarole minerals, most of which are moreoxidized phases that we assign to post-GOE formation [#45].

758

759 Stage 3b. Earth's earliest hydrosphere Earth's earliest hydrosphere, fueled by 760 ubiquitous volcanic venting of water vapor, led to cloud formation, rain, development of 761 streams and rivers, and ultimately the establishment of oceans. A robust hydrological 762 cycle was probably operating shortly after the solidification of Earth's crust, with 763 substantial bodies of water covering a large fraction of the surface by > 4.45 Ga (Wilde et 764 al. 2001; Elkins-Tanton 2011; Dong et al. 2021), and a vigorous subsurface hydrothermal 765 circulation of aqueous fluids (Heinrich and Henley 1989; Pirajno 2009). The consequent 766 water-rock interactions may have produced as many as 350 mineral phases in near-767 surface marine and terrestrial environments.

768 [#12 – Hadean hydrothermal minerals]: Subsurface Hadean hydrothermal sulfide 769 deposits, with more than 100 likely species, represent an important yet enigmatic early 770 source of mineral diversity. Today, hydrothermal systems boast more than 850 species of 771 sulfides, arsenides, antimonides, and a wide range of sulfosalts bearing relatively rare 772 elements such as Ge, Sn, Bi, Se, and Te. However, we postulate that these uncommon 773 elements were not sufficiently concentrated in Hadean hydrothermal fluids to generate 774 that wide range of compounds. In spite of longstanding interest in the secular variation of 775 sulfide-rich ore bodies (Laznicka 1973; Meyer 1988; Titley 1993; Barnes and Rose 1998; 776 Goldfarb et al. 2001; Groves et al. 2005, 2010; Huston et al. 2010; Leach et al. 2010; 777 Maier and Groves 2011; Jenkin et al. 2015), the changing mineralogies of these deposits 778 through time, especially regarding minerals containing relatively rare elements, have not

been detailed to our knowledge. We therefore limit our list of the earliest hydrothermal chalcogenides to sulfides (and in a few cases arsenides) of the commonest transition metal elements, as well as PGE elements, which are thought to concentrate in fluids associated with ultramafic lithologies owing to their highly incompatible nature (Mungall and Naldrett 2008). We list most hydrothermal ore minerals under *Stage 4* [#33], when extensive fluid-rock interactions in the upper mantle and crust associated with subduction is thought to have selected and concentrated rare elements.

[#13 – Hadean serpentinization]: Low-temperature aqueous alteration of Hadean igneous lithologies, in both near-surface and subaerial environments, must have produced hundreds of new mineral species. Most notably, serpentinization would have transformed ocean floor mafic and ultramafic rocks to familiar assemblages of brucite, serpentine, magnetite, and more than 60 other phases (Blais and Aubrey 1990; Lowell and Rona 2002; Palandri and Reed 2004; Shrenk et al. 2013; Holm et al. 2015; Menez et al. 2018).

792 [#14 - Hot springs, geysers]: The surface of the Hadean world would have featured

abundant subaerial hot springs and geysers that produced a range of geothermal minerals,

perhaps including the earliest terrestrial occurrences of calcite (CaCO<sub>3</sub>), baryte (BaSO<sub>4</sub>),

and a variety of hydrated phases. Widespread low-temperature aqueous alteration of

Hadean lithologies would also have enriched early Earth's inventory of hydrous silicates,

including clay minerals and zeolites (Deer et al. 2004; Wilson 2013).

[#15 – Black/white smokers]: A subset of more than 20 hydrothermal sulfide minerals
would have occurred with high-temperature aqueous alteration phases at Earth's earliest
"black smoker" environments, which, along with carbonate-rich "white smokers," we
consider to be a distinct seafloor paragenetic environment (Hekinian et al. 1980; Haymon

802 and Kastner 1981; Palandri and Reed 2004; Schwarzenbach and Steele-MacInnis 2020). 803 Note that the abundance and character of submarine hydrothermal vents may have 804 changed significantly through time. Compared to today, Hadean Earth had significantly 805 greater volcanic activity coupled with a larger extent of oceans. Nevertheless, the onset of 806 modern-style plate tectonics would have established continuous chains of submarine 807 vents at and near divergent plate boundaries (Lowell et al. 2008). Therefore, we conclude 808 that seafloor hydrothermal systems have been active mineral-forming environments for at 809 least 4.5 billion years.

810 [#16 - Low-T aqueous alteration]: In addition to serpentinization, low-temperature 811 aqueous alteration of Hadean subaerial and shallow subsurface lithologies produced more 812 than 80 mineral species, including dozens of zeolites, clay minerals, and other hydrous 813 silicates (Deer et al. 2004; Wilson 2013).

814 [#17 – Marine authigenic minerals]: Hadean seafloor water-rock interactions would 815 have produced a complementary suite of perhaps 50 authigenic phases in low-816 temperature marine sediments, including a variety of hydrous silicates, notably zeolites 817 (Deer et al. 2004). We have attempted to differentiate authigenic minerals, which 818 precipitated in place (typically in a sedimentary environment), from aqueous processes 819 that alter prior minerals through fluid-rock interactions (e.g., modes [#16], [#22], [#23], 820 [#46], and [#47]). Note also that we distinguish these marine authigenic minerals, which 821 began forming early in the Hadean Eon, from those that we suggest formed somewhat 822 later in a continental setting [#24].

823 [#18 – Minerals formed by freezing]: Finally, the Hadean hydrosphere must have
824 hosted the earliest terrestrial examples of minerals formed by freezing of aqueous
825 solutions, most abundantly ice (H<sub>2</sub>O), but also rare occurrences of hydrohalite (NaCl'2H<sub>2</sub>O), ikaite (CaCO<sub>3</sub>'6H<sub>2</sub>O), and possibly ernstburkeite [Mg(CH<sub>3</sub>SO<sub>3</sub>)<sub>2</sub>'12H<sub>2</sub>O], 826 827 which was discovered in an ice core from Antarctica (Genceli Güner et al. 2013). 828 829 Stage 4a. Earth's earliest continental crust The occurrence of individual zircon grains 830 with ages as great as ~4.4 Ga (Wilde et al. 2001; Cawood et al. 2013) suggests that some 831 granitic continental crust formed through partial melting of hydrated basalt within Earth's 832 first 150 million years, significantly before the onset of modern-style plate tectonics 833 according to many models (Shirey and Richardson 2011; Dhuime et al. 2012; van Hunen 834 and Moyen 2012; Tang et al. 2016; Johnson et al. 2017; Cawood et al. 2018; however, 835 see Korenaga 2021). Crustal processes during the Hadean and Archean Eons led to 836 significant mineral diversification, ultimately approaching a total of 2400 mineral species 837 (Table 1).

838 [#19 - Granitic intrusive rocks]: We list 143 primary minerals that were associated 839 with the formation of granite and related intrusive acidic rocks during the Hadean Eon – 840 mineralization that continues today. Studies of many granitic bodies point to different 841 sources of parent magmas, including mantle-derived ("M" type); sediment-derived ("S" type); deep-crustal, orogenic igneous ("I" type); and anorogenic ("A" type) granites 842 843 associated with hot spots (Chappell and White 2001; Eiler 2007; Philpotts and Ague 844 2009). Therefore, an intriguing aspect of granite mineralogy, yet to be explored, is 845 whether different types of granite might result in distinctive mineral attributes and thus 846 warrant further paragenetic subdivisions.

847	[#20 – Acidic volcanic rocks]: Rhyolite, dacite, and other quartz- and alkali feldspar-
848	bearing volcanic rocks played an important role in the evolution of continental crust
849	(Rollinson 2007b; Van Kranendonk et al. 2007b; Philpotts and Ague 2009). We identify
850	45 mineral species produced by acidic volcanism, including phases uniquely associated
851	with ash, such as quadridavyne $[(Na,K)_6Cl_2Ca_2Cl_2(Si_6Al_6O_{24})]$ , as well as its rapid
852	devitrification and hydration products [e.g., clinoptilolite-Na; Na <sub>6</sub> (Si <sub>30</sub> Al <sub>6</sub> )O <sub>72</sub> <sup>•</sup> 20H <sub>2</sub> O].
853	Note that we distinguish intrusive and extrusive acidic rocks because they have different
854	suites of minerals, as opposed to ultramafic [#7] and mafic [#8] igneous rocks.
855	[#21 – Chemical precipitates]: A wide variety of near-surface processes accompanied
856	the establishment of subaerial continental and shallow marine environments, resulting in
857	the formation of more than 1100 mineral species. Continental margins and lake
858	environments saw significant chemical precipitation of carbonates (Falkowski et al. 2000;
859	Boggs 2006), phosphorites (Baturin and Bezrukov 1979; Button 1982; Delaney 1998),
860	and iron formations (Klein 2005; Bekker et al. 2010), with at least 79 associated mineral
861	species. Extensive abiotic carbonate formations were especially consequential for
862	environmental changes in the Hadean Eon, as they rapidly became the dominant crustal
863	carbon reservoir, mirrored by a dramatic reduction of atmospheric CO <sub>2</sub> (Falkowski et al.
864	2000).
065	

[#22 – Low-T aqueous alteration]: Hydration and low-temperature aqueous alteration
of a wide range of lithologies in shallow subsurface environments must have occurred as
soon as surface rocks were exposed to water. We estimate that Hadean aqueous alteration
led to 247 mineral species, including numerous hydrous silicates, hydrated phosphates in

altered granitic rocks, and more than 50 altered Mn minerals (Savage et al. 1987; Post
1999; Nishimoto and Yoshida 2010).

[#23 – Subaerial aqueous alteration]: We record an additional 398 minerals formed by subaerial and near-surface aqueous alteration prior to significant oxidation (i.e., > 3Ga). Many of these phases, notably including hydrous silicates, carbonates, phosphates, and borates, likely form today in relatively reducing environments (Chang et al. 1996). This suite of several hundred mineral species contrasts with the almost 2000 oxidized/weathered species that appeared following the GOE (< 3 Ga) [#47].

877 [#24 - Terrestrial authigenic minerals]: We identify 74 minerals that likely formed in 878 continental sediments as authigenic species, in contrast to marine authigenic phases 879 [#17]. Most of these minerals are relatively common silicates, carbonates, or phosphates 880 that are known to form in a variety of environments (Chang et al 1996; Deer et al. 2006).

881 [#25 - Evaporites]: The emergence of dry continental crust, with the likely occurrence 882 of extensive inland seas, saw increased opportunities for deposition of diverse evaporite 883 minerals (Button 1982; Buick and Dunlop 1990). We catalog 210 plausible phases; 884 however, the relative chronology of their first appearances, which must have been 885 strongly tied to composition, is not known. Halides, notably halite (NaCl) and sylvite 886 (KCl), must have formed on Earth's first shorelines in the early Hadean Eon. Sulfates 887 may have followed soon thereafter, especially if abiotic sulfur photo-oxidation played a 888 significant role in near-surface processes (see mode [#28] below). However, it may have 889 taken much longer for significant deposition of borates or nitrates to occur (e.g., Grew et 890 al. 2011, 2015).

891 [#26 – Detrital minerals]: Numerous mechanically robust minerals, including dozens 892 of metal alloys, carbides, and chalcogenides not yet discovered in situ, are preserved in 893 sediments as detrital minerals (Baker 1962). We list 250 detrital mineral species that we 894 suggest were present by the Hadean or Archean Eons. We have attempted to assign most 895 of these phases to other paragenetic modes, as well, based on their chemistries and 896 assumed lithological associations. Note, however, that in a few instances the original 897 contexts of detrital minerals are unknown [e.g., enigmatic tantalcarbide (TaC) and 898 niobocarbide (NbC)]; therefore, in these few examples the true paragenetic mode is 899 unresolved.

900 [#27 – Radioactive processes]: A number of minor but intriguing paragenetic 901 processes led to the formation of idiosyncratic minerals in near-surface environments. For 902 example, primary uranium minerals of the Hadean Eon would have invariably formed as  $U^{4+}$ phases, most commonly uraninite (UO<sub>2</sub>) and coffinite (USiO<sub>4</sub>). However, 903 radioactive decay of  $U^{4+}$  to  $Pb^{2+}$  led to the intriguing phenomenon of "auto-oxidation," 904 by which uranium is oxidized to  $U^{6+}$  (Frondel 1958; Finch and Murakami 1999; Hazen et 905 906 al. 2009, and references therein). Auto-oxidation resulted in localized formation of a few uranyl-oxide-hydroxide phases, notably ianthinite  $[U^{4+}(U^{6+}O_2)O_4(OH)_6(H_2O)_0]$ . 907 908 schoepite  $[(UO_2)_8O_2(OH)_{12}](H_2O)_{12}]$ , and becquerelite  $[Ca[(UO_2)_3O_2(OH)_3]_2(H_2O)_8]$ , 909 as well as Pb-bearing phases such as fourmarierite  $[Pb[(UO_2)_4O_3(OH)_4](H_2O)_4]$ , 910 kasolite  $[Pb[(UO_2)(SiO_4)](H_2O)],$ vandendriesscheite and  $[Pb_{1,5}[(UO_{2})_{10}O_{6}(OH)_{11}](H_{2}O)_{11}].$ 911

40

912 [#28 - Photo-alteration]: Early Earth did not have a protective ozone layer; therefore, 913 the near-surface environment was highly susceptible to UV photo-oxidation – a 914 potentially widespread but as yet incompletely studied phenomenon. A limited number of 915 minerals are known from laboratory studies to form by photo-oxidation processes (e.g., 916 Kim et al. 2013); we list 10 of these species. In some instances, such as pararealgar (AsS), digenite (Cu<sub>1 8</sub>S), and ramsdellite (MnO<sub>2</sub>), new minerals are known to form from 917 918 prior minerals that are exposed to visible light or ultraviolet radiation. Less well studied 919 are processes by which UV oxidation produces reactive oxidized chemicals, notably the conversion of aqueous  $Fe^{2+}$  to  $Fe^{3+}$  with associated reduction of water and release of H<sub>2</sub>. 920 921 Recent experiments by Yee and colleagues (Nathan Yee, personal communication, 7 February 2021) reveal that  $Fe^{3+}$  is a strong oxidant that can lead, for example, to abiotic 922 923 production of elemental selenium and sulfate minerals from primary selenides and 924 sulfides in anoxic environments. One can infer that many analogous reactions might have 925 occurred during the Hadean and early Archean Eons, for example producing a suite of 926 arsenate and antimonate minerals from precursor arsenides and antimonides. We are not 927 yet able to definitively catalog such prebiotic oxidized phases; however, the possibility 928 remains that significant mineral diversity arose indirectly through photo-oxidation prior 929 to the Great Oxidation Event (Stage 7).

930 [#29 – Lightning minerals]: A small suite of 9 highly-reduced minerals, including
931 silicon metal, graphite, iron, moissanite, and schreibersite, is associated with the
932 production of fulgurites by lightning strikes (Essene and Fisher 1986; Grapes 2006; Pasek
933 et al. 2012; Hess et al. 2021). Pasek and Block (2009) suggested that lightning-induced

934	reduction of phosphate minerals might have provided a source of biologically useful P
935	compounds prior to life's origins – an idea amplified by Hess et al. (2021).
936	[#30 - Terrestrial impact minerals]: Bolide impacts on exposed Hadean crust must
937	have produced a range of high-pressure shock-induced phases. We tabulate 16 known
938	impact species, including high-pressure forms of carbon (diamond), SiO <sub>2</sub> (coesite,
939	stishovite, and chaoite), TiO2 (riesite and akaogiite), and other oxides and silicates
940	(Tomioka and Miyahara 2017; Tschauner 2019).
941	[#31 – Thermal alteration] Thermal metamorphism of diverse lithologies, including
942	sediments, igneous rocks, and metal-rich deposits, led to more than 1500 new phases
943	produced in subsurface continental environments. We tabulate 356 minerals that were
944	produced by thermal alteration reactions of prior phases with aqueous fluids, notably
945	those rich in carbonate and phosphate, as well as sulfate, borate, halogens, and other
946	solutes. Continents with deep roots and active hydrological cycles also hosted mineral
947	formation in more extreme subsurface environments, for example by high-temperature
948	alteration through metasomatism and/or contact metamorphism of carbonates (Falkowski
949	et al. 2000), phosphates (Button 1982), and iron formations (Klein 2005; Kappler et al.
950	2005).

[#32 – Ba/Mn/Pb/Zn deposits]: A remarkable variety of high-temperature minerals
containing Ba, Mn, Pb, and/or Zn, including contact and regional metamorphic deposits,
arose through processes in the continental crust (Post 1999; Leach et al. 2010). We
tabulate 412 species, including 185 phases unique to these environments, from classic
localities, including the Wessel manganese mine, South Africa (Caincross and Beukes
2013); Broken Hill, Australia (Spry et al. 2008); Fresno County, California (Alfors et al.

1965); and Franklin, Sussex County, New Jersey (Peters et al. 1983). These occurrences
underscore the importance of a few mineral-rich localities in establishing Earth's mineral
diversity.

960 [#33 – Hydrothermal deposits]: Subsurface hydrothermal deposition of metal-rich 961 veins and other bodies, primarily in environments associated with volcanism, produced  $\sim$ 962 800 mineral species, most of which are unique to hydrothermal origins. Significant 963 uncertainties remain regarding the sequence and timing of the earliest appearances of 964 these varied, economically important sulfide, arsenide, selenide, antimonide, and related 965 sulfosalt minerals (Meyer 1988; Guilbert and Park 2007; Bradley 2011). We suggest that 966 early Hadean formations contained a relatively small subset of these phases, primarily 967 sulfides and some arsenides of the commoner transition elements; accordingly, we list 968 129 mineral species associated with mode [#12]. Note that prior to enhanced 969 mineralization associated with plate tectonics (Hazen et al. 2014; Groves et al. 2015), 970 most volcanism was likely associated with hot spots and vertical tectonics – processes 971 that may have been accompanied by far less crustal fluid-rock interactions than today's 972 subduction-related volcanism. We suggest that relatively few large-scale, metal-rich 973 hydrothermal systems would have occurred in those settings, analogous to what is 974 observed today at mineral-poor hot spots such as Hawaii, Yellowstone, and Iceland 975 (Sherman et al. 1968; Plazo-Toledo 2019). We conclude that the advent of subduction-976 related volcanism, the appearance of volcano-hosted massive sulfide deposits, and the 977 associated concentrations of relatively rare elements were essential for the dramatic 978 expansion of hydrothermal mineral diversity.

979

980 Stage 4b. Highly evolved igneous rocks Significant mineral diversity is associated 981 with the concentration of rare, incompatible elements in late-stage magmas. "Complex" 982 granitic pegmatites, agpaitic and miaskitic pegmatites, carbonatites and related carbon-983 rich magmas, and layered igneous intrusions with concentrations of PGE elements 984 account for 1476 mineral species, 662 of them unique to those compositionally 985 idiosyncratic environments. All of these lithologies were present by the end of the 986 Archean Eon; however, the timing of the most ancient of these formations is uncertain, as 987 their relative rarity, coupled with erosional loss, results in an imperfect record.

988 [#34 - Complex granite pegmatites]: We tabulate 564 mineral species that are found in 989 granite pegmatites, with 238 rare species known only from that environment. The earliest 990 documented occurrences of pegmatite minerals with essential Li and Be are from 991 Mesoarchean formations (~3.1 to 3.0 Ga), including the Barberton greenstone belt, South 992 Africa, and the Zishineni pegmatite, Swaziland (Tkachev 2011; Grew et al. 2017, 2019), 993 while the oldest known complex pegmatite with Cs mineralization is the 2.64 Ga Tanco 994 Mine in Manitoba, Canada (Černý 2005; London 2008). These earliest known examples 995 of complex granitic pegmatites may reflect the extreme concentration of rare elements 996 that was facilitated by subduction and consequent fluid-rock interactions in large volumes 997 of the crust and upper mantle, and thus only possible < 3 Ga.

998 [#35 – Ultra-alkali rocks]: Pegmatites from ultra-alkaline agpaitic and miaskitic rocks
999 display even greater mineral diversity than granite pegmatites, with 726 recorded species
1000 (Marks and Markl 2017; Mikhailova et al. 2019), including 377 minerals that are unique
1001 to agpaitic formations. Much of this mineralogical diversity results from the unusual

1002 concentration of Be, Ti, Y, Zr, Nb, and rare earth elements in these alkali-rich, silica-poor 1003 rocks.

1004 [#36 - Carbonatites]: Carbonatites, kimberlites, and related carbonate-bearing igneous 1005 rocks (Mitchell 1986; Jones et al. 2013) hold 291 distinctive minerals, including dozens 1006 of rare carbonate and phosphate species. These intriguing lithologies have been present 1007 since at least the Mesoarchean Era. Among the oldest known occurrences are the 3.01 Ga 1008 Tupertalik carbonatite from Greenland (Downes et al. 2012), and the 2.6 Ga Siilinjarvi 1009 carbonatite from Finland (Patchett et al. 1982; Woolley and Kjarsgaard 2008), whereas 1010 the Ol Doinyo Lengai volcano in Tanzania continues to erupt low-viscosity carbonate 1011 lavas.

1012 [#37 – Layered igneous intrusions]: In spite of their affinity to other mafic/ultramafic 1013 igneous rocks, we consider layered igneous intrusions to be a separate paragenetic 1014 environment because of their unusual homogeneous aerial extent, their predominantly 1015 Precambrian ages, and the common occurrence of metal-enriched layers with more than 1016 100 PGE and Cr-Ti-Fe oxide mineral species, including a number of phases reported 1017 from no other source (Naldrett et al. 1987; Maier and Groves 2011; Zientek 2012; 1018 O'Driscoll and VanTongeren 2017). The oldest known PGE deposit is the 3.2 Ga Baula 1019 deposit in India (Augé et al. 1993), with several Neoarchean and Paleoproterozoic 1020 examples recorded from localities in Africa and North America (Maier and Groves 1021 2011).

1022

1023 <u>Stage 5. Initiation of plate tectonics</u> Crustal recycling via plate tectonics was a 1024 significant driver of mineral diversification, largely as a consequence of mineralization

associated with orogenesis, as well as the first subaerial exposure of otherwise hidden
high-*P*-*T* lithologies. We adopt the prevailing view that modern-style, subduction-driven
plate tectonics was well established prior to 3 Ga, as manifest in the Wilson cycle and
episodes of supercontinent assembly (Huston et al. 2010; Nance et al. 2014; Hazen et al.
2014), with significant earlier crustal recycling perhaps before 4 Ga (e.g., Condie and
Pease 2008; Korenaga 2021).

1031 [#38 – Ophiolites]: The stranding of ophiolites, which are highly altered sequences of 1032 mafic and ultramafic rocks from the deep oceanic lithosphere, provides an important 1033 glimpse of the mineralogy and petrology of the crust-mantle boundary (Moores 2002; 1034 Dilek 2003; Kusky 2004). Ophiolites, which hold at least 108 mineral species, are in 1035 many respects similar to ocean floor rocks altered by serpentinization. However, their 1036 unique subaerial settings and exposures of mantle lithologies warrants a separate 1037 paragenetic category. Furthermore, the mineralogically unique Luobusha ophiolite from 1038 the Shannan Prefecture of Tibet is of special interest for a remarkable suite of dozens of 1039 UHP (e.g., diamond and moissanite) and highly-reduced phases, including native 1040 elements (Al, Cr, Cu, Fe, Ti, W), carbides, nitrides, and phosphides (Bai et al. 2011). In 1041 addition, the Luobusha occurrence features many PGE metal alloys (including 1042 approximately 30 as yet undescribed metal phases), in association with chromitite zones 1043 that are reminiscent of assemblages in layered intrusions [#37]. We include these diverse, 1044 rare minerals under ophiolites, though details of their paragenesis remain obscure. 1045 Debates persist regarding the oldest ophiolite. Furnes et al. (2007) suggested that an

altered dike complex in Greenland's 3.8 Ga Isua supracrustal belt represents a "vestige"

1047 of oceanic crust, though this interpretation was questioned by Nutman and Friend (2007).

The next oldest claim, the 2.5 Ga Dongwanzi ophiolite complex from China (Kusky et al. 2001) has also been debated (Zhai et al. 2002; Kusky and Zhai 2012); however, descriptions of several altered Neoarchean examples support an origin by at least 2.5 Ga (Kusky 2004), providing further evidence that plate tectonics was well established in the late Archean Eon.

1053 [#39 - High-P metamorphism]: Plate tectonic processes have also led to the exposure 1054 of metamorphic rocks from environments with unusually low geothermal gradients, 1055 estimated to be < 10 °C/km – conditions only possible during relatively rapid subduction 1056 of crustal rocks to great depths, with subsequent buoyant uplift (Chopin 1984; Hacker 1057 2006; Palin and White 2016; Zheng and Chen 2017). These rocks include blueschist 1058 facies with glaucophane, jadeite, and/or lawsonite (formed at depths to 30 km); eclogite 1059 with pyrope and omphacite (> 45 km); and so-called "ultra-high pressure" (UHP) 1060 formations, featuring the dense coesite form of SiO<sub>2</sub> (> 80 km). The oldest known 1061 examples of these high-pressure lithologies are from the Neoarchean Era (~2.8 Ga), 1062 which coincides with models of plate tectonics commencing at  $\sim 3$  Ga (Jahn et al. 2001; 1063 Brown 2007). We list 70 associated minerals, of which a dozen species, including 1064 trinepheline (NaAlSiO<sub>4</sub>), kokchetavite (KAlSi<sub>3</sub>O<sub>8</sub>), and barioperovskite (BaTiO<sub>3</sub>), are 1065 unique to these high-pressure metamorphic rocks.

1066 [#40 – Regional metamorphism]: Regional metamorphism associated with burial, 1067 alteration, and uplift of thick accumulations of sediments, volcanic rocks, and intrusive 1068 igneous rocks led to 319 mineral species. Most regional metamorphic rocks formed at 1069 depth under an average geothermal gradient of > 15 °C/km to  $\sim$ 30 °C/km (Vernon 2008; 1070 Philpotts & Ague 2009). These rocks are usually subdivided into facies based on the

1071 appearance of new minerals with increasing P and T: greenschist (typically with chlorite, 1072 serpentine, and epidote), amphibolite (amphibole and plagioclase), and granulite 1073 (pyroxene and plagioclase). We lump all of these regional metamorphic facies in this 1074 study, while acknowledging that further subdivision of metamorphic lithologies may be 1075 warranted in future studies.

1076 [#41 – Mantle metasomatism]: Mantle metasomatism (Luth 2003; O'Reilly and 1077 Griffin 2012) is a high-pressure hydrothermal metamorphic process that alters the 1078 chemistry of existing ultramafic minerals and produces new phases through interactions 1079 with deep C-O-H fluids (Manning and Frezzotti 2020). We identify 16 oxide and silicate 1080 mineral species formed through mantle metasomatism, all but 1 of which (nixonite,

1081 Na<sub>2</sub>Ti<sub>6</sub>O<sub>13</sub>; Anzolini et al. 2019) are well known from other environments.

1082 [#42 – Seafloor nodules]: Seafloor nodules are enigmatic Mn- and Fe-rich concretions 1083 with significant Ni, Cu, and Co content that form extensive (and potentially economic) 1084 deposits on some regions of the ocean floor. While these nodules may be authigenic and 1085 thus a subset of mode [#17], their mineralogy and morphology appear to be distinct. If, as 1086 some contend, they form through microbial mineralization (Lysyuk 2008; Blöthe et al. 1087 2015) or through precipitation in an oxic environment (Fike et al. 2006; Wegorzewski 1088 and Kuhn 2014), then this paragenetic category should be moved to Stage 6 or Stage 7. 1089 If, on the other hand, they form by the release and reprecipitation of metals from 1090 ultramafic rocks and/or from hydrothermal vents (Hlawatsch et al. 2002), then metal-rich 1091 nodules may have appeared as early as Stage 3 or Stage 4. We provisionally assign 1092 seafloor nodules to Stage 5, based on the assumption that Mn release increased at

spreading centers following the crustal recycling of plate tectonics. We list 15 mineralsfrom Mn nodules, 11 of which are Mn or Fe oxides/hydroxides.

1095 [#43 – Shear-induced minerals]: Minerals produced by significant shear strain 1096 represent a distinct paragenetic mode, most commonly associated with polished fault 1097 surfaces known as slickensides (Passchier and Trouw 2005; Trouw et al. 2009). The closely related term "mylonite" relates to an altered zone of minerals recrystallized 1098 1099 through shearing. We list 9 minerals produced in shear zones, 8 of which are common 1100 oxides and silicates, that are reported to form anew in silicate rocks through this process. 1101 It is difficult to assess when shear-generated minerals first occurred. We associate them 1102 primarily with plate tectonic processes, but some examples must have occurred as early 1103 as the Hadean Eon. On the other hand, of special interest is the rare chlorite group 1104 mineral, donbassite [Al<sub>2</sub>(Si<sub>3</sub>Al)O<sub>10</sub>(OH)<sub>2</sub>·Al<sub>2.33</sub>(OH)<sub>6</sub>], which has been reported from 1105 slickensides of coal (Anthony et al. 1990-2003), and therefore must have formed at < 3501106 Ma.

1107

1108 Stage 6. Anoxic biosphere [#44]: The rise of microbial life ~4.0 billion years ago appears 1109 to have had little initial impact on mineral diversity. However, some microbes learned to 1110 exploit minerals, using carbonates and phosphates as sturdy stromatolitic habitats 1111 (Chauhan 1979; Burnett and Riggs 1990; Cook and Shergold 1990; Grotzinger and Knoll 1112 1999; Van Kranendonk et al. 2003), as well as employing iron (and possibly other 1113 transition metal) oxide and sulfide phases as reliable sources of redox energy (Lowell et 1114 al. 2009; Ishibashi et al. 2015). We propose a conservative list of 11 microbially-1115 mediated minerals, all of them common phases found in five or more paragenetic

1116 environments, that are thought to have formed in anoxic environments prior to the rise of 1117 atmosphere that characterized Stage 7. Numerous instances of Archean minerals 1118 precipitated through microbial action have been documented (Grotzinger and Knoll 1999; 1119 Konhauser et al. 2002; Frankel and Bazylinski 2003; Schulz and Schulz 2005; Southam 1120 and Saunders 2005; Lepot et al. 2008). Unambiguous occurrences of microbial carbonate 1121 stromatolites occur as early as 3.5 Ga in rocks of the Pilbara craton in Western Australia 1122 (Van Kranendonk et al. 2003; Allwood et al. 2006), pointing to calcite, dolomite, and 1123 siderite as likely early biominerals. Similarly, biogenic phosphatic stromatolites 1124 (Banerjee 1971; Schulz and Schulz 2005) and iron formations (Anbar and Holland 1992; 1125 Akai et al. 1999) have been described from multiple localities as early as the Neoarchean 1126 Era.

1127

1128 <u>Stage 7. Great Oxidation Event</u> The single most significant factor in Earth's mineral 1129 diversification was the Neoarchean/Paleoproterozoic rise of atmospheric oxygen, which 1130 we estimate led to the introduction of more than 2300 new mineral species. However, at 1131 least two significant uncertainties relate to the timing and nature of the formation of 1132 Earth's diverse oxidized mineral species.

A persistent question relates to the timing of atmospheric oxygenation (Canfield 2014; Lyons et al. 2014). A number of early Earth scenarios point to the Great Oxidation Event as a prolonged period of atmospheric change, often modeled as occurring after 2.4 Ga, when global-scale oxygenic photosynthesis became well established. Important evidence for this change in atmospheric composition is found in the sulfur isotope record, notably in the disappearance of mass-independent S isotope fractionation in near-surface sulfur-

1139 bearing minerals (Farquhar et al. 2000, 2001, 2007). This rather abrupt change in the 1140 character of S isotope ratios is thought to have occurred owing to formation of a UV-1141 blocking ozone layer. However, other researchers point to intervals or oases of elevated 1142 atmospheric oxygen as early as 3.1 Ga based on pulses of redox-sensitive trace elements 1143 in Archean sediments (e.g., Anbar et al. 2007), as well as the postulated early appearance 1144 of microbial oxygen-utilizing enzymes (Jabłońska and Tawfik 2021). Those results 1145 suggest that a subset of oxidized mineral species might have formed prior to 2.4 Ga, 1146 during the Neoarchean Era.

1147 A second uncertainty relates to the extent of Archean *abiotic* processes that might 1148 have produced relatively oxidized mineral species. In particular, as noted above (see [#28]), UV-photooxidation of  $Fe^{2+}$  to  $Fe^{3+}$ , followed by production of sulfates and other 1149 1150 relatively oxidized minerals, may have been a ubiquitous process in near-surface waters 1151 prior to the establishment of a UV-blocking ozone layer. If this UV-mediated process was 1152 commonplace prior to the rise of atmospheric oxygen, then some of the relatively 1153 oxidized phases we ascribed to post-GOE biological O<sub>2</sub> production may have emerged 1154 much earlier through abiotic chemistry. With those caveats in mind, we consider three 1155 important mineral-forming environments that were strongly influenced by atmospheric 1156 oxygenation.

1157 [#45 – Volcanic fumarole minerals, oxidized]: Volcanic fumaroles produce a 1158 remarkable variety of more than 420 relatively oxidized mineral species, 168 of which 1159 are unique to volcanic vents. We suggest that these minerals could only have been 1160 precipitated after the GOE (e.g., Vergasova and Filatov 2016). Accordingly, we tabulate 1161 253 fumarolic sulfates, arsenates, selenates, and antimonates, as well as 171 other vent

1162 minerals, including carbonates, borates, nitrates, phosphates, and other phases 1163 incorporating a range of transition metals (V, Cr, Zn, Mo, W), halogens, and ammonia. 1164 We include intriguing pneumatolytic minerals [e.g., itelmenite, Na<sub>2</sub>CuMg<sub>2</sub>(SO<sub>4</sub>)<sub>4</sub>; 1165 Nazarchuk et al. 2018] that are thought to form by reactions of prior minerals with 1166 fumarolic gases (Krivovichev et al. 2013). 1167 [#46 – Oxidized hydrothermal minerals]: The formation of more than 2000 secondary 1168 minerals by near-surface "weathering," primarily oxidation and hydration, is by far the 1169 greatest single contributor to Earth's mineral diversity. The majority of these minerals 1170 form in direct contact with the oxygenated atmosphere (see [#47]). However, we 1171 distinguish 52 oxidized secondary minerals, primarily sulfates and arsenates, that are 1172 described as forming in hydrothermal environments. Thus, for example, mansfieldite 1173 [AlAsO<sub>4</sub>'2H<sub>2</sub>O] is reported by Anthony et al. (2003) to be "a hydrothermal product in altered and mineralized andesitic pyroclastic rocks" (and thus included in [#46]), in 1174 contrast to its Fe-bearing isomorph scorodite [Fe<sup>3+</sup>AsO<sub>4</sub>'2H<sub>2</sub>O], which is described as "a 1175 1176 secondary mineral formed by oxidation of As-bearing sulfides" and therefore assigned to 1177 [#47]. Similarly, cesanite [Ca<sub>2</sub>Na<sub>3</sub>[(OH)(SO<sub>4</sub>)<sub>3</sub>] (Cavarretta et al. 1981) and grandaite [Sr<sub>2</sub>Al(AsO<sub>4</sub>)<sub>2</sub>(OH)] (Cámara et al. 2014) are described as hydrothermal minerals, 1178 1179 whereas compositionally similar antofagastaite [Na<sub>2</sub>Ca(SO<sub>4</sub>)<sub>2</sub>·1.5H<sub>2</sub>O] (Pekov et al. 1180 2019) and arsenogoyazite [SrAl<sub>3</sub>(AsO<sub>4</sub>)(AsO<sub>3</sub>OH)(OH)<sub>6</sub>] (Walenta and Dunn 1984) are 1181 presented as secondary oxidized phases. We have adopted the reported paragenetic 1182 modes of these oxidized minerals - hydrothermal versus secondary - while recognizing

1183 that a continuum exists for secondary oxidized minerals formed in near-surface 1184 environments.

1185 [#47 – Oxidative weathering]: We catalog 1998 secondary minerals formed in lowtemperature weathering environments in contact with a post-GOE oxygenated 1186 1187 atmosphere. Given this large number of secondary, oxidized minerals (35 % of all IMA-1188 approved species), we subdivide these phases into 9 overlapping compositional groups. 1189 Near-surface hydration of prior minerals is the largest subset, with 1649 species (82.5 % 1190 of [#47]). Other subsets include sulfates and sulfites (409 species); carbonates, 1191 phosphates, borates, and nitrates (577); arsenates, antimonates, selenates, and bismuthinates (505); vanadates, chromates, and manganates (372); uranyl (U<sup>6+</sup>) minerals 1192 1193 (236); and halogen-bearing minerals (236). A significant subset (316 species) of these 1194 weathering products are dehydrated, which may point to multiple stages of subaerial 1195 weathering that included both hydration and dehydration reactions. Finally, we include 1196 28 minerals known to form through the terrestrial weathering of meteorites [#47i], 1197 including 10 Fe, Ni, and/or Cr phases only known from the alteration of iron meteorites 1198 (Rubin and Ma 2021).

An intriguing finding is that 1119 of these minerals – 20 % of Earth's total mineral diversity – are known to form only through paragenetic mode [#47]. These species formed exclusively by oxidative weathering are not equally divided among the 9 compositional groups (Table 1). Approximately 45 % of sulfates and 51 % of halogenbearing phases in [#47] are only known as weathering phases, whereas 73 % of uranyl minerals are uniquely found in those near-surface oxidized environments.

1205

Stage 8. Intermediate Ocean A billion years of the Proterozoic Eon (~1.8 to 0.8 Ga) was
evidently a period of gradual oxygenation of the "intermediate ocean" (Anbar and Knoll
2002), coupled with reduced mineralization (Hazen et al. 2008; Bradley 2011; Hazen et
al. 2014; Liu et al. 2019). Sometimes called the "boring billion" (Brasier 1998, 2012;
Buick et al. 1995), this extended Proterozoic interval appears to have seen no new
paragenetic processes.

1212

1213 Stage 9. Snowball Earth The end of the Neoproterozoic Era (~0.8 to 0.6 Ga) featured at 1214 least three major episodes of global glaciation, when ice was the dominant mineral over 1215 most of Earth's surface from poles to Equator (Kirschvink 1992; Hoffman et al. 1998). 1216 Each of these "snowball Earth" events saw the cessation of significant continental 1217 weathering, while atmospheric carbon dioxide increased through the contributions of 1218 continuous volcanic emissions – changes that in turn led to an exaggerated greenhouse 1219 effect, global warming, and episodes of relatively rapid loss of ice cover. These 1220 intervening "hothouse Earth" periods must have seen enhanced surface weathering, 1221 notably accompanied by carbonate formation. However, no new paragenetic processes 1222 are associated with Stage 9 of mineral evolution.

1223

Stage 10a. Neoproterozoic oxygenation and the Phanerozoic biosphere The Phanerozoic
 Eon (< 541 Ma) has been characterized by significant mineralogical novelty, with almost</li>
 600 new minerals, notably those arising from biological (including anthropogenic)
 processes. Distinctions have been proposed by Perry et al. (2007) between "biominerals"
 (produced by living cells) and "organominerals" (derived from organic molecules, often

taphonomic, but not directly produced by cellular activity). We further recognize
thousands of biologically-mediated mineral species that form indirectly as a consequence
of environmental changes caused by cellular activity – most significantly oxygenic
photosynthesis.

1233 [#48 – Soil minerals]: Research in paleopedology – the study of ancient soils or 1234 "paleosols" – reveals that Earth's terrestrial surface has been coated by fine-grained 1235 detrital material for much of its history (Johnson and Watson-Stegner 1987; Holland and 1236 Rye 1997; Rye and Holland 1998; Retallack 2001). However, prior to the emergence of 1237 land plants at  $\sim 0.4$  Ga soils tended to be thin and mineralogically tied to their host 1238 lithologies and subaerial weathering. Soils became much deeper and more complex 1239 mineral-producing zones in concert with the development of root systems, most notably 1240 following the evolution of symbiotic mycorrhizal fungi (Remy et al. 1994; Paris et al. 1241 1995, 1996; Ueshima and Tazaki 1998). The terrestrial formation of clay minerals in 1242 soils, in particular, is estimated to have increased by an order of magnitude with the 1243 advent of root systems (Schwartzman and Volk 1989; Barker et al. 1998). We list 71 soil 1244 minerals that we attribute to Phanerozoic processes, including more than a dozen Al-rich 1245 hydrous minerals from leaching zones in laterites and bauxites (e.g., Helgren and Butzer 1246 1977; Hill et al. 2000).

1247 [#49 – *Biominerals*]: Phanerozoic biomineralization encompasses a fascinating 1248 collection of 77 minerals formed by a variety of metabolic processes. Much attention has 1249 focused on the biological nano-sculpting of mineral-organic composite materials that play 1250 structural roles in organisms. Examples include hydroxylapatite and fluorapatite in 1251 directed biomineralization of vertebrates (teeth and bones), inarticulate brachiopod shells,

1252 and stinging nettles; the calcite or aragonite forms of CaCO<sub>3</sub>, for example in corals,

1253 mollusks, and foraminifera; and of silica employed by such diverse organisms as diatoms, 1254 sponges, and spinifex grass (Lowenstam and Weiner 1989; Weiner and Wagner 1998; 1255 Dove et al. 2003; Dove 2010; Aparicio and Ginebra 2016; Ensikat et al. 2016; Kattimani 1256 et al. 2016; Endo et al. 2018). Lichtenegger et al. (2002) also reported an unusual 1257 occurrence of atacamite [Cu<sub>2</sub>(OH)<sub>3</sub>Cl] in jaws of the bloodworm, *Glycera dibranchiate*. 1258 Microbially-induced oxidation-reduction reactions have resulted in a variety of mineralization pathways, including reduction of aqueous  $U^{6+}$  to uraninite (UO<sub>2</sub>; Suzuki 1259 1260 and Banfield 1999; Fayek et al. 2005; Hazen et al. 2009; Wufuer et al. 2017); formation 1261 of autunite group uranium phosphates (Liang et al. 2015; Beazley et al. 2017); oxidation 1262 or reduction of iron oxide, sulfide, or sulfates (Pósfai et al. 1998; Akai et al. 1999; Coker 1263 et al. 2008; Boyd and Druschel 2013; Berg et al. 2014, 2020), notably to form internal 1264 "magnetosomes" of magnetite or greigite for microbial navigation (Chang and 1265 Kirschvink 1989; Lins et al. 2007; Pósfai et al. 2013; Uebe and Schüler 2016); and 1266 manganese redox reactions to form a number of species [e.g., vernadite, todorokite, 1267 birnessite, rhodochrosite; Carmichael and Bräuer 2015; Kelm et al. 2015; Li et al. 2016; 1268 Zerfaß et al. 2019; Galezowski et al. 2020; Lu et al. 2021). Some researchers have suggested that seafloor manganese nodules form, at least in part, through such 1269 1270 biochemical processes (Lysyuk 2008; Blöthe et al. 2015). Microbes have also been 1271 invoked in the precipitation of gold (Au; Reith et al. 2006, 2009; Johnston et al. 2013; 1272 Sanval and Shuster 2021), copper carbonate (Li and Gadd 2017), anatase (TiO<sub>2</sub>; Ping et 1283

1284 in the past ~300 million years (Saxby 2000; Schweinfurth 2016). As might be expected,

been identified from coal and/or oil shale minerals, all of which must have been formed

1285 several of these phases are crystallized hydrocarbons, such as idrialite (C<sub>22</sub>H<sub>14</sub>) and

1286 evenkite (C<sub>23</sub>H<sub>48</sub>), or ammonia-bearing minerals, including letovicite [(NH<sub>4</sub>)<sub>3</sub>H(SO<sub>4</sub>)<sub>2</sub>]

1287 and godovikovite  $[(NH_4)Al(SO_4)_2]$ . Of note is abelsonite  $(NiC_{31}H_{32}N_4; Milton et al.$ 

1288 1978), the only known geoporphyrin mineral, from the Green River oil shale of Utah.

1289 Coal and oil shale also contain a wide range of fine-grained clay minerals,

1290 oxide/hydroxides, and hydrous sulfates, as well as minerals incorporating relatively rare

1291 metals that are known to concentrate in hydrocarbon-rich deposits, for example

1292 strangersite (SnGeS<sub>3</sub>), patronite (VS<sub>4</sub>), Laphamite (As<sub>2</sub>Se<sub>3</sub>), cadmoindite (CdIn<sub>2</sub>S<sub>4</sub>),

1293 moschelite (HgI), tugarinovite (MoO<sub>2</sub>), bismuthinite (Bi<sub>2</sub>S<sub>3</sub>), and stibnite (Sb<sub>2</sub>S<sub>3</sub>). Note

<sup>1273</sup> al. 2016), vanadium minerals (Carpentier et al. 2003), and polymorphs of sulfur (Douglas

<sup>1274</sup> and Yang 2002; Gleeson et al. 2011; Cron et al. 2019).

<sup>1275</sup> Biochemical processes also lead to a variety of incidental minerals (Yoder 2002; 1276 Hazen et al. 2008, their Table 3), as manifest in dozens of human-precipitated minerals, 1277 for example in kidney stones (Rogers 1997; Giannossi et al. 2009) and gallstones (Endo 1278 1962; Dumitrascu et al. 1984). Finally, the only example of a mineral known to form 1279 exclusively by biomineralization is hazenite [KNaMg<sub>2</sub>(PO<sub>4</sub>)<sub>2</sub>·14H<sub>2</sub>O; Yang et al. 2011], 1280 which is excreted by microbes living in the alkali- and P-saturated hypersaline waters of 1281 Mono Lake, California. 1282 [#50 - Coal minerals]: An eclectic assortment of 273 different mineral species has

1294 that we also list 234 minerals produced in coal mine fires as a separate anthropogenic 1295 paragenetic process [#54]. However, we cannot rule out possible overlaps between those 1296 phases and the varied coal minerals listed here [#50]. 1297 [#51 - Pyrometamorphic minerals]: Pyrometamorphic minerals of the Hatrurim

1298 Formation in the Middle East (Israel and Jordan) include a fascinating suite of 128 1299 species that formed when natural ignition of near-surface hydrocarbon deposits 1300 (presumably both natural gas and petroleum) subjected sediments to temperatures in 1301 excess of 1000 °C (Gross 1977; Grapes 2006; Sokol et al. 2019). Two-thirds of these 1302 minerals are Ca-bearing, some closely resembling phases in cement "clinkers," including 1303 such unusual silicates as larnite (Ca<sub>2</sub>SiO<sub>4</sub>), hatrurite (Ca<sub>3</sub>SiO<sub>5</sub>), grossite (CaAl<sub>4</sub>O<sub>7</sub>), and 1304 rankinite (Ca<sub>3</sub>Si<sub>2</sub>O<sub>7</sub>). The Hatrurim Formation also hosts unusual Ca sulfides such as 1305 oldhamite (CaS) and dzierzanowskite (CaCuS<sub>2</sub>), and other phases such as vapnikite 1306  $(Ca_3UO_6)$ , chromatite  $(CaCrO_4)$ , and lakargite  $[Ca(Zr,Sn)O_3]$ . The Hatrurim Formation 1307 is mineralogically unique, but pyrometamorphism has been a persistent phenomenon 1308 throughout Earth history, with high-T, low-P alteration occurring in lava xenoliths [#9], 1309 during lightning strikes [#29], and as a consequence of anthropogenic coal mine fires 1310 [#54] and smelting [#56]. 1311 [#52 – Guano/urine minerals]: A curious assortment of 72 mineral species are derived 1312 from the urine and guano of birds and bats – minerals preserved primarily in caves and on

islands in arid environments. A few of these minerals form directly from excreta, including guanine  $[C_5H_3(NH_2)N_4O]$ , urea  $[CO(NH_2)_2]$ , and uricite  $(C_5H_4N_4O_3)$ . Other minerals result from reactions between urine and prior minerals; spheniscidite

 $[(NH_4)Fe^{3+}_{2}(PO_4)_2(OH)^{2}H_2O]$  (Wilson and Bain 1986), a fascinating example, is 1316 formed when the urine of penguins (order Sphenisciformes, hence the mineral name) 1317 1318 reacts with layer silicates in soils beneath a rookery on Elephant Island in the British 1319 Antarctic Territory. 1320 [#53 – Taphonomic minerals]: We identify 117 other biologically-derived minerals 1321 with presumed taphonomic origins, mostly formed during the decay and/or fossilization 1322 of wood and other plant material (Behrensmeyer et al. 2000, Mustoe 2018). Plant decay 1323 leads to several distinctive organic minerals (e.g., Garvie 2003), notably oxalates, such as 1324 weddellite  $(CaC_2O_4 2H_2O)$  and uroxite  $[(UO_2)_2(C_2O_4)(OH)_2(H_2O)_2H_2O]$ , and acetates, including paceite [CaCu(CH<sub>3</sub>COO)<sub>4</sub>·6H<sub>2</sub>O] and hoganite [Cu(CH<sub>3</sub>COO)<sub>2</sub>·H<sub>2</sub>O]. Decay 1325 1326 may also release ammonia, which plays a role in producing 20 taphonomic minerals that 1327 by reaction with NH<sub>4</sub>-bearing fluids, for example ammoniozippeite form 1328  $[(NH_4)_2(UO_2)_2(SO_4)O_2 \cdot H_2O]$  and ambrinoite  $[(K,NH_4)_2(As,Sb)_6(Sb,As)_2S_{13} \cdot H_2O]$ . An 1329 intriguing geochemical consequence of wood fossilization is the concentration of 1330 vanadium-rich fluids and formation of at least 14 minerals, including vanoxite  $(V_{4}^{4+}V_{2}^{5+}O_{13}\cdot 8H_2O)$  and dreyerite (Bi<sup>3+</sup>V<sub>2</sub>O<sub>4</sub>). Burial of organics also commonly 1331 results in subsurface reduction reactions, including  $U^{6+}$  to  $U^{4+}$  [e.g., coffinite, 1332  $(U^{4+}SiO_4 \cdot nH_2O)$ ] and  $V^{5+}$  to  $V^{4+}$  or  $V^{3+}$  [berdesinskiite,  $(V^{3+}_2TiO_5)$ ]. 1333

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1335 <u>Stage 10b. Anthropogenic minerals</u> The most recent (and ongoing) stage of mineral 1336 evolution, spanning the past few thousand years but intensifying since the Industrial

Revolution, encompasses a wide range of more than 600 human-mediated minerals 1337 1338 (Catheart 2011; Corcoran et al. 2014; Hazen and Zalaziewski 2019; Zeng et al. 2020). 1339 Hazen et al. (2017) reviewed the mineralogy of the so-called "Anthropocene Epoch," 1340 tabulating 208 mineral species approved by the IMA-CNMNC that occur principally or 1341 exclusively through human activities. The present more inclusive survey of the 1342 paragenetic modes of all minerals points to more than 600 mineral species that may form 1343 as byproducts of human industry, though most of those phases occur by natural 1344 processes, as well.

1345 [#54 - Mine fire minerals]: Of more than 500 anthropogenic minerals arising as a 1346 consequence of mining activities, we tabulate 234 minerals that form in coal mine fires, 1347 both by thermal metamorphism of prior phases and by condensation of new species 1348 (Lapham et al. 1980; Grapes 2006; Kruszewski 2013; Stracher et al. 2015). All but 6 of 1349 these minerals are also known to form by other processes, mostly by pyrometamorphism 1350 [#51] and at volcanic fumaroles ([#11] or [#45]). Those six species unique to coal fires 1351 include two intriguing molecular crystals: freitalite  $(C_{14}H_{10})$  is composed of the 1352 polycyclic aromatic hydrocarbon, anthracene, which is a major component of coal tar; 1353 acetamide (CH<sub>3</sub>CONH<sub>2</sub>) is the simplest amide (NH<sub>2</sub>) compound, which has important 1354 uses in the plastics industry.

1355 [#55 – *Mine-derived minerals*]: Mining activities lead to a wealth of new minerals, 1356 including those formed by weathering of mine dump phases, formation on mine tunnel 1357 walls, precipitation from acid mine drainage systems, and alteration associated with mine 1358 timbers and artifacts (Hazen et al. 2017). We recognize 264 of these mining-derived 1359 minerals, all but 5 of which also form in natural environments. In particular, 227 of these

1360 species (86 %) also arise through natural near-surface oxidative weathering processes 1361 [#47]. We acknowledge that it is often difficult to discriminate between phases that arose 1362 exclusively because of human mining activities and those that may have formed naturally 1363 through alteration of surface exposures. 1364 [#56 - Slag minerals]: The alteration of minerals through smelting, and in some 1365 instances the subsequent alteration of slag, results in 143 mineral species. This high-1366 temperature processing represents a form of pyrometamorphism (Grapes 2006; Sueoka 1367 and Sakakibara 2013; Kierczak and Ettler 2021), and it produces a similar suite of phases 1368 to other pyrometamorphic mechanisms (e.g., [#9], [#11], [#29], and [#54]). We did not

1369 identify any mineral species that occurs exclusively in slag.

1370 [#57 - Other human-mediated minerals]: Finally, we record an eclectic list of 49 1371 mineral species produced via other human-mediated processes. These phases include 1372 corrosion products of bronze, lead, and other ancient archaeological artifacts, notably Pb-1373 and Cu-bearing phases that have been found associated with the ancient slag deposits at 1374 Laurium, Greece [e.g., atacamite, Cu<sub>2</sub>Cl(OH)<sub>3</sub>, and laurionite, PbCl(OH); Smith and 1375 Prior 1899]; precipitation of phases in geothermal piping and other mineral-rich 1376 subsurface water systems, such as nasinite [Na2B5O8(OH)·2H2O] and santite 1377 [KB<sub>5</sub>O<sub>6</sub>(OH)<sub>4</sub>·2H<sub>2</sub>O] from Tuscany (Merlino and Sartori 1970); alteration of archived 1378 drill cores or minerals stored in museum drawers, including calclacite 1379 [Ca(CH<sub>3</sub>COO)Cl·5H<sub>2</sub>O; coalingite Van Tassel 1945] and [Mg<sub>10</sub>Fe<sup>3+</sup><sub>2</sub>(CO<sub>3</sub>)(OH)<sub>24</sub>·2H<sub>2</sub>O; Mumpton et al. 1965]; minerals suspected of forming 1380 by grinding [itoite, Pb<sub>3</sub>GeO<sub>2</sub>(SO<sub>4</sub>)<sub>2</sub>(OH)<sub>2</sub>; Anthony et al. 2003]; fertilizer byproducts 1381

1382 such as metavariscite [Al(PO<sub>4</sub>)·2H<sub>2</sub>O; Anthony et al. 2003]; and alteration of discarded

lead batteries (e.g., cerussite, PbCO<sub>3</sub>). However, none of these phases is unique to 1383 1384 anthropogenic activities, in accord with rules of the IMA-CNMNC (e.g., Nickel and 1385 Grice 1998). Other minerals that might join this list in the future include alteration phases 1386 of discarded lithium batteries, semiconductors and phosphors employed in electronics, 1387 and other technological materials (e.g., Galezowski et al. 2020). 1388 In addition, purposeful human productions of countless thousands of synthetic phases 1389 not considered as minerals by the IMA-CNMNC have dramatically increased the 1390 diversity and distribution of mineral-like compounds at or near Earth's surface. Many of 1391 these compounds, including building materials, semiconductors, laser crystals, specialty 1392 alloys, synthetic gemstones, and possibly even plastic debris, are likely to persist for 1393 millions of years in the geologic record, thus providing a clear sedimentary horizon that 1394 marks the Anthropocene Epoch (Catheart 2011; Corcoran et al. 2014; Hazen et al. 2017; 1395 Hazen and Zalaziewski 2019).

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## **IMPLICATIONS**

This study of the paragenetic modes of minerals points to several important trends in mineral evolution, while raising significant unanswered questions. Our preliminary database records 5659 minerals approved by the IMA-CNMNC assigned to 57 paragenetic modes, resulting in 10,556 combinations of a mineral species and a paragenetic mode (Supplementary Table 1). This open-access database is an evolving resource; we welcome additions and corrections.

1404 Several conclusions can be drawn from these data:

• Water is the principal driver of mineral diversity: Of the 5659 minerals examined,

4583 (81.0 %) either incorporate essential OH and/or H<sub>2</sub>O, accounting for 3150 of 1406 1407 5659 mineral species (55.7 %), or they are anhydrous phases that were formed 1408 through the action of aqueous fluids, for example, by dehydration, evaporation, or 1409 precipitation from a hydrothermal solution (1433 species). No other physical or 1410 chemical factor comes close to this dominant role of water in creating mineral 1411 diversity. Near-surface hydration and weathering represent obvious major factors in 1412 this variety, resulting in more than 2000 species. We suggest that subduction and the 1413 deep-water cycle is comparable in its influence on Earth's mineralogy as a 1414 consequence of the selection and concentration of rare elements through water-rock 1415 interactions in large volumes of the upper mantle and crust. The sharp contrast 1416 between Earth's large complement of minerals and the relative mineralogical 1417 parsimony of the Moon and Mercury, as well as the modest diversity found on Mars, 1418 stems from differing influences of water.

1419 Biology plays an important role in Earth's mineral diversity (but not quite as much 1420 as previously suggested): We find that almost half of Earth's minerals -2707 of 1421 5659 species (47.8 %) – are known to form as a result of biological influences. More 1422 than 2000 of these minerals occur as a consequence of near-surface oxidative 1423 weathering/alteration of prior phases, which is the second most important factor in 1424 Earth's mineral diversity after water. Many minerals also form as coal-related and 1425 other taphonomic minerals (597 species), as well as anthropogenic minerals, for 1426 example as byproducts of mining (603 minerals). However, contrary to previous 1427 estimates, we find that only  $\sim 34\%$  of mineral species form *exclusively* as a 1428 consequence of biological processes, in contrast to the estimate of two-thirds by 1429 Hazen et al. (2008). Nevertheless, the appearance of more than 1900 minerals 1430 exclusively through biology represents a significant, pervasive planetary biomarker 1431 (Chan et al 2019; Johnson et al. 2020).

1432 Rare elements are important in Earth's mineral diversity: As noted above, 41 rare 1433 mineral-forming elements present in Earth's crust at less than 5 ppm are essential constituents in ~2400 mineral species. These scarce elements, including 1434 1435 chalcogenides (As, Sb, Se, Te), transition metals (Cd, Mo, Sn), platinum group 1436 elements, and rare earth elements, represent fewer than 1 in 10,000 crustal atoms, yet 1437 they are essential in 42.4 % of minerals (https://rruff.info/ima, accessed 7 March 1438 2021) – far more species than might be expected based on crustal abundances. We 1439 suggest that interactions between host rocks with trace amounts of these elements 1440 and aqueous fluids, especially with varied anionic species that select and concentrate 1441 metal ions, are the principal factor in creating local chemical anomalies with extreme

rare element concentrations. This observation is consistent with the occurrence of mineral-rich localities associated with complex granite pegmatites, agpaitic rocks, and many hydrothermal ore deposits. In the context of evolving complex systems, including natural ecosystems and human societies, it is significant that the presence of a few exotic individuals has the potential to greatly enhance system diversity.

1447 Most minerals are known to form by only one paragenetic process: Most mineral 1448 species -3349 of 5659, or 59.2 % – are known to form by only one of our proposed 1449 57 paragenetic modes. An additional 1372 species (24.2 %) have two known 1450 paragenetic modes, whereas 458 species (8.1 %) have three. By contrast, only 26 1451 minerals, all of them familiar and abundant rock-forming phases (Deer et al. 1982-1452 2013), are known to form in 12 or more different ways. Pyrite boasts the most 1453 diverse range, with 21 paragenetic modes, including igneous, metamorphic, 1454 sedimentary, and alteration host lithologies; encompassing both abiotic and biotic 1455 processes; and spanning more than 4.5 billion years of Earth history, from pre-1456 terrestrial asteroid alteration (> 4.56 Ga) to precipitation in anthropogenic mine 1457 environments (< 1 Ka). Other widely-forming species with 14 or more paragenetic 1458 modes include albite, corundum, hematite, and magnetite (with 18 modes); calcite, 1459 hornblende, and rutile (17); baryte (15); and forsterite, ilmenite, quartz, and 1460 sphalerite (14). Note that the previous finding that most minerals are rare, found at 1461 five or fewer localities (Hazen et al. 2015; Hystad et al. 2015; Hazen and Ausubel 1462 2016), mirrors the observation that most minerals have only one mode of formation. 1463 Our work also raises intriguing questions that will drive future investigations:

1464 Does Earth's mineralogy display a progressive rise in complexity? Pioneering work 1465 on the quantification of mineral complexity by Sergey Krivovichev (2012, 2013, 1466 2014, 2015, 2016) facilitates testing of hypotheses related to increased mineral 1467 complexity through time. Preliminary work has been applied to suites of minerals 1468 based on composition (Grew et al. 2016, 2017; Morrison et al. 2020) or approximate 1469 stage in Earth history (Krivovichev et al. 2018). The present study offers the 1470 opportunity to probe the evolving complexity of Earth's minerals in greater depth, by 1471 considering the relative influences of temperature, pressure, and/or composition in 1472 various mineral-forming environments as a function of geological time. Quantitative 1473 investigations of whether, and if so how, mineral system complexify over time could 1474 provide a rigorous test of models of cosmic chemical evolution (e.g., Zaikowski and 1475 Friedrich 2007).

1476 What mineral-forming environments occur on the Moon, Mars, and other terrestrial 1477 worlds? Enumerating paragenetic modes, and placing each mineral species into one 1478 or more of those categories, offers an opportunity to evaluate extraterrestrial 1479 mineralogy with a new perspective. If Mars had (or still has) a hydrological cycle, 1480 what mineralogical manifestations might we expect? For example, are there Martian 1481 hydrothermal sulfide deposits and, if so, were a variety of metals mobilized? On the 1482 other hand, if the Moon is truly dry, then what paragenetic processes are excluded? 1483 And do extraterrestrial bodies display paragenetic processes not seen on Earth, such 1484 as cryo-volcanism on Titan (Maynard-Casely et al. 2018; Hazen 2018).

• *What role did minerals play in the origins of life?* A recurrent theme in recent studies 1486 of Earth's evolving crust, in particular the tectonic processes that shaped it, is

1487 speculation that many of Earth's mineral-forming processes may have occurred 1488 much earlier than previously modeled. The formation of oceans prior to 4.45 Ga, the 1489 extensive development of continental crust by 4.4 Ga, and perhaps even the initiation 1490 of some form of subduction-driven plate tectonics in the early Hadean Eon, push 1491 many important mineral-forming processes to Earth's first 250 million years. These 1492 scenarios, if true, have important implications for models of mineral evolution and 1493 the origins of life. Previous estimates of Hadean mineral diversity posited 1494 mineralogical parsimony, with only a few hundred species prior to 4 Ga (Hazen 1495 2013; Morrison et al. 2018). However, this new tabulation of mineral paragenetic 1496 modes, coupled with recent estimates of the rapid evolution of a dynamic early 1497 Hadean Earth, suggest that as many as 3534 mineral species - the collective 1498 inventories of modes [#1] through [#43] – may have formed within Earth's first 250 1499 million years. If so, then most of the geochemical and mineralogical environments 1500 invoked in models of life's origins would have been present by 4.3 Ga. If life is rare 1501 in the universe, requiring an idiosyncratic sequence of unlikely chemical reactions 1502 (Hazen 2017), then this view of a mineralogically diverse early Earth provides many 1503 more plausible reactive pathways over a longer timespan than previous models. If, 1504 on the other hand, life is a cosmic imperative that emerges on any mineral- and 1505 water-rich world, then these findings support the hypothesis that life on Earth 1506 emerged rapidly, in concert with a vibrant, diverse Mineral Kingdom, in the earliest 1507 stages of planetary evolution.

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Paragenetic Mode*	Age (Ga)	#Species**	#Unique***	Ref.****
Pro townstrial "I be minorals"	> 1 57	20	Q	
<u>Pre-terrestrial "Ur-minerals"</u>	<u>&gt; 4.57</u>	<u>30</u> 22	<u>8</u> 1	1-4
<ol> <li>Stellar atmosphere condensates</li> <li>Interstellar condensates****</li> </ol>		8	1 7	1-4 5-7
2. Interstenar condensates		8	/	5-7
Stage 1: Primary nebular phases	<u>4.567—4.561</u>	<u>86</u>	<u>14</u>	
3. Solar nebular condensates (CAIs, AOAs, URIs)	> 4.565	<u>86</u> 48	<u>14</u> 14	7-12
4. Primary chondrule phases	4.566-4.561	47	0	11-15
Stage 2: Planetesimal differentiation & alteration	<u>4.566—4.550</u>	<u>261</u>	<u>65</u>	
5. Primary asteroid phases	4.566-4.560	94	<u>65</u> 21	11,13,16-18
6. Secondary asteroid phases	4.565-4.550	205	44	11,12,16,17,19-21
Stage 3a: Earth's earliest Hadean crust	> 4.50	<u>400</u>	<u>61</u>	22-26
7. Ultramafic igneous rocks		123	10	22,23,27
8. Mafic igneous rocks		93	6	22,23,28-33
9. Lava/xenolith minerals (hornfels, sanidinite facies)		127	8	22,23,34
10. Basalt-hosted zeolite minerals		107	30	35
11. Volcanic fumarole minerals; reduced phases (see also	[#45])	36	7	34,36
Stage 3b: Earth's earliest hydrosphere	> 4.45	<u>350</u>	<u>64</u>	37,38
12. Hadean hydrothermal subsurface sulfide deposits (see	e also [#33])	129	48	39-43
13. Hadean serpentinization		67	8	44-49
14. Hot springs, geysers, and other subaerial geothermal minerals		61	1	35,50
15. Black/white smoker minerals and other seafloor hydrothermal minerals		32	0	46,51,52
16. Low-T aqueous alteration of Hadean subaerial lithologies (see also [#23])		83	3	35,50
17. Marine authigenic Hadean minerals (see also [#24])		51	1	35
18. Minerals formed by freezing		4	3	53

## Table 1. Chronological list of 57 paragenetic modes of minerals, with numbers of associated mineral species

Stage 4a: Earth's earliest continental crust	<u>&gt; 4.4—3.0</u>	<u>2385</u>	<u>1031</u>	22,23,54-57
Igneous rocks		<u>160</u>	<u>17</u>	
19. Granitic intrusive rocks		143	15	22,23,58
20. Acidic volcanic rocks		45	2	22,23,59
Near-surface Processes		<u>1113</u>	<u>244</u>	
21. Chemically precipitated carbonate, phosphate, iron form	nations	79	$\frac{244}{3}$	28,60-67
22. Hydration and low-T subsurface aqueous alteration (see		247	85	37,38,68-70
23. Subaerial aqueous alteration by non-redox-sensitive flu	ids (see also [#47]	) 398	79	71
24. Authigenic minerals in terrestrial sediments (see also [#	[17])	74	0	35,71,72
25. Evaporites (prebiotic)		210	65	57,62,73
26. Hadean detrital minerals		250	7	74
27. Radioactive decay; auto-oxidation		9	0	75-77
28. Photo-alteration, pre-biotic		10	0	78
29. Lightning-generated minerals		7	0	34,79-81
30. Terrestrial impact minerals		16	5	82,83
High-T alteration and/or metamorphism		1505	<u>770</u>	
31. Thermally altered carbonate, phosphate, and iron forma	tions	356	89	28,60-66
32. Ba/Mn/Pb/Zn deposits, including metamorphic deposits	5	412	185	40,68,84
33. Minerals deposited by hydrothermal metal-rich fluids (s	see also [#12])	797	496	37-43
Stage 4b: Highly evolved igneous rocks	<u>&gt; 3.0</u>	<u>1476</u>	<u>662</u>	
34. Complex granite pegmatites	<u> </u>	<u>1770</u> 564	238	85-88
35. Ultra-alkali and agpaitic igneous rocks		726	377	89,90
36. Carbonatites, kimberlites, and related igneous rocks		291	40	40,91-97
37. Layered igneous intrusions and related PGE minerals		135	7	40,98-100
Stage 5: Initiation of plate tectonics	<u>&lt; 3.5—2.5</u>	<u>458</u>	<u>56</u>	101-107
38. Ophiolites		108	7	108-111
39. High-P metamorphism (blueschist, eclogite, ultrahigh P	facies)	70	12	112-118

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40. Regional metamorphism (greenschist, amphibolite, granulite	e facies)	319	36	117,118
41. Mantle metasomatism		16	1	119,120
42. Sea-floor Mn nodules		15	0	121-123
43. Shear-induced minerals (including mylonite/slickensides)		9	0	124,125
<u>Stage 6: Anoxic biosphere</u>	<u>&lt; 4.0</u>			
44. Anoxic microbially-mediated minerals (see also [#44])		11	0	126-141
Stage 7: Great Oxidation Event	<u>&lt; 2.4</u>	<u>2328</u>	<u>1298</u>	142-147
45. Oxidized fumarolic minerals (see also [#11])		424	168	34,36
a. [Sulfates, arsenates, selenates, antimonates]		[253]	[116]	
b. [Other oxidized fumarolic minerals]		[276]	[108]	
46. Near-surface hydrothermal alteration of minerals (see also [#	#22])	52	11	37,38,148
47. Low-T subaerial oxidative hydration, weathering (see also [#	#16] & [#23]	) <u>1998</u>	<u>1119</u>	148
a. [Near-surface hydration of prior minerals]		[1649]	[956]	
b. [Sulfates and sulfites]		[409]	[185]	
c. [Carbonates, phosphates, borates, nitrates]		[577]	[338]	
d. [Arsenates, antimonates, selenates, bismuthinates]		[505]	[361]	
e. [Vanadates, chromates, manganates]		[372]	[229]	
f. [Uranyl $(U^{6+})$ minerals]		[236]	[173]	149,150
g. [Halogen-bearing surface weathering minerals]		[236]	[121]	
h. [Near-surface oxidized, dehydrated minerals]		[316]	[176]	
i. [Terrestrial weathering of meteorites]		[28]	[10]	11
Stage 8: "Intermediate Ocean" (no new paragenetic modes)	1.9—0.9	0	0	151-155
Stage 9: "Snowball Earth" (no new paragenetic modes)	0.9—0.6	0	0	156-158
Stage 10a: Neoproterozoic oxygenation/terrestrial biosphere	< 0.6	<u>597</u>	<u>72</u>	
48. Soil leaching zone minerals	< 0.6	71	4	159-166
49. Oxic cellular biomineralization (see also [#44])	< 0.54	77	1	136,167-186

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50. Coal and/or oil shale minerals	< 0.36	273	4	187,188
51. Pyrometamorphic minerals (see also [#54] & [#56])	< 0.36	128	32	34,189,190
52. Guano- and urine-derived minerals	< 0.4	72	17	191,192
53. Other minerals with taphonomic origins	< 0.4	117	14	193,194
<u>Stage 10b: Anthropogenic minerals</u>	< 10 Ka	<u>603</u>	<u>11</u>	195-199
54. Coal and other mine fire minerals (see also [#51] & [#56])		234	6	34,200-202
55. Anthropogenic mine minerals		264	5	197
56. Slag and smelter minerals (see also [#51] & [#55])		143	0	34,197,203-204
57. Other minerals formed by human processes		49	0	195-199, 205,206

\* The 10 stages of mineral evolution refer to temporal divisions, as outlined in Table 1 of Hazen et al. (2008). The 57 paragenetic modes are designated by [#1] to [#57].

\*\* Numbers of species are based on paragenetic modes identified for 5659 IMA-approved mineral species, as tabulated on https://rruff.info/ima (accessed 23 November 2020).

\*\*\* Numbers of species known to form only by that paragenetic mode.

\*\*\*\* **References:** General references for mineral paragenetic modes include Anthony et al. (1990-2003); Deer et al. (1982-2013); and <a href="https://rruff.info/ima">https://rruff.info/ima</a> and <a href="https://rruff.info/ima">https://rruff.info/ima</

Numbered references: 1 = Hazen & Morrison (2020); 2 = Zinner (2014); 3 = Nittler & Ciesla (2016); 4 = Boujibar et al. (2021); 5 = Whittet (2003); 6 = Gibb et al. (2004); 7 = Morrison & Hazen (2020); 8 = MacPherson (2014); 9 = Krot et al. (2004); 10 = El Goresy et al. (2002); 11 = Rubin & Ma (2021); 12 = Brearley & Jones (1998); 13 = Krot et al. (2014); 14 = Russell et al. (2018); 15 = Hazen et al. (2021); 16 = Mittlefehldt et al. 1998; 17 = Mittlefehldt (2014); 18 = Morrison & Hazen (2021); 19 = McSween et al. (1988); 20 = Hazen & Morrison (2021); 21 = Tomioka & Miyahara (2017); 22 = Rollinson (2007b); 23 = Van Kranendonk et al. (2007b); 24 = Papineau (2010); 25 = Hazen (2013); 26 = Morrison et al. (2018); 27 = Korenaga (2021); 28 = Isley & Abbott (1999); 29 = O'Neil et al. (2008); 30 = Ashwal (1993); 31 = Prokoph et al. (2004); 32 = O'Neil et al. (2008); 33 = Ernst (2014); 34 = Grapes (2006); 35 = Deer et al. (2004); 36 = Vergasova & Filatov (2016); 37 = Heinrich and Henley (1989); 38 = Pirajno (2009); 39 = Laznicka 1973; 40 = Meyer (1988); 41 = Groves et al. (2005); 42 = Huston et al. (2010); 43 = Jenkin et al. (2015); 44 = Blais & Auvray (1990); 45 = Lowell & Rona (2002); 46 = Palandri & Reed (2004); 47 = Shrenk et al. (2013); 48 = Holm et al. (2015);

49 = Menez et al. (2018); 50 = Wilson (2013); 51 = Hekinian et al. (1980); 52 = Haymon and Kastner (1981); 53 = Genceli Güner et al. (2013); 54 = Wilde et al. (2001); 55 = Kemp & Hawkesworth (2003); 56 = Kemp et al. (2007); 57 = Johnson et al. (2017); 58 = Chappell & White (2001); 59 = Sugitani et al. (2003); 60 = Falkowski et al. 2000; 61 = Klein (2005); 62 = Button (1982); 63 = Anbar & Holland (1992); 64 = Bekker et al. (2010); 65 = Barturin & Bezrukov (1979); 66 = Delaney (1998); 67 = Crevello et al. (1989); 68 = Post (1999); 69 = Savage et al. (1987); 70 = Nishimoto & Yoshido (2010); 71 = Chang et al. (1996); 72 = Holland & Rye (1997); 73 = Buick & Dunlop (1990); 74 = Baker (1962); 75 = Frondel (1958); 76 = Finch & Murakami (1999); 77 = Hazen et al. (2009); 78 = Kim et al. (2013); 79 = Essene & Fisher (1986); 80 = Pasek et al. (2012); 81 = Hess et al. (2021); 82 = Tomioka & Miyahara (2017); 83 = Tschauner (2019); 84 = Leach et al. (2010); 85 = London (2008); 86 = Tkachev (2011); 87 = Grew et al(2017); 88 = Grew et al. (2019); 89 = Marks & Markl (2017); 90 = Mikhailova et al. (2019); 91 = Patchett et al. (1982); 92 = Bell (1989); 93 = Downes et al. (2012); 94 = Jones et al. (2013); 95 = Mitchell et al. (2019); 96 = Woolley & Kjarsgaard (2008); 97 = Heaman et al. (2019); 98 = Naldrett et al. (1987); 99 = Maier & Groves (2011); 100 = Zientek (2012); 101 = Stern (2005); 102 = Rollinson (2007a); 103 = Condie & Pease (2008); 104 = Shirey et al. (2008); 105 = Shirey and Richardson (2011); 106 = Bauer et al. (2020); 107 = Reimink et al. (2019); 108 = Bai et al. (2011); 109 = Moores (2002); 110 = Dilek (2003); 111 = Furnes et al. (2007); 112 = Chopin (1984); 113 = Hacker (2006); 114 = Palin and White (2016); 115 = Zheng and Chen (2017); 116 = Brown (2007); 117 = Philpotts & Ague (2009); 118 = Vernon (2008); 119 = Luth (2003); 120 = O'Reilly and Griffin (2012); 121 = Hlawatsch et al. (2002); 122 = Fike et al. (2006); 123 = Wegorzewski & Kuhn (2014); 124 = Passchier & Trouw (2005); 125 = Trouw et al. (2009); 126 = Van Kranendonk et al. (2003); 127 = Akai et al. (1999); 128 = Allwood et al. (2006); 129 = Anbar & Holland (1992); 130 = Banerjee (1971); 131 = Frankel & Bazylinski (2003); 132 = Grotzinger & Knoll (1999); 133 = Konhauser et al. (2005); 134 = Kappler et al. (2005); 135 = Schulz & Schulz (2005); 136 = Southam & Saunders (2005); 137 = Lepot et al. (2008); 138 = Marshall (1979); 139 = Burnett & Riggs (1990); 140 = Chauhan (1979); 141 = Cook & Shergold (1990); 142 = Sverjensky & Lee (2010); 143 = Hazen (2015); 144 = Jabłońska & Tawfik (2021); 145 = Canfield (2014); 146 = Lyons et al. (2014); 147 = Anbar et al. (2007); 148 = Anthony et al. (1990-2003); 149 = Nash et al. (1981); 150 = Hazen et al. (2009); 151 =Brasier (1998); 152 = Buick et al. (1995); 153 = Anbar & Knoll (2002); 154 = Brasier (2012); 155 = Mukherjee et al. (2018); 156 = Kirschvink (1992); 157 = Hoffman et al. (1998); 158 = Bodiselitsch et al. (2005); 159 = Johnson and Watson-Stegner (1987); 160 = Holland and Rye (1997); 161 = Rye and Holland (1998); 162 = Retallack (2001); 163 = Kennedy et al. (2006); 164 = Ueshima & Tazaki (1998); 165 = Helgren and Butzer (1977); 166 = Hill et al. (2000); 167 = Yoder (2002); 168 = Dove et al. (2003); 169 = Dove (2010); 170 = Lowenstam & Weiner 1989; 171 = Ensikat et al. (2016); 172 = Endo et al. (2018); 173 = Lichtenegger et al.

5

(2002); 174 = Zuzuki & Banfield (1999); 175 = Wufuer et al. (2017); 176 = Beazley et al. (2017); 177 = Berg et al. (2020); 178 = Pósfai et al. (2013); 179 = Uebe and Schüler (2016); 180 = Kelm et al. (2015); 181 = Reith et al. (2009); 182 = Li & Gadd (2017); 183 = Cron et al. (2019); 184 = Carmichael & Bräuer (2015); 185 = Weiner & Wagner (1998); 186 = Kattimani et al. (2016); 187 = Saxby (2000); 188 = Schweinfurth (2016); 189 = Gross (1977); 190 = Sokol et al. (2008); 191 = Audra et al. (2017); 192 = Audra et al. (2018); 193 = Behrensmeyer et al. (2000); 194 = Mustoe (2018); 195 = Catheart (2011); 196 = Corcoran et al. (2014); 197 = Hazen et al. (2017); 198 = Hazen & Zalaziewski (2019); 199 = Zeng et al. (2020); 200 = Lapham et al. (1980); 201 = Kruszewski (2013); 202 = Stracher et al. (2015); 203 = Sueoka & Sakakibara (2013); 204 = Kierczak & Ettler (2021); 205 = Merlino & Sartori (1970); 206 = Smith & Prior (1899)

\*\*\*\*\* Includes 7 interstellar ice phases that are not IMA-approved mineral species.